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## Highlights

- Experimental study on rheological behavior of MWCNT-CuO/SAE50 nanolubricant
- Measuring viscosity at several concentrations, temperatures and the shear rates
- The highest viscosity reduction (-15%) occurred in a volume fraction of 0.0625%.
- A comparison was performed between the viscosities of different nanofluids.
- The viscosity sensitivity analysis for temperature and volume fraction has been performed.

ACCEPTED MANUSCRIPT

# Simultaneous effects of multi-walled carbon nanotubes and copper oxide nanoparticles on the rheological behavior of cooling oil: applicable for refrigeration systems

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## Abstract

This study discussed the rheological behavior of MWCNT-CuO(30%-70%)/SAE50 nanofluid experimentally at the volume fractions of 0-1 percent in the temperature range of 25-50 °C and the shear rate of 666- 7998s<sup>-1</sup>. Due to a nonlinear relationship between the shear rate and shear stress, nanofluid behavior can be considered close to the non-Newtonian fluids. Nanofluid viscosity reduces by adding nanoparticles at the volume fractions of 0.0625%, 0.125%, and 0.25% (maximum reduction of 15%). Nanofluid viscosity increases at the volume fractions higher than 0.5%, as the maximum viscosity increase, i.e. 14%, occurs at the volume fraction of 1%. Reduction of viscosity at low volume fractions and a small increase of viscosity at higher volume fractions show that this nanofluid can be used in the applications other than lubrication. This study presents for the first time a new mathematical relation for the relative viscosity of MWCNT-CuO(30%-70%)/SAE50. In the relation provided to predict the relative viscosity of nanofluid,  $R^2=0.9453$ , which indicates its accuracy. This study also discussed the sensitivity of the relative viscosity of nanofluid to temperature and volume fraction.

**Keywords:** Viscosity; Hybrid nano-lubricant; Experimental correlation; sensitivity analysis; Pumping power;

## 1. Introduction

Nanofluids are colloid solutions in which solid nanoparticles are suspended. The appropriate wettability and dispersion level of nanoparticles in base fluids leads to an improvement in the productivity of a nanofluid relative to a base fluid.

The results of the tests on heat transfer method for several nanofluids prove that performance of nanofluids in heat transfer generally exceeds what has been predicted theoretically. This fact is a fundamental discovery in the heat transfer problem [1-11]. Moreover, clustering of nanoparticles affects viscosity, pressure drop, and pumping power, which block flow and reduce nanofluid thermal performance. Many researchers studied viscosity of nanofluids [12-20], some of which are presented by Table 1.

Table 1. The result of the study of some researchers on viscosity variations

Researcher	nanofluid	Viscosity changes
A. Aghaei et al [21]	MWCNT-CuO/5w50	Max = +41% @ T= 55 °C, $\varphi=1\%$ Min = +0.11% @ T= 45 °C, $\varphi=0.05\%$
A. Alirezaie et al [22]	MWCNT(COOH-functionalized)-MgO/SAE50	Max = 9.2% @ T= 50 °C, $\varphi=1\%$ Min = +1.2% @ T= 50 °C, $\varphi=0.25\%$
M. H. Esfe et al [23]	Al <sub>2</sub> O <sub>3</sub> /10W40	Max = 131% @ T= 55 °C, $\varphi=2\%$ Min = +14% @ T= 65 °C, $\varphi=0.25\%$
M. Afrand et al [24]	MWCNT-SiO <sub>2</sub> /SAE40	Max = +75% @ T= 50 °C, $\varphi=1\%$ Min = +0.19% @ T= 25 °C, $\varphi=0.0625\%$
A. Asadi et al [25]	MWCNT- MgO/SAE50	Max = +65% @ T= 40 C, $\varphi=2\%$ Min = +14.4% @ T= 25 C, $\varphi=0.25\%$

The rheological behavior (a Newtonian/non-Newtonian model) of oil-based fluids changes by adding nanoparticles, which affect the performance of nanofluids. Determination of the rheological behavior of nanofluids allows avoiding significant design errors and neutralizing undesirable performance of oil in a system.

