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Optimising The Mixing Ratio of Hybrid Nanofluids Based on Their Thermal Conductivity and Dynamic Viscosity Properties Using Design of Experiment Method

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ABSTRACT

The hybrid nanofluid's synergetic effect is proportional to the mixing ratio. However, the One Factor at A Time (OFAT) method only displays the optimised mixing ratio when the mixing ratio used in the respective experiments is specified. Thus, the purpose of this study is to optimise the nanoparticle mixing ratio using Design of Experiment (DOE) in Design Expert 11 to cover the entire range of mixing ratios with the fewest experiments possible. The prepared hybrid nanofluid comprises Titanium Dioxide (TiO_2) and Graphene Nanoplatelets (GNP) at a concentration of 0.3vol%, a mixing ratio ranging from 1:9 to 9:1, and temperature ranging from 30°C to 60°C. However, the DOE method generates only 1:9, 1:1, and 9:1 mixing ratios. ANOVA analysis was used to generate a model for thermal conductivity. Additionally, the optimisation results indicate that a mixing ratio of 1:4 (TiO_2-GNP) and a temperature of 40°C are the optimised parameters. The difference between the measured and predicted thermal conductivity values was less than 5%.

1. Introduction

Despite increasing nanofluids' thermal properties, many real-world applications require trade-offs between various nanofluid characteristics/properties. For example, metal oxide nanoparticles exhibit good chemical inertness and stability, while metallic nanoparticles like Aluminum, Copper, Silver possess higher thermal conductivities but are chemically reactive and unstable [1]. Thus, a new type of nanofluid called hybrid nanofluid is created by hybridising two different nanoparticles with unique properties. A hybrid nanofluid is a mixture of two distinct nanoparticles dispersed in a base fluid. The purpose of a hybrid nanofluid is to improve thermal properties compared to a base fluid or a mono-nanofluid through synergistic effects [2,3]. For heat transfer applications, hybrid nanofluid is expected to improve the heat transfer performance, pressure drop, and stability by trade-off the

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advantages and disadvantages of the nanoparticles used [4,5]. Based on a review on hybrid nanofluid done by Sarkar *et al.,* [2] proper hybridisation of hybrid nanofluid shows likely for heat transfer enhancement. Further work is recommended in preparation and stability, characterisation, and applications to address long-term stability, increased pressure drop or pumping power, high viscosity, and unavailability of a suitable thermal conductivity model [6].

Typically, the thermophysical properties of nanofluids depend on the dispersed nanoparticle type, size, shape, concentration, base fluid, operating temperature, and surfactant addition [7-13]. On the other hand, the hybrid nanofluid's synergetic effect is proportional to the mixing ratio. [1,2, 14]. A good synergetic effect of hybrid nanofluid is significant to enhance the thermal properties of the prepared hybrid nanofluid. As a result, the synergetic effect of hybrid nanofluid can be determined by optimising the mixing ratio of the nanoparticles [15-17].

Most researchers use the One factor at a time (OFAT) method to determine the best mixing ratio of hybrid nanofluid. OFAT method is a method of designing experiments involving the testing of factors, or causes, one at a time instead of multiple factors simultaneously. The traditional OFAT approach for optimisation has three serious downsides, which are (a) leading to an unnecessarily large number of experimental runs, (b) unable to study interactions among the factors, (c) time consuming to conduct a large number of experiments [18-20]. As a result, the best response value from the selected mixing ratio shows the best mixing ratio for the respective hybrid nanofluid. Therefore, this study aims to comprehensively optimise the mixing ratio of hybrid nanofluid based on thermal conductivity and viscosity based on statistical analysis.

2. Methodology

2.1 Preparation of Hybrid Nanofluid

A two-step method was used to produce the hybrid nanofluid using Eq. (1). First, a balance is used, AS 310.R2 PLUS Analytical Balance (Figure 1), to prepare the hybrid nanofluid. Various concentrations of 0.3vol% with different ratios of TiO_2 and GNP, which were 1:9 up to 9:1 dispersed in distilled water. Three mixing ratios of hybrid nanofluid had been prepared for each concentration which are 1:9, 1:1, and 9:1. The amount of surfactant used was 1:10 based on the amount of TiO_2.

