STUDY OF A NOVEL TERNARY SECOND-ORDER VISCOSITY MODEL ON Al_2O_3-WATER NANOFLUID

by

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In terms of heat dissipation, nanofluids with strong thermal conductivity are becoming more and more common, attracting more and more research and attention date. In this study, Al_2O_3-water nanofluids were studied using molecular dynamics simulations and model parameterization. The dynamic viscosity distribution patterns were obtained at various temperatures (290~360 K), nanoparticle volume fractions (1.24~6.2%), and particle sphericity (0.69~1.0), and an efficient ternary second-order polynomial viscosity prediction model was proposed on the basis of these results. The findings demonstrate the model's goodness-of-fit with a coefficient of determination over 0.96 and a root mean square error under 0.05, as well as its high predictive ability with a maximum relative error between simulated and predicted values under 9%. Using this viscosity prediction model, a subsequent parametric sensitivity study shows that the volume fraction had the most significant impact on viscosity, exhibiting not just a second order effect but also an interacting effect with temperature and sphericity. The relative nanofluid viscosity, which is the ratio of nanofluid viscosity to aqueous base fluid viscosity, exhibits a convex parabolic growth at constant temperature and sphericity and increases more quickly at the same volume fraction the higher the temperature. The viscosity of the nanofluid increases by up to 34% when the volume fraction is equal to 6.28% and the particle sphericity is equal to 1. An efficient viscosity prediction model makes it easier to control important variables to reduce energy consumption during flow and increase its capacity to dissipate heat.

Key words: *dynamic viscosity, alumina-water nanofluid, prediction model, goodness-of-fit*

Introduction

The amount of thermal power used per unit volume of electronic devices likewise rises quickly as a result of their increasing high integration, miniaturization, and sophisticated functionalities. As of right now, the chip's greatest operating temperature can even reach 200 °C, and its heat flow is growing by 7% annually [1, 2]. The reliability and performance of electronic components will be impacted by high temperatures. This calls for the development

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of effective cooling technology. If not, heat failure will occur throughout the entire electrical product [3-5].

Due to their high thermal conductivity and difficulty in blocking, nanofluids – a novel type of heat transfer medium – have gained increasing attention in the field of heat transfer over the past few years [6-9]. They are created by suspending a specific concentration of solid nanoparticles with a size of less than 100 nm in a base fluid. One of the most crucial thermal qualities, viscosity, controls the pumping effort and pressure drop during flow as well as the nanofluid's flow state. Viscosity is an important thermal property which influences the the fluid's resistance and leads to an increase in pump work as the viscosity increases. Therefore, a precise prediction of viscosity is essential for the use of heat transfer fluid medium in these and other fields [10] to build the thermal system and calculate the necessary pumping power [11].