However, the rheological behaviors (a Newtonian/non-Newtonian model) of engine oil may relatively be separated from each other after using nanoparticles. Determination of rheological parameters may cause significant errors for designing and neutralizing the excessive performance of an engine system. It should be noted that a linear shear stress is variable by a Newtonian behavior. Many researchers conducted studies on the rheological behavior of nanofluids and could identify the type of nanofluid behavior [26-34]. Since each fluid has a unique rheological behavior and their rheological behaviors should be evaluated separately, table 2 shows some of their studies.

Table 2. The studies of some researchers on the rheological behavior of nanofluids

Researcher	Nanofluid	Range of Shear Rate ( $s^{-1}$ )	Rheological Behavior
M. H. Esfe et al [35]	Zno/EG	20-140	Newtonian
H. Eshgarf et al [36]	MWCNT-SiO <sub>2</sub> /Eg-Water	0-120	Non-Newtonian
Chen et al [37]	TiO <sub>2</sub> /EG	0.1-1000	Newtonian
M. H. Esfe et al [38]	MWCNT/10W40	666-13333	Non-Newtonian
M. H. Esfe et al [39]	ZnO/10W40	666-11997	Non-Newtonian
Jamal-Abad[40]	ZnO/Oil	100-1000	Non-Newtonian

Meanwhile, Hemmat *et al.* [41] identified the non-Newtonian behavior of MWCNT-CuO(10%-90%)/10W40 nanofluid through measuring shear stress. The measurements were made at the temperature range of 5-55°C and the volume fractions of 0-10%. S. Ghasemi and A. Karimipour [42] examined the effect of volume fraction and temperature of CuO nanoparticles on the dynamic viscosity of a liquid paraffin base fluid and realized that the impact of temperature on viscosity is much higher than the volume fraction on the viscosity of nanofluids.

This research identified the non-Newtonian behavior of MWCNT-CuO (30%-70%)/SAE50 nanofluid for better performance in the system. To identify the best performance, the viscosities of different nanofluids were compared and the best nanofluid was introduced. A theoretical correlation was proposed within the range of the tests to understand better the behavior of nanofluid viscosity at varied volume fractions and temperatures.

## 2. Experimentation

## 2.1. Sample Preparation

This research produced the composition of MWCNT and CuO nanoparticles in a two-stage method using Equation 1 at the volume ratio of 30:70 in SAE 50 oil at the volume fractions of 0, 0.0625, 0.125, 0.25, 0.5, 0.75, and 1%. The homogenization results include lack of clusterization, homogeneity, and stability of nanofluid. Table 3 shows the specification of CuO and MWCNT. The size of nanoparticles was examined by an X-ray diffraction (XRD). Fig. 1 shows the XRD patterns of CuO nanoparticles and MWCNTs, and photograph of prepared nanofluid.