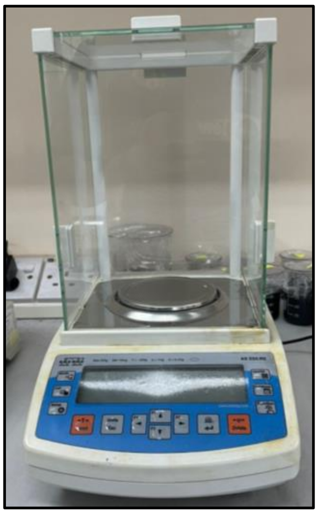

Fig. 1. Analytics Balance

Firstly, the required amount of surfactant was mixed with distilled water until it dissolved completely in water using a magnetic stirrer. The colour of the solution becomes apparent as the surfactant is completely dissolved. After that, the required amount of TiO_2 and GNP was mixed and let the solution stir for 15 minutes. The presence of surfactant dissolved in the distilled water help the nanoparticles and distilled water become homogeneous. However, using the magnetic stirrer was not enough to make the solution homogeneous. Therefore, the mixture was mixed using Ultrasonic Probe (FS-1200 N, frequency: 20 kHz, power output: 1200 W, 18mm probe) for 90 minutes to achieve a better homogeneous solution. The setup for the ultrasonic probe is shown in Figure 2.

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

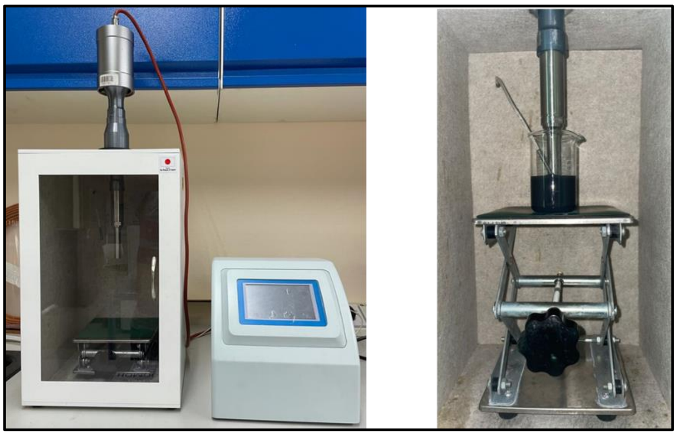

Fig. 2. Ultrasonic probe

2.2 Design Expert 11 Setup

In this study, the experimental design of the operating conditions is performed by the Design Expert 11 software. In this software, a set of DOE is developed to study the relationship of more than one variable that affects a response or several responses. This method is more efficient than OFAT because more information can be collected with fewer experiments. There are three steps involved in DOE: planning, execution, and analysis.

At first, Full Factorial Design is used to develop the DOE. During the planning process, two responses were used: thermal conductivity and viscosity, and two variables were used: the amount of TiO_2 (mixing ratio) and temperature, as shown in Table 1. The mixing ratio is determined by the amount of $TiO_{2,}$ which is calculated based on Eq. (1). Two levels were used for each variable. Each independent variable is varied over two levels. Each variable's low and high levels were -1 and +1, respectively.

Furthermore, used five replications with a center point to increase the experiment's sensitivity and the precision of the developed model. Therefore, the software listed 25 experiments, as shown in Table 2. After collecting the response, analysis is done by following these steps

i. Chose a transformation if desired. Otherwise, leave the option at "None".
ii. Used the appropriate model. The Fit Summary button displays the sequential F-tests, lack-of-fit tests, and other adequacy measures to assist in selecting the appropriate model.
iii. Perform the analysis of variance (ANOVA).
iv. Inspect various diagnostic plots to validate the model statistically.
v. If the model looks good, generate model graphs.

