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ORIGINAL RESEARCH PAPER

Modeling of Effective Thermal Conductivity and Viscosity of Carbon Structured Nanofluid

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Abstract

This paper was aimed to address the modeling of effective thermal conductivity and viscosity of carbon structured nanofluids. Response surface methodology, D_optimal design (DOD) was employed to assess the main and interactive effects of temperature (T) and weight percentage (wt %) to model effective thermal conductivity and viscosity of multi wall and single wall carbon nanotube, CVD and RGO Graphene and nanoporous Graphene sheet. The second-order polynomial regression model was proposed for effective thermal conductivity and viscosity as a function of relevant investigated parameters. Effective thermal conductivity and viscosity of nanofluids measured using an accurate transient short hot wire system and a viscometer, respectively. nanofluids was prepared using two-step method and showed a desirable stability. In general, Graphene nanosheets have more effective thermal conductivity and viscosity compared to carbon nanotube because of variation in shape and likely size.

1. Introduction

Searching for new techniques to improve cooling performance of conventional heat transfer fluids performs a key role in various industrial applications including power generation, chemical processes, heating or cooling processes, and microelectronics. Using solid millimeter or micrometer-sized particles, Maxwell [1,2] present the well known idea of enhancing the thermal conductivity of fluid.

But particles of millimeter or micrometer size cause some problems such as severe pressure drops, sedimentation, clogging of channels.

Therefore, it seems that they are not applicable for many practical applications. Nanotechnology provides with an opening to synthesis a new generation of heat transfer fluids called nanofluid.

Indeed, nanometer-sizesd particle have been dispersed in conventional heat transfer fluid in order to improve performance of cooling devices. Choi [3] introduced an engineered Nanofluids with superior thermal properties compared to the conventional heat transfer fluids. Many researchers reported many experimental and and theoretical works which dedicated to determine thermal conductivity of nano-

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fluids [4 -12].

Das et al. [5] reported the thermal conductivity of Al_2O_3 and CuO nanoparticles suspended in water as a function of temperature. The results showed that thermal conductivity increased with increasing nanofluids temperature as well as particle concentrations.

Moreover, for Al_2O_3/water nanofluids, the results showed that the predicted value by the Hamilton–Crosser model (H–C model) [13] agreed well with the measured value at room temperature only. For CuO/water nanofluids, the H–C model gives a value less than that of the measured value at room temperature. At elevated temperature, both Al_2O_3/water and CuO/water nanofluids disagree with the H–C model.

Kumar et al. [14] suggested a comprehensive model which describes the mechasim of in thermal conductivity enhancement of a nanofluid with considering variation in particle size, particle volume fraction, and temperature. This enhancement is inversely and linearly proportional to the radius of the particle and nanoparticle concentration respectively. Leong et al. [15] proposed a new model for predicting thermal conductivity of nanofluids.

They demonstrated that the interfacial layer and the particle size are one of the major mechanisms for increasing the thermal conductivity of nanofluids.

Ko et al. [16] examined the pressure drop and viscosity of carbon nanotubes (CNT) dispersed in distilled water flowing through a horizontal tube. The results showed that the nanofluids prepared by the acid treatment (TCNT) have much smaller viscosity than the ones made with surfactant (PCNT). Corcione [12] proposed two empirical correlations for predicting the effective thermal conductivity and dynamic viscosity of nanofluids, based on a high number of experimental data available in the literature, are proposed and discussed. The results show that the ratio between the thermal conductivities of the nanofluid and the pure base liquid increases as the nanoparticle volume fraction and the temperature are increased, and the nanoparticle diameter is decreased. carbon nanoparticles (CNT and Graphene) can bundle together easily because of their non-reactive surface properties, high van der Waals interaction forces [17,18],[19] (Park et al. 2002) [19] (Park et al. 2002) large specific surface areas [19].

Therefore, the stability of carbon structured nanofluid could be a critical issue of nanofluid preparation. This study attempts to propose an empirical model witch predicted the effective thermal conductivity and viscosity of carbon structured nanofluids containing single and multi wall carbon nanotubes, CVD, RGO and nano-porous Graphene using statistical design of experiments. Response surface methodology-D-O ptimal model has been utilized to investigate the influence of weight percentage, temperature and five type of carbon structured nanofluid on the effective thermal conductivity and viscosity.

2. Methodology

Experimental design is widely used for controlling the effects of parameters in many processes. Its usage decreases the number of experiments, time and material resources. Furthermore, the analysis performed on the results is easily realized and the experimental errors are minimized. Statistical methods measure the effects of change in operating variables and their mutual interactions on the process [20]. Response surface methodology (RSM) is a group of mathematical and statistical techniques based on an efficient experimental strategy that is used for developing, improving, optimizing the processes and modeling and analyzing engineering problems.