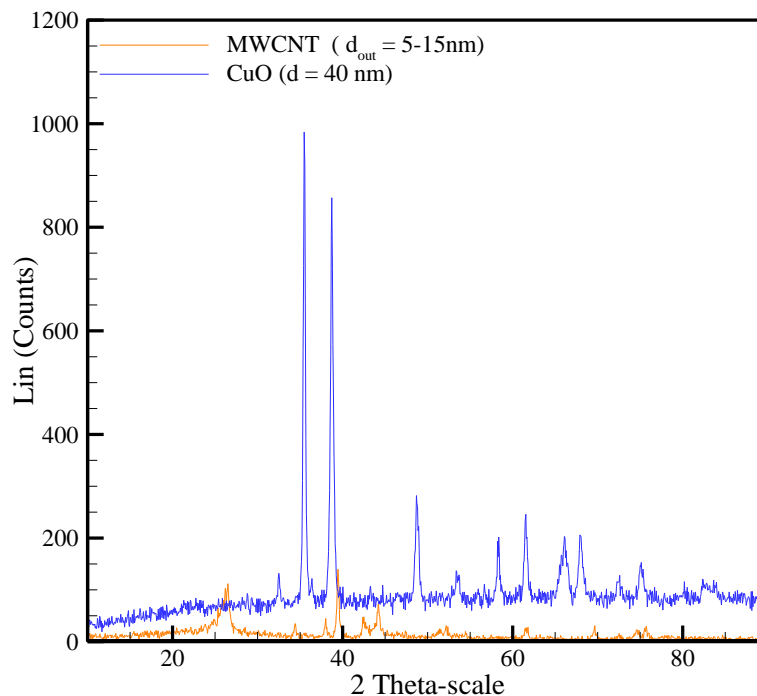
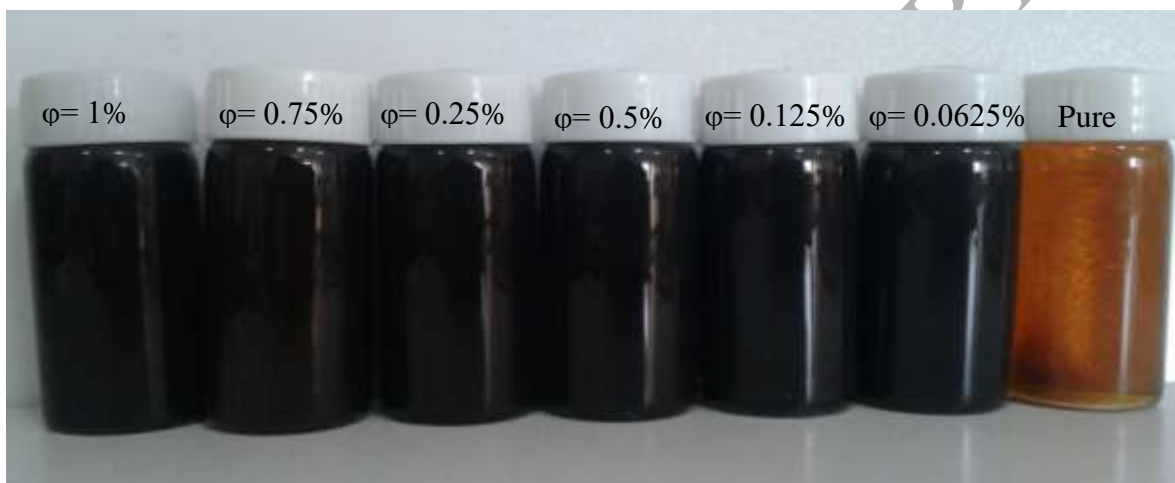


Figure 1. XRD patterns of CuO nanoparticles and MWCNTs, and photograph of prepared

nanofluid

Table 3- Specifications of MWCNT and CuO nanofluids

Parameter	MWCNT	CuO
Purity	95+%	99+%
Outside diameter	5-15 nm (from HRTEM, Raman)	40 nm
Inside diameter	3-5 nm	
SSA	233 m <sup>2</sup> /g (BET)	20 m <sup>2</sup> /g
Color	Black	Black
Ash	1.5wt% (TGA)	-
Electrical conductivity	>100 s/cm	-
True density	~ 2.1 g cm <sup>-3</sup>	6.4 g/cm <sup>3</sup>

$$\varphi(\%) = \frac{\left[ \frac{W_{MWCNT}}{\rho_{MWCNT}} \right] + \left[ \frac{W_{CuO}}{\rho_{CuO}} \right]}{\left[ \frac{W_{MWCNT}}{\rho_{MWCNT}} \right] + \left[ \frac{W_{CuO}}{\rho_{CuO}} \right] + \left[ \frac{W_{SAE50}}{\rho_{SAE50}} \right]} \times 100 \quad (1)$$

In this equation,  $\rho$  is the density,  $\varphi$  is the volume fraction of nanoparticles, and  $w$  is the weight of nanoparticles.

A transmission electron microscope (TEM) was used to identify the morphological properties of nanoparticles.