Currently, numerical simulations and experimental research are the two main methods used to study viscosity. Garoosi [12] put forward two new empirical formulas to calculate the effective dynamic viscosity of nanofluids and presented the relationship between the effective dynamic viscosity and particle volume fraction ($0\% \leq \psi \leq 12\%$), particle size ($10\ \text{nm} \leq d_p \leq 5\ \mu\text{m}$), particle type (Ag, Cu, Al_2O_3, TiO_2, CuO, SiO_2, *etc*.), base liquid temperature and thermophysical properties, and the data for establishing the equations depended on a large number of experimental data available in the literature. Furtherly, Which were found that the two models were more accurate than the frequently used classical models (*e.g*. Brinkman and Maxwell-Garnett models) in literature. Moldoveanu *et al*. [13] got the conclusion that Al_2O_3-water based nanofluids possessed a non-Newtonian behavior and the viscosity of which was higher compared with SiO_2 nanofluid. An equation that varied with temperature was proposed, with a relationship that the viscosity decreased with the increase of temperature. Moldoveanu *et al.* [14] obtained the relationship between relative viscosity and the volume fraction of nanofluids using regression analysis and curve fitting methods in the experimental study on the viscosity of stabilized Al_2O_3, TiO_2 nanofluids and their hybrid. Minakov *et al*. [15] systematically studied the viscosity of more than 30 kinds of nanofluids such as Al_2O_3 and TiO_2. Aamong them, the base fluids were distilled water, ethylene glycol and engine oil, respectively, the volume concentration of nanoparticles was 0.25-8%, the size of nanoparticles ranged from 5-150 nm, and the temperature changed from 25-60 ℃. The results showed that the viscosity of nanofluids increased with the decrease of nanoparticle size, and the higher the viscosity of the base fluid, the higher the increment of the viscosity of nanofluids. Alawi *et al.* [16] studied the thermal conductivity and viscosity of various metal oxides with the 1-5% volume concentration of nanoparticles at the temperature of 300-320 K. The results indicated that the viscosity increased significantly with the increase of nanoparticle concentration. Elcioglu *et al*. [17] carried out an experimental study on Al_2O_3 nanofluids by Taguchi experimental design plan. Data analysis made clear that the interaction between temperature and nanoparticle volume fraction on viscosity was obvious, and which was reflected in the viscosity equation. Vakilinejad *et al.* [18] measured the kinematic viscosities of three nanofluids including Al_2O_3, TiO_2 and graphene water based nanofluids, and calculated the viscosity ratio to deionized water of all liquids. Polynomial models were proposed for each nanofluid, the percentage error can be as low as 0.06% for Al_2O_3 nanofluid among them. Esfe *et al.* [19] used the experimental data of MWCNT (10%)-Al_2O_3 (90%) hybrid nanofluid to establish an artificial neural network model for predicting viscosity. It was found that temperature had the greatest influence on which and the R^2 values for the ANN was 0.9998.

As mentioned previously, some research has been done on predictive models for viscosity up until this point. However, the form and even the parameters of these viscosity model

expressions vary. Some studies only look at the effect of volume fraction on viscosity and ignore the temperature dependence [20-25], others look at volume concentration and temperature but ignore the effect of particle size [26-30]. Still others don't look at the effect of particle shape on viscosity [31-34], and so on. Further investigation is required to precisely anticipate the viscosity of Al_2O_3 nanofluids, which will facilitate their application.

Therefore, the study aimed to present a three-variable second-order polynomial model with good predictive accuracy. Which consists of three parts mainly, including Molecular dynamics simulation on the viscosity of Al_2O_3-water nanofluid, Construction of a viscosity prediction model and results discussion. The following will be developed and studied in detail in the text.

Viscosity study on Al_2O_3-water nanofluid by molecular dynamics simulations

The Al_2O_3-water nanofluid molecular dynamics simulation

With the ongoing advancement of the theory, methodology, and computer technology of molecular simulation, it has become the third way to comprehend the universe from the molecular and atomic levels, treating it as a virtual laboratory [35]. In this study, the numerical analysis of Al_2O_3-water nanofluid viscosity is carried out using the molecular dynamics (MD) approach with the classic MD software LAMMPS. Here a box of size 3 nm × 3 nm × 6 nm is constructed and the particle size of nanoparticles is 10.86 Å. The simulations are performed using periodic boundaries with a truncation radius of 9 Å. The potential function used is the water molecule SPC/E model parameter and the CLAYFF force field is used for the Al_2O_3 nanoparticles. The model was emulated using the NPT and NVT system synthesis.

Figure 1 shows the Al_2O_3 particles micro-structure by SEM, and the structural properties and parameters are listed in tab. 1.