Table 1
Independent variable of DOE

Level of value	(A) Mixing Ratio (TiO_2:GNP)	(B) Temperature (°C)
-1	1:9	30
+1	9:1	60

Table 2
DOE for Full Factorial Design 0.3% concentration

Run	Mixing Ratio		Temperature (°C)
	TiO_2(g)	Mixing Ratio (TiO_2:GnP)	
1	0.475276	9:1	30
2	0.0528084	1:9	60
3	0.475276	9:1	60
4	0.264042	1:1	45
5	0.0528084	1:9	30
6	0.264042	1:1	45
7	0.475276	9:1	60
8	0.475276	9:1	30
9	0.475276	9:1	60
10	0.0528084	1:9	60
11	0.0528084	1:9	30
12	0.0528084	1:9	60
13	0.0528084	1:9	30
14	0.264042	1:1	45
15	0.475276	9:1	60
16	0.264042	1:1	45
17	0.264042	1:1	45
18	0.475276	9:1	30
19	0.475276	9:1	30
20	0.0528084	1:9	30
21	0.0528084	1:9	60
22	0.0528084	1:9	60
23	0.475276	9:1	30
24	0.0528084	1:9	30
25	0.475276	9:1	60

ANOVA analyses the data to study the relationship between variables and responses. A good model must be significant, and the lack of fit must be insignificant. The Adjusted R^2 value should not be less than 0.2 with the Predicted R^2 value to ensure the accuracy of the generated model terms. Furthermore, if there is a curvature presence in the results, the design needs to be upgraded to a Response Surface Design (RSM). If the augmentation is needed, choose Central Composite Design (CCD) with one run per axial (star) point, zero center point as the center point had been added from the previous design, and face-centered alpha where the point selected is within the level set before.

2.3 Thermal Conductivity Measurement

The thermal conductivity of nanofluid was measured using KD2 Pro, which was a transient hot-wire method. This study used a KS1 sensor, and it had an uncertainty of ±10.0% as it was the most suitable sensor for liquid analysis. Before starting the thermal conductivity measurement, the sample was put in an ultrasonic bath (Elmasonic S100H) to ensure that the sample was homogeneous to have an accurate measurement. The temperature involved was 30 to 60°C. The water bath function was to control the temperature of the sample. The sample test tube was put in a water bath to ensure the temperature was in an equilibrium state. Collect a total of five readings for each sample. The thermal conductivity setup is as in Figure 3.

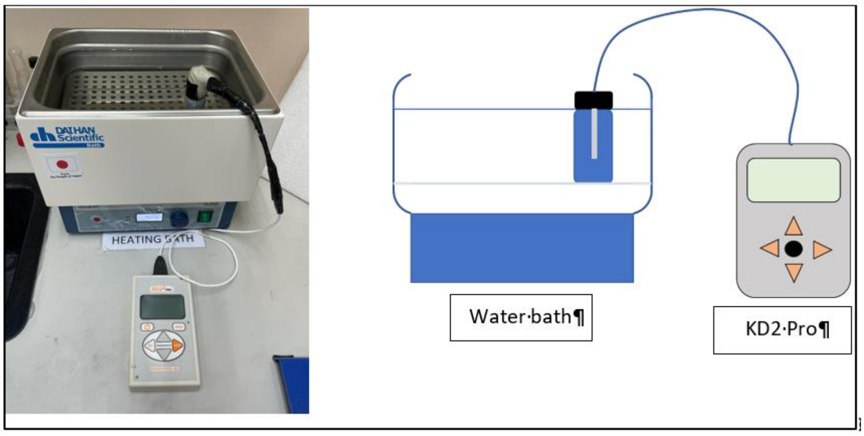

Fig. 3. Thermal conductivity measurement setup

2.4 Dynamic Viscosity Measurement

This experiment used Daihan Scientific Viscometer WVS-2M to measure the viscosity of hybrid nanofluid. The viscometer setup is as Figure 4. Based on the manual given by the manufacturer, the sample with low viscosity should use a large dimensioned rotor (No.0 – No.2) and fast rotating speed. Therefore, in this experiment, the rotor SP is No. 0, the rotating speed 60RPM, 30 seconds reading with 10mPa.s maximum capacity. The spindle LV1 used is suitable for low-viscosity liquid. The volume of the liquid sample must be sufficient to immerse the spindle to ensure the measurement's precision, as shown in Figure 4. The viscometer was compared with distilled water at 30°C for calibration. The percentage error is only 5% with the actual dynamic viscosity.