## 2.2. Viscosity measurement

Fluids have different rheological characteristics that can be described by Viscometer measurements. Viscosity of MWCNT-CUO (30% -70%) / SAE50 was measured in solid volume fractions of 0.0625%, 0.125%, 0.25%, 0.5%, 0.75% and 1% and in temperature range of 25°C -50°C using the CAP 2000+ Brookfield Viscometer. The specification of Brookfield's viscometer is given in Table 4. The CAP 2000+ Viscometer rotates a precisely machined cone spindle over a temperature controlled plate shearing the test sample over a range of speeds from 5 to 1000 RPM. This provides a comprehensive capability to analyze materials for viscosity behavior as a function of both shear rate and temperature. The CAP 2000+ is a

variable speed viscometer. The CAP 2000+ Viscometer is repeatable to  $\pm 0.5\%$  of the full scale viscosity range (FSR).

The viscometer is calibrated with the base fluid at ambient temperature before measurement viscosity of nanofluids. To ensure the results of the tests and repeatability tests, each measurement was performed 3 times and the calculated error was lower than 1%.

Table 4. Viscometer characteristics

Specification	CAP 2000+
Meets Industry Standards	ASTM D4287, ISO 2884 and BS 3900
Range of measurement	0.2-15000 Poise
speed of rotation	5-1000 RPM
shear rate	10-13000 $\text{sec}^{-1}$
Temperature range	5-75 $^{\circ}\text{C}$
shear rate constant	13.33 N

### 3. Results and Discussion

This study discussed experimentally the rheological behavior and temperature and volume fraction impacts on MWCNT-CuO(30%-70%)/SAE50 hybrid nanofluid viscosity. The test used a multi-wall carbon nanotube and copper oxide at the volume ratio of 30:70. The tests were conducted at six temperatures within the range of 25 $^{\circ}\text{C}$  -50  $^{\circ}\text{C}$  at the volume fractions of 0.0625, 0.125, 0.25, 0.5, 0.75, and 1.

#### 3.1. Newtonian Behavior

The behavior of non-Newtonian fluids is studied in two different modes: One mode is according to the fluid response to an applied external pressure and the other is according to the effects caused by a shear stress. The fluids are classified to compressible and incompressible fluids. It denotes whether the volume of an element of the fluid depends on pressure. The flow of gasses and liquids is usually considered as a incompressible fluid and their response to stress is the more important than the pressure response.

A non-Newtonian fluid is a fluid whose flow diagram (shear stress versus shear rate) is non-linear and/or do not flow through the origin of the coordinate axis. In other words, apparent viscosity, shear stress on shear rate, is not fixed at constant temperature and pressure. Such cases can be classified into three major groups:

- 1) They consist of the fluids in which shear rate at any point is only determined by the value of shear stress at the same moment. These fluids are known in different references as time-independent, fully viscose, non-elastic, or NGF fluids.
- 2) They include further complicated fluids in which the relationship between shear stress and shear rate also depends on shear application duration and its history. They are called time-dependent fluids.
- 3) These materials, which have the characteristics of both ideal fluids and elastic solids, are converted into the initial state after deformation. They are categorized as viscoelastic fluids. This categorization is optional and they are often presented in most real cases by combining two or all the three non-Newtonian characteristics. They can be classified into the following three groups:

- 1) Shear-thinning or quasi-plastic
- 2) Viscoplastic
- 3) Shear-thickening or dilatant

Most models have been presented for a quasi-plastic behavior. Some of them have a theoretical basis, others have an experimental basis, and they attempt to perform curve fitting. It is possible to express the non-Newtonian characteristics of the prepared nanofluid through data processing using the Ostwald-de Waele numerical model [43]. The relationship between shear stress and shear rate (drawn as a two-logarithmic curve) in a shear-thinning fluid can be achieved by a direct line within a stress or shear rate range. The following expression can be used:

$$\tau_{yx} = m(\dot{\gamma}_{yx})^n \quad (2)$$

Therefore, the apparent viscosity for the power law (Ostwald de Waele) is as follows:

$$\mu = \frac{\tau_{yx}}{\dot{\gamma}_{yx}} = m(\dot{\gamma}_{yx})^{n-1} \quad (3)$$

For  $n < 1$ , the fluid has pseudoplastic characteristics.

For  $n=1$ , the fluid has a Newtonian behavior.