It also applies to evaluate the relative significance of some affecting factors even in the presence of complex interactions [20, 21]. Two important models are commonly used in RSM as follow: i) the first-degree model,

$$Y = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \varepsilon \qquad (1)$$

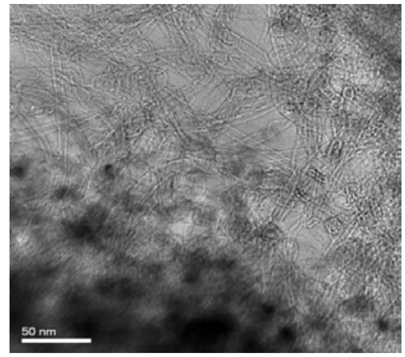
And, ii) the second–degree model,

$$Y = \beta_0 + \sum_{i=1}^{k} \beta_i x_i + \sum_{1<i<j}^{k} \beta_{ij} x_i x_j + \sum_{<i<j}^{k} \beta_{ii} x_i^2 + \varepsilon \qquad (2)$$

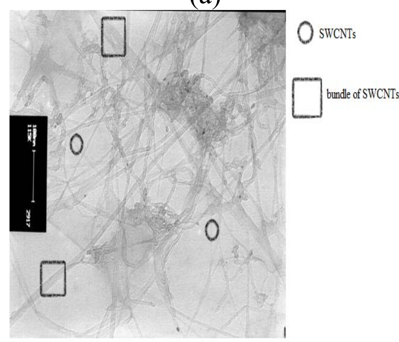
where k, β_0, β_i, x_i, β_{ii}, β_{ij} and ε represent the number of variables, constant term, coefficients of the linear parameters, variables, the coefficients of the quadratic parameter, the coefficients of the interaction parameters and residual associated to the experiments, respectively [22]. Therefore, in the present work experiments were designed on the basis of experimental design technique using response surface design method.

3. Carbon nanoparticles

The size and morphology of the carbon nanoparticles were examined by using TEM images. The TEM image of the different carbon nanoparticle is shown in figure 1. In general, it appears that structure of the all carbon nanoparticles have a nano at one dimension at least. Size of MWCNTs is 10-30 nanometers in diameter and few microns in length. Individual SWCNT which points out using circles has a diameter less than 3 nm and few microns in length. Transparent region in Fig. 1c points out few layered Graphene formed while dark one indicated that the more layers of Graphene sheets were grown. Generally, for CVD Graphene, observed layer is less than five numbers. Number of layers for RGO Graphene sheet is between 8-10 (it’s not shown in the figure).



(a)



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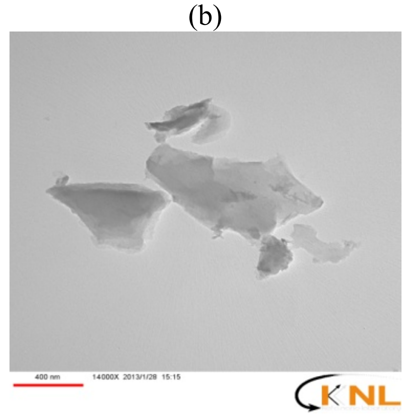

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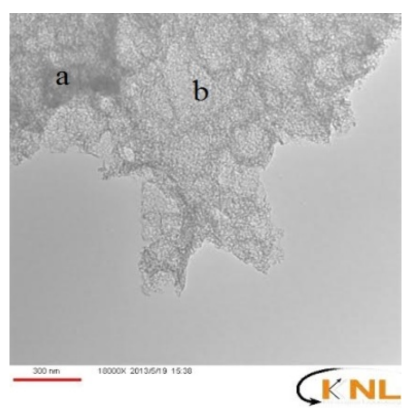


(d)

Fig. 1. TEM image of image of nanoparticle (a) MWCNTs (b) SWCNTs (c) CVD Geraphen (d) nanoporous Geraphen

4. Nanofluid preparation

A two-step process was utilized in order to prepare the sample of CNT and Graphene nanofluids. Nanoparticles including multi-wall carbon nanotube (MWCNT), single-wall carbon nanotube (SWCNT), CVD Graphene sheet (G_1), RGO Graphene sheet (G_2) and nano-Porous Graphene sheet (G_3) with the 0.5 and 1% weight percentages mixes up with distilled water. Then, that was placed in the ultrasonic bath for 30 minutes. According to figure 2, MWCNT/Water sample has a good stability and dispersion for MWCNT in water. Other samples were similarly examined to check the stability and the quality of nanoparticle dispersion in the base fluid. Briefly, good stability was observed after passing 5 days (120 hours).

In general, it appears that structure of the all carbon nanoparticles have a nano at one dimension at least. Size of MWCNTs is 10-30 nanometers in diameter and few microns in length.

Individual SWCNT which points out using circles has a diameter less than 3 nm and few microns in length. Transparent region in Fig. 1c points out few layered Graphene formed while dark one indicated that the more layers of Graphene sheets were grown. Generally, for CVD Graphene, observed layer is less than five numbers. Number of layers for RGO Graphene sheet is between 8-10 (it's not shown in the figure).