Fig. 4. Viscometer measurement setup

3. Results and Discussions

3.1 Overall Results and ANOVA

The experiments were done by following the sequence of runs provided by Design Expert 11 to prevent bias in the results obtained. The thermal conductivity and viscosity of the hybrid nanofluid prepared are shown in Table 3. Those results will be used in the Analysis of Variance (ANOVA) to develop models for each concentration.

Table 3
DOE for Full Factorial Design 0.3% concentration

Run	Mixing Ratio		Temperature (°C)	Thermal Conductivity (W/m.K)	Viscosity (mPa.s)
	TiO_2(g)	Mixing Ratio (TiO_2:GNP)			
1	0.475276	9:1	30	0.818	0.9
2	0.0528084	1:9	60	1.147	0.83
3	0.475276	9:1	60	1.016	0.8
4	0.264042	1:1	45	0.9552	0.86
5	0.0528084	1:9	30	0.958	0.94
6	0.264042	1:1	45	0.9361	0.86
7	0.475276	9:1	60	1.048	0.79
8	0.475276	9:1	30	0.748	0.89
9	0.475276	9:1	60	1.0526	0.8
10	0.0528084	1:9	60	1.032	0.82
11	0.0528084	1:9	30	0.983	0.93
12	0.0528084	1:9	60	1.043	0.85
13	0.0528084	1:9	30	1.212	0.95
14	0.264042	1:1	45	0.871	0.86
15	0.475276	9:1	60	1.0985	0.82
16	0.264042	1:1	45	1.086	0.87
17	0.264042	1:1	45	1.2472	0.88
18	0.475276	9:1	30	0.964	0.89
19	0.475276	9:1	30	0.783	0.89
20	0.0528084	1:9	30	0.979	0.91
21	0.0528084	1:9	60	0.984	0.83
22	0.0528084	1:9	60	1.152	0.83
23	0.475276	9:1	30	0.895	0.87
24	0.0528084	1:9	30	0.951	0.94
25	0.475276	9:1	60	0.999	0.81

All terms available for the thermal conductivity ANOVA analysis are shown in the half-normal plot in Figure 5. The available terms are A – TiO_2, B – Temperature, and AB. The terms are selected from the largest effect to the lowest (right to the left). Therefore, the ANOVA generated is shown in Table 4. P-values less than 0.0500 indicate model terms are significant. In this case, A and B are significant model terms. Values greater than 0.1000 indicate that the model terms are not significant. Therefore, AB is not significant and can be removed to improve the model.

Furthermore, curvature appears insignificant and needs to remove to simplify the analysis. After removing the insignificant term, the final ANOVA for analysis is shown in Table 5. The Model F-value of 6.52 implies that the model is significant. The Lack of Fit F-value of 1.50 implies that the Lack of Fit is not significant relative to the pure error. Non-significant lack of fit is good. The Predicted R^2 of 0.2191 is in reasonable agreement with the Adjusted R^2 of 0.3152, where the difference is less than 0.2. Adeq Precision measures the signal-to-noise ratio. A ratio greater than 4 is desirable. This model can be used to navigate the design space. In Eq. (2), the thermal conductivity of hybrid nanofluid is suggested in volume fraction and temperature. The equation can be used to make predictions about the response for given levels of each factor.