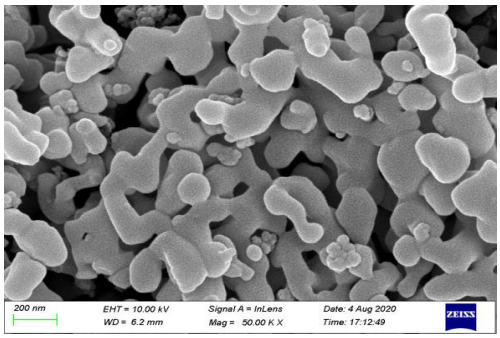


Figure 1. The micro-structure of Al_2O_3 particles by SEM

Table 1. The structural properties and parameters used in MD simulation

Unite	Value	Unite	Value
O mass	15.999 [gmol^{-1}]	LJ sigma of O_1O_3	3.013 [Å]
H mass	1.008 [gmol^{-1}]	LJ epsilon of O_1Al, O_2A_l	0.096 [kcalmol^{-1}]
Al mass	26.982 [gmol^{-1}]	LJ sigma of O_1A_l, O_2A_l	3.447 [Å]
O_3 charge	–0.840 (e)	LJ epsilon of A_lA_l	0.040 [kcalmol^{-1}]
H charge	0.420 (e)	LJ sigma of A_lA_l	4.053
LJ epsilon of O_1O_1, O_1O_2, O_2O_2	0.228 [kcalmol^{-1}]	LJ epsilon of O_3Al	0.078 [kcalmol^{-1}]
LJ sigma of O_1O_1, O_1O_2, O_2O_2	2.859 [Å]	LJ sigma of O_3Al	3.609 [Å]
LJ epsilon of O_3O_3	0.155 [kcalmol^{-1}]	r_0 of OH bond	1.000 [Å]
LJ sigma of O_3O_3	3.166 [Å]	r_0 of O_1Al, O_2Al bond	1.775 [Å]
LJ epsilon of O_1O_3	0.188 [kcalmol^{-1}]	theta of HOH angle	109.470 [°]

The equilibrium molecular dynamics (EMD) method is employed to calculate the viscosity with the Green-Kubo equations 1:

$$\mu_s = \frac{V}{k_B T}\int_0^\infty \left\langle P_{\alpha\beta}(t)P_{\alpha\beta}(0)\right\rangle \mathrm{d}t \tag{1}$$

where μ_s is the shear viscosity, V – the molecular model box volume, k_B – the Boltzmann constant, T – the temperature, and $P_{\alpha\beta}$ – the off-diagonal components of the pressure tensor.

Method verification

The viscosity of water is computed using the same formula and the MD analysis technique to confirm the reliability of the EMD approach. The MD simulation data at temperatures between 300 K and 360 K at atmospheric pressure were used to determine the viscosity of water. Figure 2 shows a comparison of the water viscosity simulation data with the experimental data and the National Institute of Standards and Technology of US (NIST) data, where the viscosity experimental data are obtained by rotational viscometer DHJ-9S at the same temperature. It is discovered that the water viscosity simulation value closely matches the experimental value and the NIST value, with a maximum relative error between the two values of no more than 5%. It also demonstrates how the viscosity of nanofluids can be determined using the EMD approach and the Green-Kubo equation.

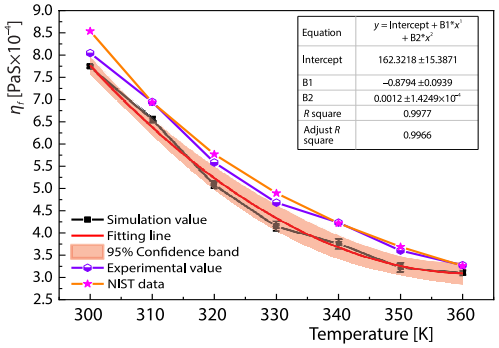


Figure 2. Viscosity comparison between the experimental result, the NIST value, and the simulation value

Viscosity analysis of Al_2O_3-water nanofluid

The macroscopic characteristics of base fluid and nanoparticles will have an impact on the thermophysical characteristics of nanofluids. The interactions between the particles, the base liquid, as well as agglomeration, sedimentation, and varied distributions all have an impact on the properties of the material. Here, the effects of temperature, T, from 290-360 K, volume fraction, φ, from 1.24% to 6.28%, and various forms, given in tab. 2, were explored to determine how viscosity of nanofluids changed. The L stands for length, D for diameter, H for height, and W for width.

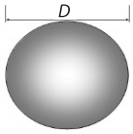
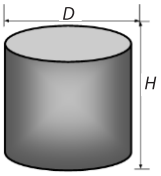
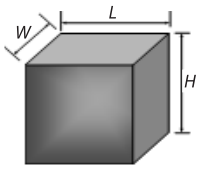
Figure 3 depicts the viscosity data distribution according to temperature, and figs. 4 and 5 illustrate the distribution state with sphericity, α, and volume fraction, respectively. The diverse particle forms are classified by sphericity, and the closer a particle is to a ball in shape, the closer its sphericity is to 1. The computation of sphericity is illustrated in eq. (2).

$$\alpha = \frac{4\pi\left(\dfrac{3V_F}{4\pi}\right)^{2/3}}{S_p} \tag{2}$$

where V_F is the particle volume, S_p – the particle surface area, and α – the particle sphericity.