(a) (b)

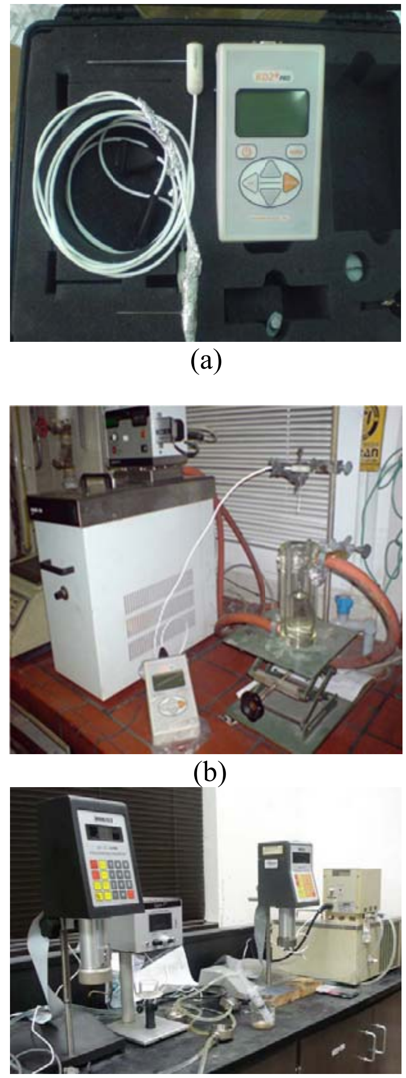
Fig. 2. sample (a) immediately after preparation (b) after 5 days

5. Thermal conductivity and viscosity measurement

The experimental apparatus was calibrated by measuring the thermal conductivity of deionized water. The sample temperature was monitored using a thermocouple inside the vessel. A vessel containing the sample nanofluid was placed in the bath of thethermalconductivity measurement. The sample nanofluid was kept for further 30 min to the bath temperature to ensure heat equilibrium. figure 3a and 3b illustrates KD_2 set up for measuring the thermal conductivity.

In order to understand rheological behavior of the samples, they were examined using Visco Elite set up fabricated by Fungilab Company.

Table 1 shows the variation of viscosity against the shear rate at highest concentration (1wt %) of nanoparticles. No significant variation is observed for the value of samples viscosity. Therefore, one can be concluded that the samples behave as a Newtonian fluid. Then, the viscosity of samples according to design of experiments were measured using a viscometer set up as pictured in figure 3c.



(a)

(b)

(c)

Fig. 3. Experimental set up for measuring (a) and (b) effective thermal conductivity (c) effective viscosity

6. Results and discussion

Experiments designed by surface response method have been performed over a range of temperature and particle weight percentage in order to investigate the effective thermal conductivity and viscosity of carbon structured nanofluid. The samples consist of multi and single wall carbon nanotube, CVD, RGO and nano-porous Graphene sheet dispersed in distilled water. The set of experiments designed by Response surface methodology- D-Optimal method and Responses (the effective thermal conductivity and viscosity) to obtain a model is presented in table 2. The limitations of model are as follows: (1) temperature range between

15^{0}C and 35 ^{0}C and (2) weight percentage range between 0.01 wt% and 1 wt%. All the measurements were obtained after adjusting the KD_2 and viscometer instrument with water. As shown in figure 4, relatively good agreement at temperatures of 15^{0}C, 25^{0}C and 35^{0}C between the measured and the reference values are observed. The results which predicted by model are present and discuss in this section.

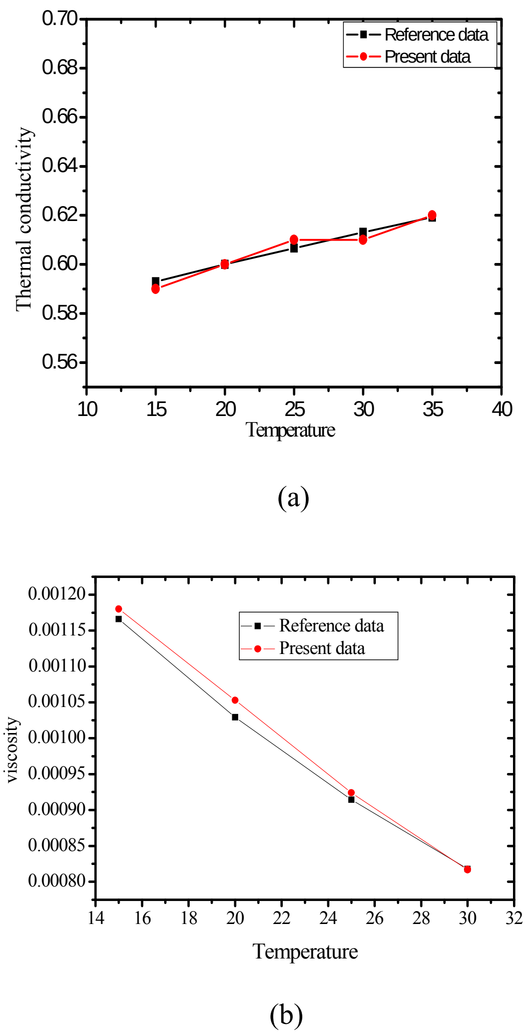


Fig. 4. Comparison of KD_2 and viscometer with references

6.1. Verification of the models

The adequacy verification of the model includes the test for significance of the regression model, test for Significance on model coefficients, and test for lack of fit. Therefore, the adequacy of the proposed models have been tested by the analysis of variance and shown in table 3 and table 4. Finally, Actual measured data according to design of experiment against to predicted model are pictured in figure 5. As clearly observed, the experimental data (actual) are in good agreement with corresponding predicted. Therefore the model is reliable and can predict effective thermal conductivity and viscosity of samples.