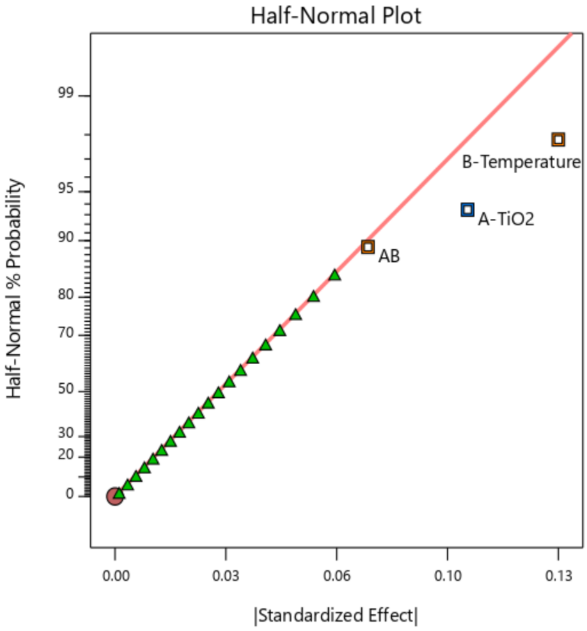

Fig. 5. Half-Normal plot for thermal conductivity analysis 0.3% concentration

Table 4
Thermal conductivity ANOVA for 0.3% Concentration

Source	Sum of Squares	df	Mean Square	F-value	p-value	Source
Model	0.1607	3	0.0536	5.45	0.0066	significant
A-TiO_2	0.0519	1	0.0519	5.28	0.0325	
B-Temperature	0.0821	1	0.0821	8.35	0.0091	
AB	0.0267	1	0.0267	2.72	0.1147	
Curvature	0.0027	1	0.0027	0.2740	0.6064	
Pure Error	0.1965	20	0.0098			
Cor Total	0.3599	24				

Table 5
Selected thermal conductivity ANOVA for 0.3% Concentration with fit statistic

ANOVA						
Source	Sum of Squares	df	Mean Square	F-value	p-value	
Model	0.1340	2	0.0670	6.52	0.0060	significant
A-TiO_2	0.0519	1	0.0519	5.05	0.0349	
B-Temperature	0.0821	1	0.0821	7.99	0.0098	
Residual	0.2259	22	0.0103			
Lack of Fit	0.0294	2	0.0147	1.50	0.2479	not significant
Pure Error	0.1965	20	0.0098			
Cor Total	0.3599	24				
Fit Statistics						
R^2	0.3722					
Adjusted R^2	0.3152					
Predicted R^2	0.2191					
Adeq Precision	25.0860					

$$Thermal\ conductivity = 0.9983 - 0.0509A + 0.0641B \quad (2)$$

Next, for the dynamic viscosity ANOVA analysis, all terms available are shown at the half-normal plot as shown in Figure 6. The available terms are A – TiO_2, B – Temperature, and AB. The terms are selected from the largest effect to the lowest (right to the left). Therefore, the ANOVA generated is shown in Table 6. P-values less than 0.0500 indicate model terms are significant. In this case, A and B are significant model terms. Values greater than 0.1000 indicate that the model terms are not significant. Therefore, AB is not significant and can be removed to improve the model. Furthermore, curvature appears insignificant and needs to remove to simplify the analysis. After removing the insignificant term, the final ANOVA for analysis is shown in Table 7. The Model F-value of 99.96 implies that the model is significant. The Lack of Fit F-value of 0.42 implies that the Lack of Fit is not significant relative to the pure error. Non-significant lack of fit is good. The Predicted R^2 of 0.8683 is in reasonable agreement with the Adjusted R^2 of 0.8919, where the difference is less than 0.2. Adeq Precision measures the signal-to-noise ratio. A ratio greater than 4 is desirable. This model can be used to navigate the design space. In Eq. (3), the dynamic viscosity of hybrid nanofluid is suggested in terms of volume fraction and temperature. The equation can be used to make predictions about the response for given levels of each factor.