According to fig. 3, viscosity decreases as temperature rises. The decrease is more pronounced at low temperatures between 290 K and 320 K and tends to be mild at high tem-

Table 2. Structural parameters for different forms of particles

Particle shape	Graphical representation	*D*/*L* [Å]	*H* [Å]	*W* [Å]
Sphere		10.858	–	–
Columnar		10.330	8	–
		11.930	6	–
		14.610	4	–
Cubic		8.743	8.743	8.743
		13.880	6.938	6.938
		20.000	5.780	5.780

peratures between 320 K and 360 K. Even at temperatures as high as 350 K, the trend of viscosity declines slowly, and the rate of change can even approach zero.

Figure 4 shows that the viscosity increases as the volume fraction increases, with the effect being more pronounced when the volume fraction exceeded 3.72%. As the volume fraction and particle count increase, the molecular forces between the particles and the base liquid rise, which causes the friction force of the overall fluid-flow to rise. In fig. 5, the

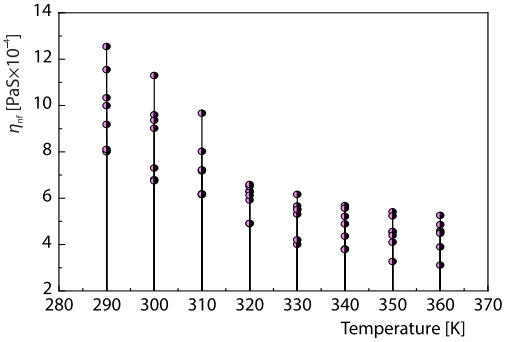


Figure 3. The distribution of viscosity data under the temperature

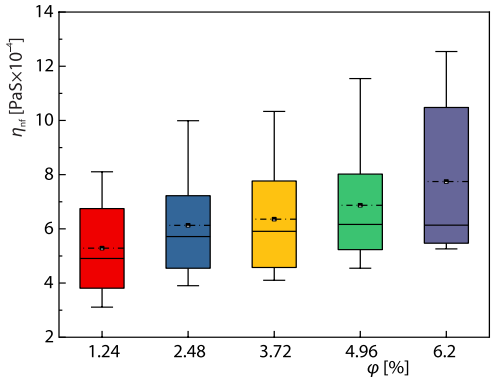


Figure 4. The distribution of viscosity data under φ

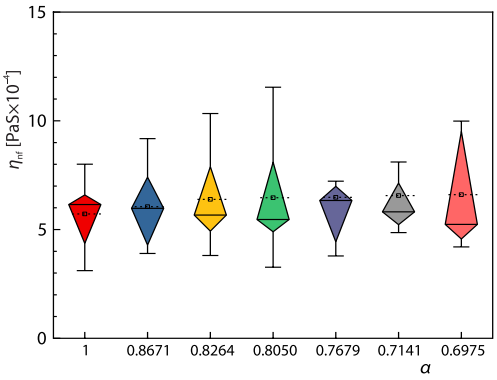


Figure 5. The distribution of viscosity data under α

viscosity likewise rises as the particular surface area changes, but the pattern of change is not very predictable. The contact area and hence viscosity increase as the surface area of the nanoparticles increases.

In the process of analyzing viscosity data, it is discovered that numerous parameters interact with one another. Figures 6 and 7 illustrate the relationship on η_{nf} between temperature, volume fraction, and as well as the relationship between particle sphericity and temperature.

According to fig. 6, at the same temperature, the viscosity increases with a higher volume fraction, and at a higher temperature, the effect of volume fraction on viscosity decreases. fig. 7 shows that the effect of particle sphericity on viscosity is nearly constant across a range of temperatures, however there is a bigger fluctuation when the sphericity is less than 0.85.