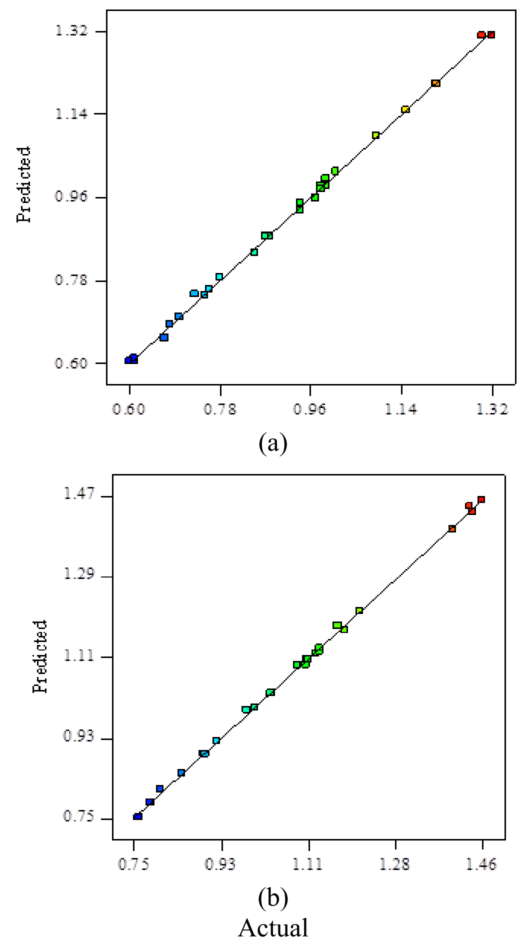
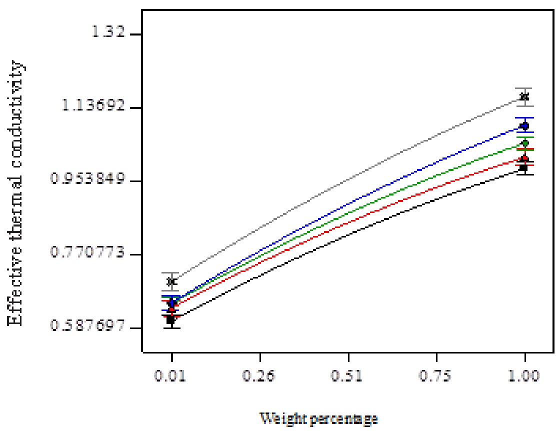


Fig. 5. Comparison of Predicted versus actual data (a) Effective thermal conductivity (b) viscosity

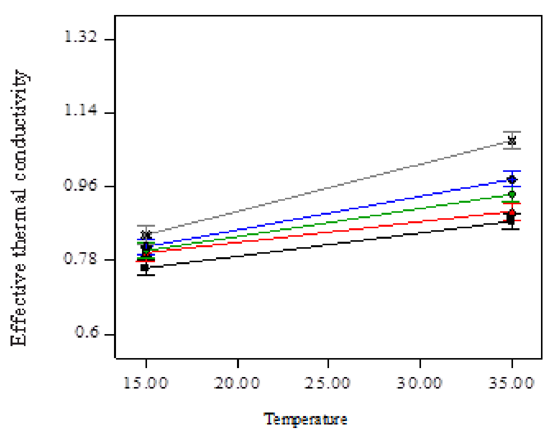
6.2. Effective thermal conductivity

Figure 6a shows the prediction of effective thermal conductivity variation with the weight percentage for different carbon structured nanofluids. Weight percentage of nanoparticles linearly increases the thermal conductivity of all sample nanofluids. Higher weight percentage augments the number of nanoparticle per unit of volume and therefore, results in elevated interface area between nanoparticle and base fluid. At such situation, mixing action, micro convections and more local fluctuation arised from high temperature causes stronger interaction and collision among nanoparticles.

Therefore, all these factors cause an enhancement of ballistic conduction heat transport compared to the base fluid. Comparison of two CNT indicates that the shape of single SWCNT (1-2 nm in diameter) is similar to the MWCNT (20-30 nm in dimeter).