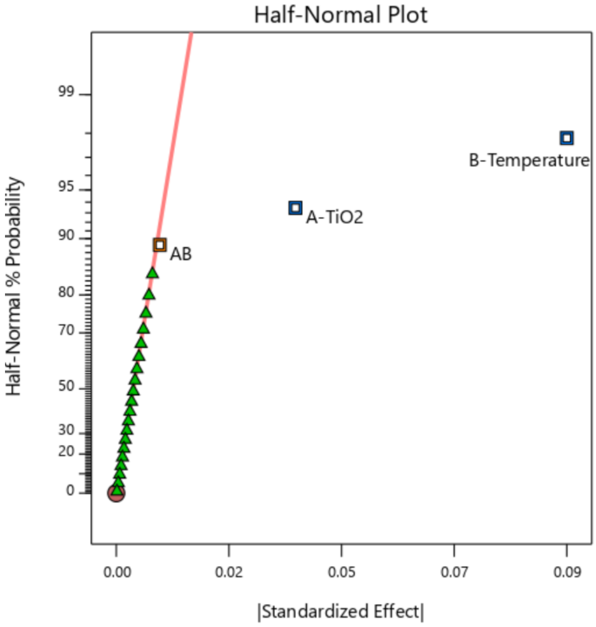

Fig. 6. Half-Normal plot for dynamic viscosity analysis 0.3% concentration

Table 6
Dynamic viscosity ANOVA for 0.3% Concentration

Source	Sum of Squares	df	Mean Square	F-value	p-value	Source
Model	0.0505	3	0.0168	123.76	< 0.0001	significant
A-TiO_2	0.0068	1	0.0068	50.33	< 0.0001	
B-Temperature	0.0432	1	0.0432	317.98	< 0.0001	
AB	0.0004	1	0.0004	2.98	0.0998	
Curvature	9.000E-06	1	9.000E-06	0.0662	0.7996	
Pure Error	0.0027	20	0.0001			
Cor Total	0.0532	24				

Table 7
Selected dynamic viscosity ANOVA for 0.3% Concentration with fit statistic

ANOVA						
Source	Sum of Squares	df	Mean Square	F-value	p-value	
Model	0.0501	2	0.0250	175.81	< 0.0001	significant
A-TiO_2	0.0068	1	0.0068	48.05	< 0.0001	
B-Temperature	0.0432	1	0.0432	303.57	< 0.0001	
Residual	0.0031	22	0.0001			
Lack of Fit	0.0004	2	0.0002	1.52	0.2425	not significant
Pure Error	0.0027	20	0.0001			
Cor Total	0.0532	24				

Fit Statistics	
R^2	0.9411
Adjusted R^2	0.9358
Predicted R^2	0.9220
Adeq Precision	31.4423

$$Dynamic\ viscosity\ =\ 0.8648 - 0.0185A - 0.0465B \tag{3}$$

3.2 Optimisation

The objective of the optimisation is to determine which mixing ratio of hybrid nanofluid from each concentration has the best performance based on thermal conductivity and dynamic viscosity. Therefore, the optimisation condition is set as shown in Table 8. The level of importance of a goal can be changed based on the optimisation objectives. The default is for all goals to be equally important at three pluses (+++), and the most important is five pluses (+++++). To achieve the best parameter for the mixing ratio and temperature of hybrid nanofluid, the target for optimisation is high thermal conductivity with a high level of importance and low dynamic viscosity with the default setting of the level of importance.

Table 8
Condition for optimization

Parameter	Target	Level of Importance
A: TiO_2 (g)	In range	3
B: Temperature (°C)	In range up to 40°C	3
Thermal conductivity (W/m K)	Maximize	5
Dynamic Viscosity (mPa.s)	Minimize	3

The optimal value suggested by the software for each concentration is shown in Table 9. Solution number 5 was the best value for each concentration. These values were picked from the previous model generated and goals set. From this table, the best parameter is 0.3vol% with 0.1053g of TiO_2 equivalent to 1:4(TiO_2:GNP) mixing ratio because it has the highest thermal conductivity compared to others. Dynamic viscosity is considered less important than thermal conductivity for this selection despite higher viscosity than the other solutions.

Table 9
The optimal values for 0.3vol% suggested by the software

Solution	TiO_2(g)	Temperature (°C)	Thermal conductivity (W/m K)	Dynamic viscosity (mPa.s)
1	0.1228	40	1.0111	0.8927
2	0.1213	40	1.0114	0.8928
3	0.1244	40	1.0107	0.8925
4	0.1303	40	1.0093	0.8920
5	0.1053	40	1.0153	0.8942
6	0.1420	40	1.0064	0.8910
7	0.1525	40	1.0039	0.8901
8	0.2004	40	0.9923	0.8859
9	0.2391	40	0.9830	0.8825