It is important to process the viscosity data and develop the relational expression between viscosity and factors like temperature, volume fraction, and particle sphericity in order to represent the relationship between them.

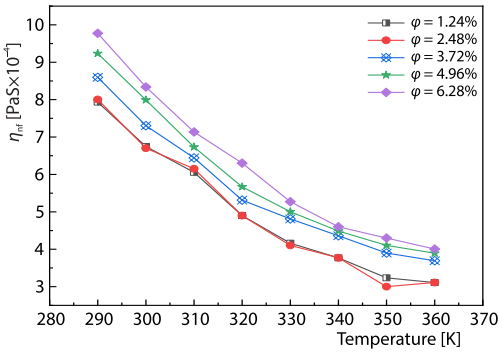


Figure 6. Effect of volume fraction and temperature on η_{nf} at $\alpha = 1$

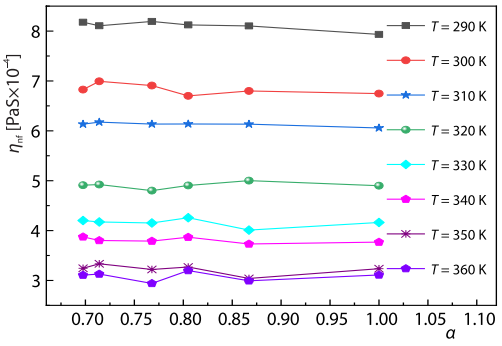


Figure 7. Effect of particle sphericity and temperature on η_{nf} at $\varphi = 1$

Viscosity model study on Al_2O_3-water nanofluid

The viscosity of nanofluids can be precisely predicted using a competent parametric model. A parameterized polynomial model of viscosity is preferred as eq. (3) in light of the aforementioned data distribution. To find the relevant expressions and calculated coefficients, this equation needs to be further examined. The a_0, a_1, ... a_{10} is the coefficient terms. The specific process is to determine an optimal model expression and the coefficients in the expression using a stepwise forward regression method for the constructed ternary second-order full-effects model, examining the changes in the four indicator terms R^2, RMSE, AIC_c, and BIC, as well as the contribution of the introduction of each variable to the least-squared sum of effects, weighing the complexity of the model against the accuracy of the prediction.

$$\frac{\eta_{nf}}{\eta_f} = a_0\varphi^2 + a_1T^2 + a_2\alpha^2 + a_3\varphi T + a_4\varphi\alpha + a_5\alpha T + a_6\varphi + a_7T + a_8\alpha + a_{10} \tag{3}$$

Equation (4) calculates the effect size based on the least squares, the independent effects of the three variables, the cross effects between them, and the quadratic effects of which are picked as the legitimate parameters (effect size > 2). In the formula, P-value is the significance level, and the values above two are significant at $P = 0.01$. As seen from fig. 8, the α, T^2, and αT in the formula can be ignored:

$$\text{Effect size} = -\log10(P) \tag{4}$$

The viscosity prediction model is then given as eq. (5) using the stepwise regression method, fully taking into account the higher R^2, lower RMSE, lower AICc, and lower BIC, as seen in tab. 3. This equation consists of six variable terms with a goodness of fit up to 0.96. It is clear that φ significantly affects the changes in viscosity.

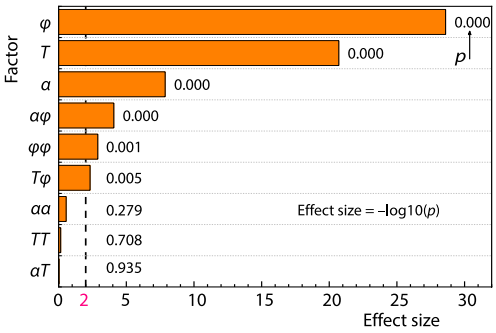


Figure 8. Parameter effects screening by least squares method

Table 3. Convergence table of the parameters of the stepwise regression method

Variables	R^2_{max}	$RMSE_{min}$	$AIC_{c,min}$	BIC_{min}
1	0.024	0.209	–12.567	–6.891
2	0.851	0.082	–117.390	–109.990
3	0.925	0.059	–153.840	–144.810
4	0.943	0.052	–167.620	–157.040
5	0.953	0.048	–175.140	–163.130
6	0.961	0.044	–183.720	–170.380
7	0.962	0.044	–182.330	–167.770