(a)



(b)

Fig. 6. Thermal conductivity variations of samples vs. (a) wt %. At 25 ^{0}C (b) temperature At wt=0.51%

But there is a difference in size. This causes intensified fluctuation and mixing action through the nanofluid smaller nanoparticle (SWCNT). Moreover, there is greater interfacial area between nanoparticles and fluid in SWCNT/water compared to MWCNT/water. Therefore effective thermal conductivity of SWCNTs/water nanofluids increases compared to MWCNT/water. There are similar circumstances for G_1 and G_2. In general, it is observed an enhancement in effective thermal conductivity of Graphene compared to CNT because of variation in shape of nanparticles. Graphene has more effective surface and therefore, greater interfacial area between nanoparticles and base fluid. For a given weight percentage, porous nature of G_3 causes a significant increase in number of nanosheets. This amplifies Physical interactions between nanoparticles and base fluids which develop heat transport mechanisms of G_3/Water nanofluid compared to G_1 and G_2 one.

The results demonstrated that for wt=0.01% the use of G_3 dispersed in base fluid give 16.3% thermal conductivity enhancement compared to MWCNT in base fluid. While, it be can seen an increase about 18.2% at the thermal conductivity for wt=1%. Therefore, more interaction between G_3 particles is observed compared to MWCNT particles at higher concentration.

As shows in figure 6b, our experimental data indicate that effective thermal conductivity of the samples increases with temperature. Enhancement of effective thermal conductivity is mainly related to the intensified Brownian motions of dispersed nanoparticles. As temperature of nanofluid is elevated, the dispersed nanoparticles change residence faster in the distilled water. Consequently, energy transport through the nanofluids becomes strong and therefore, effective thermal conductivity increases.

Indeed, ballistic heat conduction of nanoparticles enhances thermal diffusion. As pictured in the figure there is less sensitivity related to type of carbon nanoparticle at T=15^{0}C. It seems at low temperatures some nano scale effects such as interaction and collision of nanoparticles caused by Brownian motions and shape of nanoparticles vanishes and other factor like interfacial area between nanoparticles and fluid play main role in limited enhancement of thermal conductivity (10.6%).

However, the collision of G_3 nanoparticles and the fluctuation of fluid improve the thermal conductivity of G_3/water compared to MWCNT/water nanofluid at T=35^{0}C about 21.7%. Finally, according to the results of figure 7, the weight percentage has more significant than the temperature in the effective thermal conductivity.

The following model with Analysis of variance tabulated at previous section is produced in order to calculate thermal conductivity of samples nanofluid:

Also, constant coefficients of the above model for the sample nanofluids are given in table 5.

$$k_{eff} = \begin{pmatrix} \beta_0 + \beta_1 Wt\% + \beta_2 T + \beta_{12} Wt\% \times T \\ + \beta_{11} Wt\%^2 \end{pmatrix} \times 10^{-3} \quad (3)$$

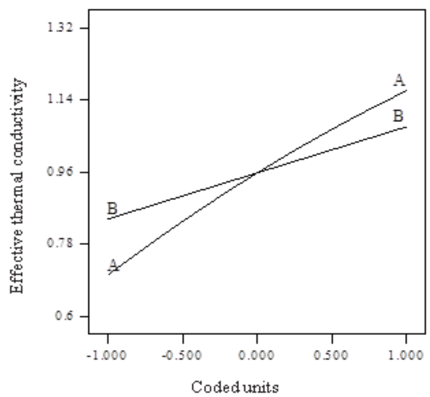
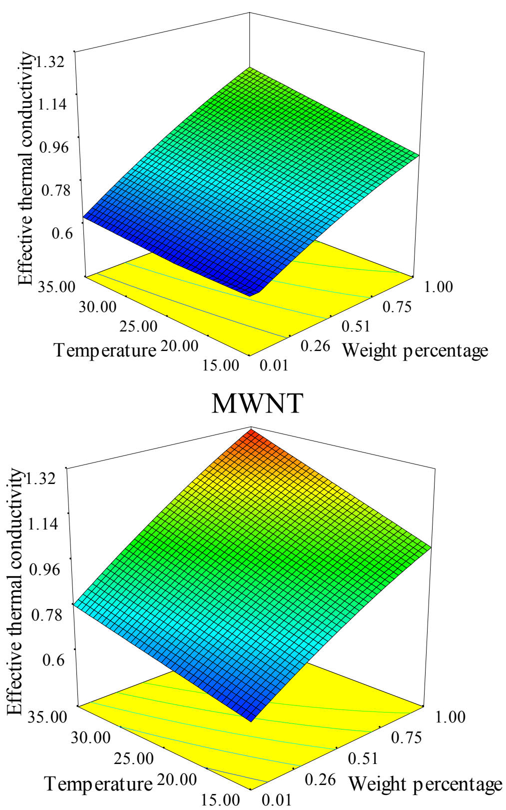


Fig. 7. Effective thermal conductivity for G_3/water vs. (A) Weight percentage and (B) Temperature (coded units)