For  $n>1$ , the fluid has a dilatant (shear-thickening) behavior.

In these equations,  $\tau$  and  $\dot{\gamma}$  indicate the direction perpendicular to the surface under shearing and the direction of force and flow, respectively. “ $m$ ” is consistency index and is a part of viscosity formula for non-Newtonian fluids that is called apparent viscosity. The apparent viscosity is  $m(\dot{\gamma}_{yx})^{n-1}$ . According to fundamental of viscosity the relation between shear stress and shear rate for non-Newtonian and Newtonian fluids are  $\tau_{yx} = m(\dot{\gamma}_{yx})^n$  and  $\tau_{yx} = m\dot{\gamma}_{yx}$  respectively.

$$\tau_{yx} = m(\dot{\gamma}_{yx})^n = m(\dot{\gamma}_{yx})^{n-1} \times (\dot{\gamma}_{yx})^1, \mu = m(\dot{\gamma}_{yx})^{n-1} \quad (4)$$

Parameters  $m$  and  $n$  are experimental coefficients and they are obtained from the curve fitting. They include consistency index and power law index, respectively. For the quasi-plastic fluid, the index varies between 0 and 1. The smaller the  $n$  value is, the higher the quasi-plastic level will be. The value of  $n$  greater than one indicates a dilatant fluid. Figure 2 shows that the nanofluid shows a behavior close to a non-Newtonian time-independent behavior; however, as the behavior is close to a Newtonian behavior, it cannot be mentioned confidently that the nanofluid has a non-Newtonian behavior.