$$\frac{\eta_{nf}}{\eta_f}\cdot 10^3 = 125.85\varphi + 2.67T + 29.09\alpha - 8.20\varphi^2 + 0.51\varphi T - 171.89\varphi\alpha - 192.10 \tag{5}$$

where η_{nf} is the nanofluid viscosity, η_f – the base fluid viscosity, φ – the volume fraction, T – the temperature, and α – the particle sphericity.

Figure 9 shows that the simulation value and the model's predicted value are very well matched, with a relative error between them of less than 9%. It indicates that molecular dynamics methods can be used well to model the dynamic viscosity of nanofluids, but the process is complex, whereas the predictive model is fast and accurate under the same conditions.

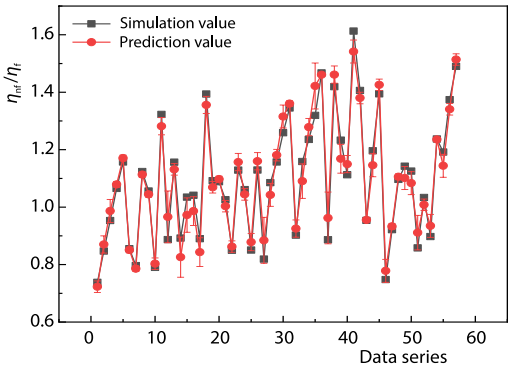


Figure 9. Comparison of predicted and simulated values

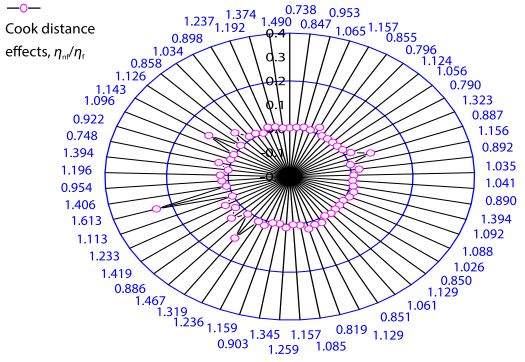


Figure 10. Model parameters Cook's distance effects

Figure 10 shows that the Cook's distance is less than 0.3 and that there are no out-of-the-ordinary data points that significantly alter the regression equation, which reflects the fact that the absence of outliers in the sample data can have a misleading effect on the accuracy of the model predictions. The viscosity prediction equation is hence reliable and precise. A good predictive model can give better and faster values for the viscosity of Al_2O_3-water nanofluids, simply by entering the values of the parameters under different conditions. It is easier to find and analyse the pattern of the effect of different parameters on the viscosity, as in section *Sensitivity analysis of viscosity prediction model*.

Results and discussion

Comparison of viscosity models

Some models have been used to estimate the viscosity of nanofluids, with the concentration of the nanofluid serving as the sole input parameter. Here, three traditional nanofluid viscosity models – discussed in tab. 4 and illustrated in fig. 11 – are utilized to compare with the model given in this research.

Table 4. A comparison table between the four models

	Equation	Subject	Range of volume fraction
[36]	$\frac{\mu_{nf}}{\mu_{bf}}=1+2.5\varphi$	Spherical shape	$\varphi < 0.2\%$
[37]	$\frac{\mu_{nf}}{\mu_{bf}}=\frac{1}{(1-\varphi)^{2.5}}$	Spherical shape	$\varphi < 0.2\%$
[38]	$\frac{\mu_{nf}}{\mu_{bf}}=1+2.5\varphi+6.5\varphi^2$	Spherical shape	$\varphi < 0.2\%$
Self-defined viscosity model	$\frac{\eta_{nf}}{\eta_f}\cdot 10^3=125.85\varphi+2.67T+29.09\alpha-8.20\varphi^2+0.51\varphi T-171.89\varphi\alpha$	Arbitrary shape	$\varphi < 6.28\%$