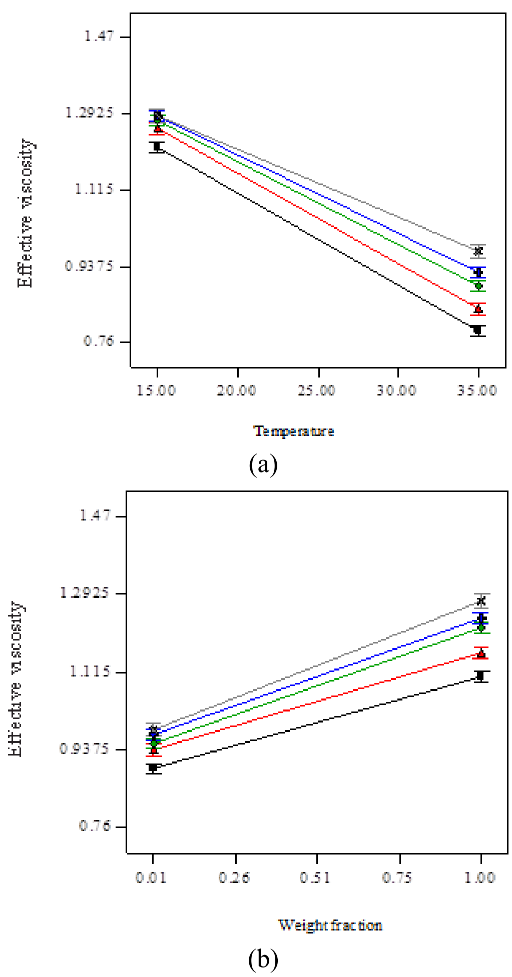
G-3

Fig. 8. Response surface plots showing the effect of two variables on Effective thermal conductivity. nanofluid containing (a) MWCNT and (b) G_3.

Further investigation of factors influence on the effective thermal conductivity carried out by the 3D response surface plots for MWCNT and G_2 dispersed in water nanofluid illustrated in figure 8. Indeed, the 3D response surface plots in the figures are simulations from Equations which describe the effect of the temperature and weight percentage on effective thermal conductivity.

6.3. Effective viscosity

Figure 9 shows the effective viscosity of sample nanofluids as a function of temperature and weight fraction. The results indicate that the viscosity of nanofluids significantly decreases with increasing nanofluid temperature.



(b)

Fig. 9. Effective viscosity variations of samples vs. (a) temperature at wt=0.51% (b) wt %. at 25 ^{0}C

All samples directly decrease with temperature. For a given weight percentage and low temperature

($T=15^0C$), there is no significant variation at the viscosity of different samples. This arises from the fact that some nano scale effects such as variation in size, shape and Brownian motion of nanoparticles for which result in an elevated momentum transfer of aqueous suspensions vanish.

In general, the thermal conductivity of fluids increases with an increase in the temperature but the trends vary for different cases. At $T=35^0C$, elevated momentum transfer of nanoparticles and therefore, enhanced effective viscosity, derived from collision and random motion of nanoparticles are achieved. Such factors could likely resists against reduction of viscosity of the base fluid (molecular momentum transfer) with temperature. Therefore, the effective viscosity of nanofluid enhances because of nano-vibration of particles. Porous nature of G_3 results in a considerable increase in number of nanosheets which present developed nano-fluctuations. Hence, at high temperature, later reason increases the effective viscosity of G_3/water nanofluid compared to other ones. For example, compared to MWCNT/water, while there is an augmentation of 6.14% at 15^0C, one encounters an enhancement of 23.65% at $T=35^0C$.

Figure 10 indicates our recent discussion on the interaction of temperature and type of nanoparticle. Dependency of effective viscosity to type of loading nanoparticles decreases at low temperature.

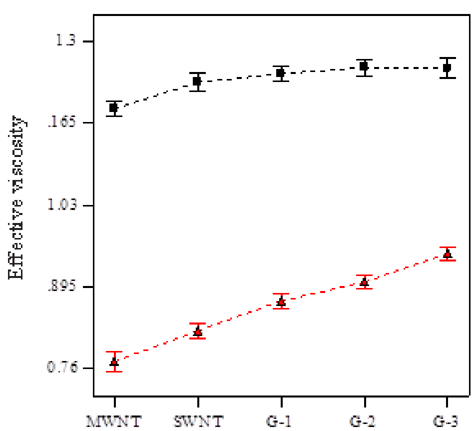


Fig. 10. effective viscosity vs. different type of carbon structured nanofluid (wt%=0.41)

The proposed model for the empirical relationship between the effective viscosity and the important factors (Temperature and weight percentage) was presented on the basis of the experimental results as follows:

$$\mu_{eff} = \left(\beta_0 + \beta_1 Wt\% + \beta_2 T + \beta_{12} Wt\% \times T\right) \times 10^{-3} \quad (4)$$

Constant coefficients are shown in table 6.

Figure11 shows 3D response surface plots extracted from Equations 4 and describe the effect of the temperature and weight percentage on effective thermal conductivity.