To validate the optimum value generated from the software, other thermal conductivity and dynamic viscosity experiments were done with 0.3vol% concentration, 1:4(TiO_2-GNP) mixing ratio, and 40°C. The results of the experiments are then compared with results generated from Eq. (2) and Eq. (3). Based on Figure 7 and Figure 8, The difference between the measured and predicted values of thermal conductivity and dynamic viscosity was less than 5%. Therefore, the model develops using this technique is acceptable. Graphene-based nanoparticles have a higher thermal conductivity compared with other nanofluids. Therefore, the presence of more GNP has a better thermal conductivity of hybrid nanofluid compared with a lower mixing ratio of GNP. A study by Vărdaru *et al.,* [21] verified that the presence of more graphene-based nanoparticles in the hybrid nanofluid has better thermal conductivity compared with other hybrid nanofluids with a lower mixing ratio of graphene-based nanoparticles.

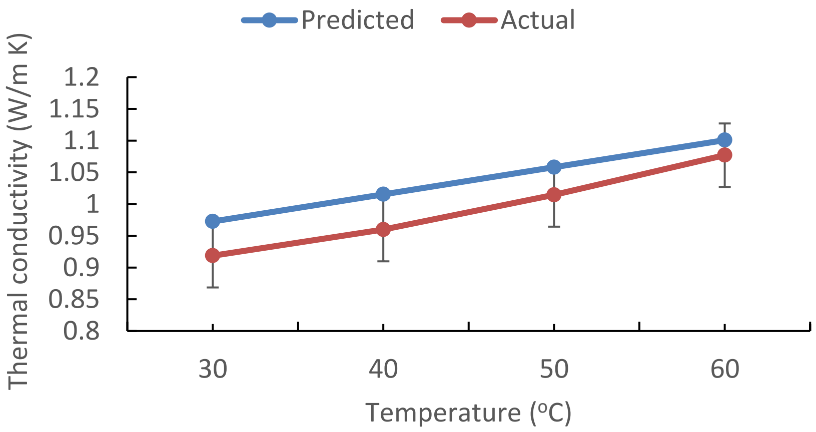

Fig. 7. Thermal conductivity predicted vs actual

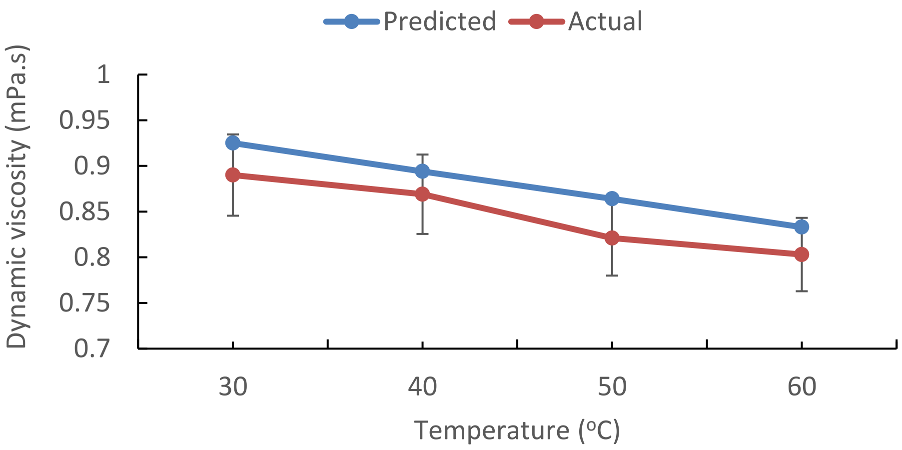

Fig. 8. Dynamic viscosity predicted vs actual

4. Conclusions

The experiment objective was to optimise the mixing ratio of hybrid nanofluid with thermal conductivity and viscosity using Design Expert 11. The optimised mixing ratio and concentration of the prepared hybrid nanofluid are 0.3vol% with a 1:4(TiO_2-GNP) mixing ratio. DOE method shows fewer experiments need to be done while having enough information for the optimisation process. The chosen mixing ratio was based on the model developed by the ANOVA. The difference between the measured and predicted values of thermal conductivity and dynamic viscosity was less than 5%. Therefore, the model develops using this technique is acceptable.

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