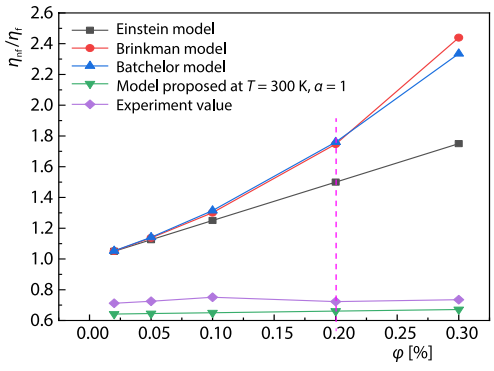


Figure 11. Comparison of the predicted values of the four models

According to the aforementioned table and graph, Einstein, Brinkman, and Batchelor models have only one independent variable of volume fraction, which is less than 0.2 percent with a good prediction. As a result, none of them can make an accurate prediction unless other effective parameters such as temperature and particle shape are included. The viscosity model proposed in this paper has a high level of prediction accuracy and agreement with experimental values, with a maximum relative error of less than 10%.

Sensitivity analysis of viscosity prediction model

– Effect of the volume concentration on the viscosity

Furthermore, a sensitivity analysis of the forecasting model is performed. The results show that the viscosity ratio has a non-linear increasing tendency, and the volume fraction of 6.5% reaches its highest value of 0.9886 K at 300 K before starting to decline. Additionally, the tipping point lags as the temperature rises. For a given volume fraction, it can be shown that the ratio increases more as the temperature rises; this trend is more obvious for volume fractions above 2%. For example, at 360 K, the viscosity ratio increases by more than 37% with the volume fraction increasing from 0.02-8%, and the increase in viscosity ratio is nearly 20% regardless of temperature change while the volume fraction is less than 2%. The nanofluid becomes more viscous as the number of particles grows, increasing both the gravitational attraction between nearby particles and the frictional resistance that must be overcome when flowing.

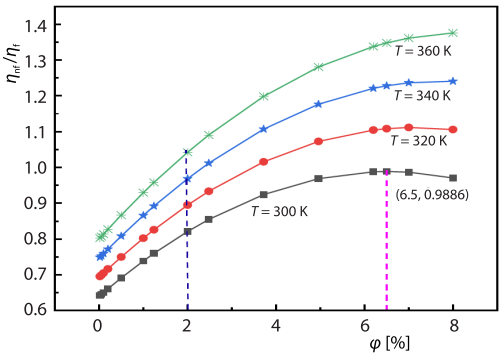


Figure 12. Variation of viscosity with volume fraction under spherical particles

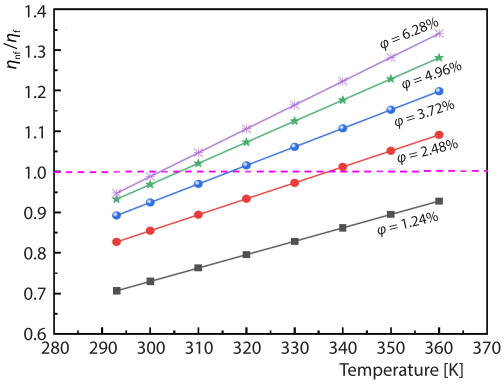


Figure 13. Variation of viscosity ratio with temperature under spherical particles

– Effect of the temperature on the viscosity

Figures 12 and 13 both demonstrate that temperature has a significant impact on the nanofluid's viscosity. Figure 13 shows that for spherical Al_2O_3-water nanofluid, an increase in temperature causes a corresponding rise in the viscosity ratio. The ratio grows to its highest value at 360 K with a volume fraction of 6.28%. Additionally, the viscosity ratio increases more quickly at the same temperature the larger the volume percentage. For instance, at 320 K, the growth rate at the volume fraction of 6.28% is higher than that at the volume fraction of 1.24% by more than 30%. In addition, the viscosity ratio of change slows down at the same temperature and is more pronounced at low temperatures.