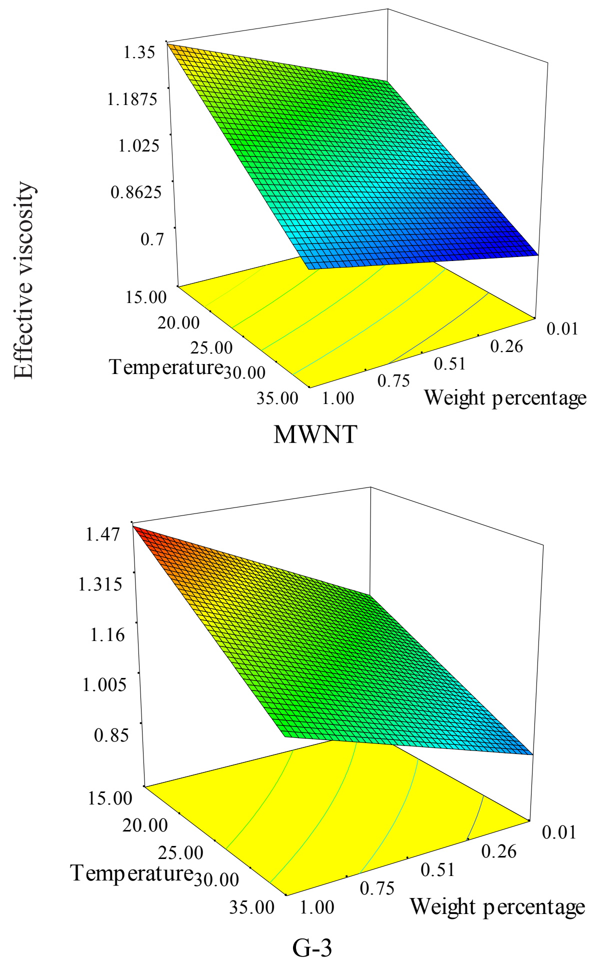


Fig. 11. Response surface plots showing the effect of two variables on Effective viscosity. nanofluid containing (a) MWCNT and (b) G_3

In figure 12, the effective viscosity of nanofluid containing MWCNT vs. wt% derived from Eq. (4) for different values of temperature compared with the predictions of the Brinkman equation [23]. Effective dynamic viscosity predicted by the Brinkman equation clearly fails when applied to the nanofluids. Other samples qualitatively show similar inconsistently.

Therefore, the conventional theory could not predict the variation of effective viscosity in nanofluid.

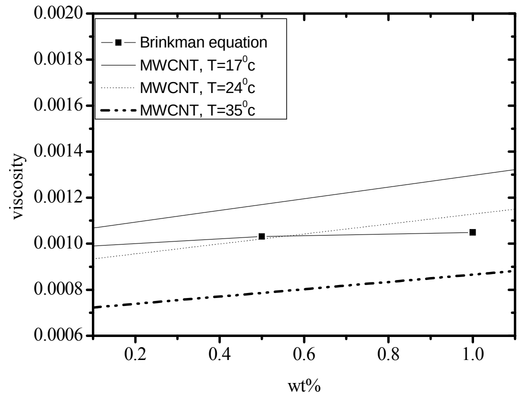


Fig. 12. Comparison of the model with Brinkman equation

7. Conclusion

According to design of experiments, experiments have been carried out to investigate the effect of weight percentage and temperature on carbon structured nanofluid. The model suggested by design expert are presented and discussed.

Nanofluids containing the SWCNT and MWCNT for which have the same shape and different size, smaller one (SWCNT) results in intensified fluctuation and mixing action through the nanofluid. Therefore effective thermal conductivity of SWCNTs/water nanofluids increases compared to MWCNT/water. More effective surface and greater interfacial area between nanoparticles and base fluid enhances effective thermal conductivity and viscosity of graphen nano sheet compared to carbon nano tubes. Porosity of nanoparticles (G_3) results in an increase in number of nanoparticles and Physical interactions between nanoparticles and base fluids. So this develops heat and momentum transfer and, therefore, an increase in effective thermal conductivity and viscosity. It could be concluded that at low temperatures some nano scale effects disappear and limited variation in effective thermal conductivity and viscosity are achieved.

Table 1
The viscosity variation of samples with the shear rate wt=1%.

Number	Shear rate(S^{-1})	MWCNT/Water kg m-1 s-1 @T=18 0C	SWCNT/Water kg m^{-1} s^{-1} @T=18 ^{0}C	G_1/Water kg m^{-1} s^{-1} @T=20 ^{0}C	G_2/Water kg m^{-1} s^{-1} @T=20 ^{0}C	G_3/Water kg m^{-1} s^{-1} @T=20 ^{0}C
1	10.38	1.23e-3	1.24e-3	1.33 e-3	1.35 e-3	1.36 e-3
2	29.48	1.20e-3	1.22e-3	1.34 e-3	1.33 e-3	1.33 e-3
3	46.61	1.19e-3	1.20e-3	1.35 e-3	1.35 e-3	1.38 e-3
4	62.72	1.18e-3	1.21e-3	1.32 e-3	1.36 e-3	1.35 e-3
5	87.84	1.18e-3	1.20e-3	1.33 e-3	1.31 e-3	1.36 e-3
6	103.9	1.18e-3	1.19e-3	1.38 e-3	1.36 e-3	1.33 e-3
7	110.0	1.21e-3	1.20e-3	1.32 e-3	1.34 e-3	1.34 e-3

Table 2
The set of designed experiments for different parameter.