The Brownian motion of the nanoparticles is improved by higher temperature, which also lessens the attraction between the particles and reduces the viscosity of the nanofluid. However, the relative viscosity of the nanofluid will increase with increasing temperature if the rate of reduction is larger than the rate at which the viscosity of the base fluid decreases with increasing temperature. Therefore, the concentration of nanoparticles must be increased or the temperature of the nanofluid must be raised in order to effectively increase the viscosity of the base fluid (viscosity ratio over 1).

– Effect of the particle sphericity on the viscosity

Irregularities in the shape of the nanoparticles can exacerbate their interaction in the base fluid. As seen in fig. 14, the viscosity ratio decreases as the nanoparticles get closer to having a spherical form, showing that spherical particles have less mutual adsorption and shear forces. The lowest viscosity ratio is found in spherical particle nanofluids, which do not sur-

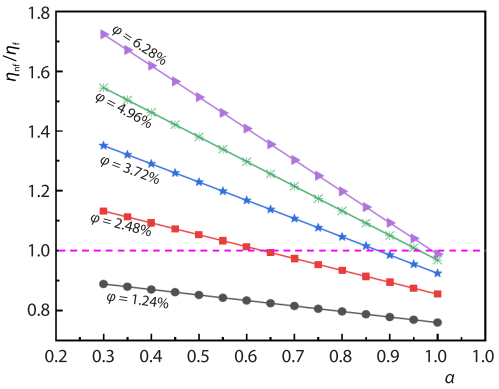


Figure 14. Variation of viscosity ratio with particle sphericity at 300 K

pass 1 even at volume fractions of 6.28%. In spherical particle nanofluids, the rate of change of the viscosity ratio decreases with increasing volume fraction, and at the same sphericity, the higher the volume fraction, the higher the viscosity ratio. For example, at a volume fraction of 6.28%, the sphericity of 0.3 has a growth rate of 72%. Additionally, the viscosity ratio begins to decrease at a slower rate with a volume fraction of more than 2.48%, and this trend continues to decline as the volume fraction rises.

Conclusions

This study uses molecular dynamics to examine how the viscosity of an Al_2O_3-water nanofluid changes as a function of parameters, including temperature (290-360 K), volume fraction (1.24-6.28%), and particle sphericity (0.3-1, a convergent variable that integrates particle shape and size). To properly supply its viscosity features, a viscosity prediction model is nicely built. The major conclusions, are as follows.

- With a prediction accuracy of over 90%, a ternary second-order viscosity prediction model of an Al_2O_3-water nanofluid is proposed, incorporating factors for volume fraction, temperature, and particle sphericity.
- The viscosity of Al_2O_3-water the nanofluid can be effectively increased by adding nanoparticles to the base fluid. The volume fraction, which has non-negligible first- and second-order effects, is the key element in the formula for estimating viscosity. The relative viscosity of the nanofluid increases with increasing volume fraction, although the increasing trend is rapid below 2% concentration and slows down and even decreases above 6.5%. Low temperatures make this effect particularly noticeable.
- The viscosity of Al_2O_3-water nanofluids is significantly influenced by temperature, and temperature and volume fraction interact more strongly. Viscosity of the base solution is more likely to grow when particle concentration in the base solution increases at high temperatures.
- The regularity of the particles added to the base liquid has an inverse relationship with the increase in the viscosity of the nanofluid. When low concentration nanoparticles are added to the base fluid, the more irregular the particles, the more strong the interaction effect, and the greater the viscosity enhancement to the nanofluids are.

Nomenclature

D – diameter
H – particle height, [Å]
L – particle length, [Å]
R^2 – determination coefficient
S_p – particle surface area, [m^2]
T – temperature, [K]
V_F – particle volume, [m^3]
W – particle width, [Å]

Greek symbols

α – particle sphericity
η – fluid viscosity, [Pa·s]
η_f – base fluid viscosity, [Pa·s]
η_{nf} – nanofluid viscosity, [Pa·s]
η_{nf}/η_n – relative viscosity
μ_s – shear viscosity, [Pa·s]
μ_{bf} – base fluid viscosity, [Pa·s]
φ – volume percent, [vol.%]

Acronyms

AIC_c – Akaike information criterion
BIC – bayesian information criterion
RMSE – Root mean square error

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