Exp. No.	Wt (%)	Temperature (^{0}C)	Type of nano particle (TNP)	Effective Thermal conductivity (K_{eff})(w m^{-1}.K^{-1})	Effective Viscosity (μ_{eff}) (kg m^{-1} s^{-1})E-3
1	0.01	25	MWNT	0.61	0.895
2	1	35	G-3	1.3	1.103
3	1	15	G-3	1.01	1.461
4	0.01	25	MWNT	0.6	0.899
5	0.7525	25	G-2	0.99	1.181
6	1	35	G-1	1.15	0.999
7	0.2575	20	G-2	0.75	1.131
8	0.2575	25	SWNT	0.73	0.982
9	1	15	G-1	0.94	1.441
10	0.01	15	G-1	0.61	1.122
11	0.505	15	MWNT	0.76	1.212
12	0.01	35	G-2	0.7	0.809
13	1	35	G-2	1.21	1.032
14	0.01	35	G-1	0.68	0.789
15	1	25	MWNT	0.99	1.104
16	1	35	G-3	1.32	1.085
17	1	25	MWNT	0.98	1.108
18	0.505	25	G-3	0.97	1.131
19	0.01	35	SWNT	0.67	0.763
20	1	15	G-2	0.98	1.436
21	0.505	35	MWNT	0.88	0.7863
22	1	15	SWNT	0.94	1.401
23	1	35	SWNT	1.09	0.921
24	0.01	35	G-3	0.78	0.852
25	0.505	35	MWNT	0.87	0.786
26	0.505	20	G-1	0.85	1.168

Table 3
Analysis of variance for response surface reduced quadratic model of effective thermal conductivity and viscosity.

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob > F	
Model	1.067297	16	0.066706	365.6537	< 0.0001	significant
A-Wt %	0.76147	1	0.76147	4174.049	< 0.0001	
B-Temperature	0.113491	1	0.113491	622.109	< 0.0001	
C-Type of nanoparticle	0.086338	4	0.021585	118.3172	< 0.0001	
AB	0.006975	1	0.006975	38.2352	0.0002	
AC	0.004057	4	0.001014	5.559319	0.0155	
BC	0.006778	4	0.001694	9.288386	0.0030	
A^2	0.002115	1	0.002115	11.59357	0.0078	
Residual	0.001642	9	0.000182			
Lack of Fit	0.001292	5	0.000258	2.952837	0.1582	not significant
Pure Error	0.00035	4	8.75E-05			
Std. Dev.	0.013507	R-Squared		0.998464		
Mean	0.898462	Adj R-Squared		0.995733		
C.V. %	1.503308	Pred R-Squared		0.949223		
PRESS	0.054277	Adeq Precision		64.64786		

Table 4
Analysis of variance for response surface reduced quadratic model of effective viscosity.

Source	Sum of Squares	Source	Mean Square	F Value	p-value Prob> F	
Model	1.133616	15	0.075574	789.9628	< 0.0001	significant
A-Wt	0.22802	1	0.22802	2383.444	< 0.0001	
B-Temperature	0.610122	1	0.610122	6377.472	< 0.0001	
C-Type of nanoparticle	0.081075	4	0.020269	211.8635	< 0.0001	
AB	0.005436	1	0.005436	56.82034	< 0.0001	
AC	0.003899	4	0.000975	10.18964	0.0015	
BC	0.005507	4	0.001377	14.39004	0.0004	
Residual	0.000957	10	9.57E-05			
Lack of Fit	0.000779	6	0.00013	2.915512	0.1598	not significant
Std. Dev.	0.009781	R-Squared		0.999157		
Mean	1.061435	Adj R-Squared		0.997892		
C.V. %	0.921491	Pred R-Squared		0.981672		
PRESS	0.020795	Adeq Precision		92.1104		

Table 5.
Value of constant coefficients for effective thermal conductivity of equation (3).

		Type of nanoparticles				
		MWNT	**SWNT**	**G-1**	**G-2**	**G-3**
coefficient	β_0	0.537463	0.586049	0.552114	0.519929	0.489377
	β_1	0.327447	0.323441	0.347649	0.389817	0.408833
	β_2	0.002465	0.001827	0.003626	0.004981	0.008296
	β_1	0.006401	0.006401	0.006401	0.006401	0.006401
	β_1	-0.10261	-0.10261	-0.10261	-0.10261	-0.10261

Table 6.
Value of constant coefficients for viscosity of equation (4).

		Type of nanoparticles				
		MWNT	**SWNT**	**G-1**	**G-2**	**G-3**
coefficients	β_0	1.360976	1.393472	1.36361	1.357974	1.308054
	β_1	0.342731	0.355397	0.398791	0.399995	0.428523
	β_2	-0.01868	-0.01831	-0.01656	-0.01552	-0.01309
	β_{12}	-0.00526	-0.00526	-0.00526	-0.00526	-0.00526

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