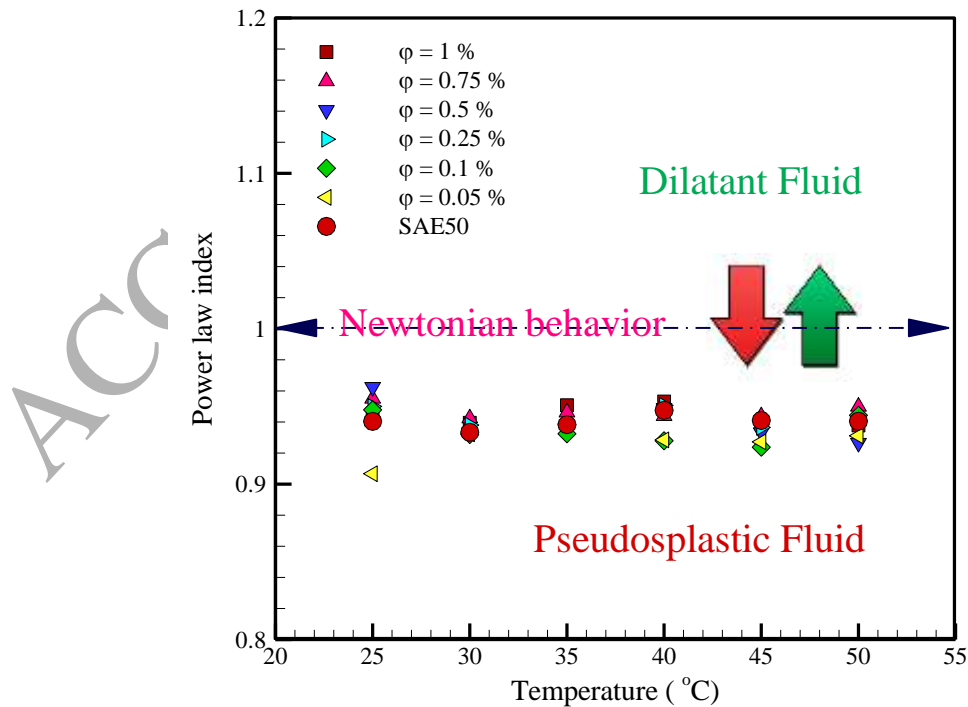
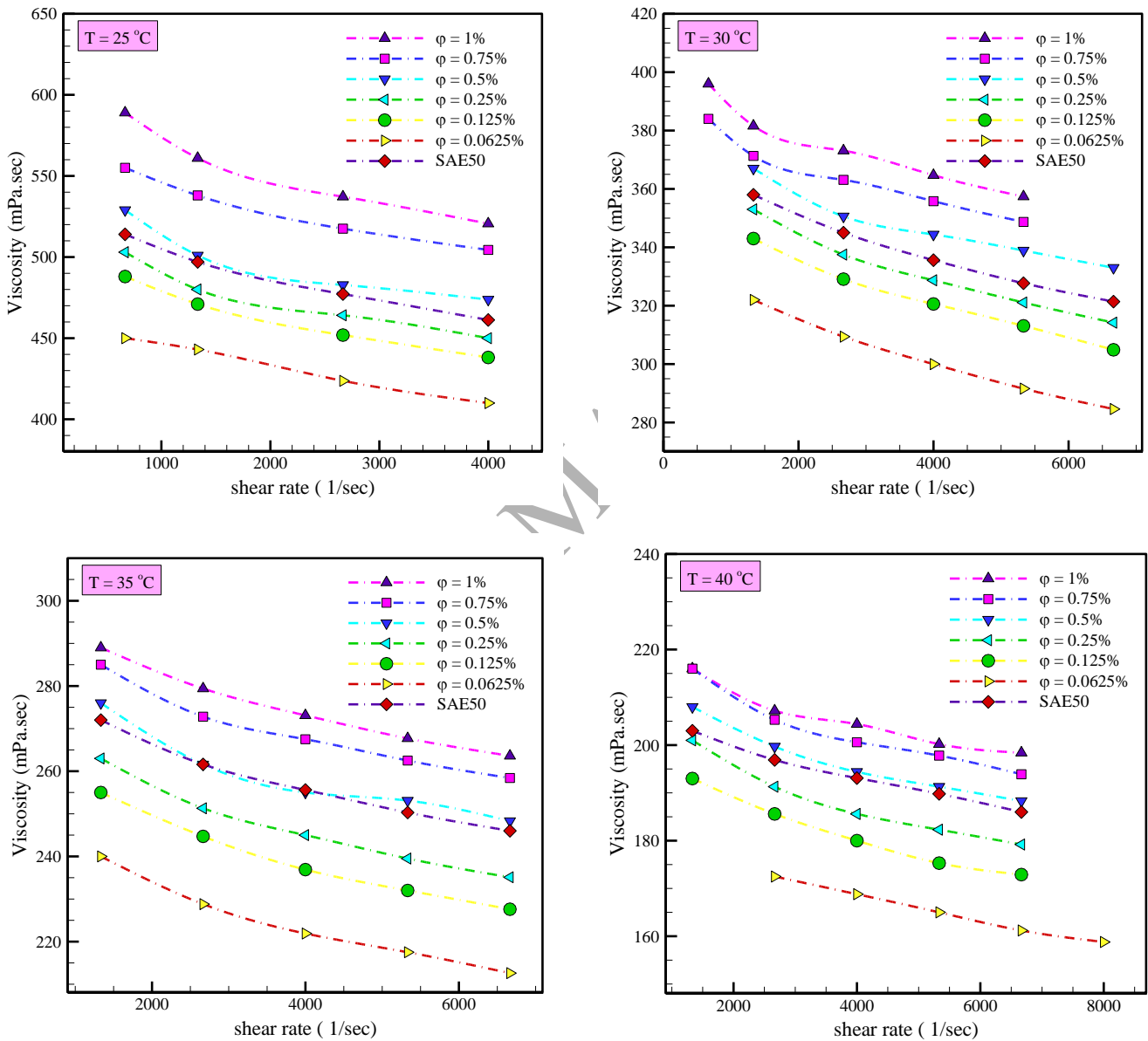


Figure 2. Power law index in terms of solid volume fraction and

temperatures

The most frequent time-independent behavior observed in non-Newtonian behavior is the shear-thinning behavior, which is characterized by a reduction in apparent viscosity with increasing shear rate. As Figure 3 shows, nanofluid viscosity changes with the shear rate increasing and this phenomenon proves the non-Newtonian behavior of the nanofluid.



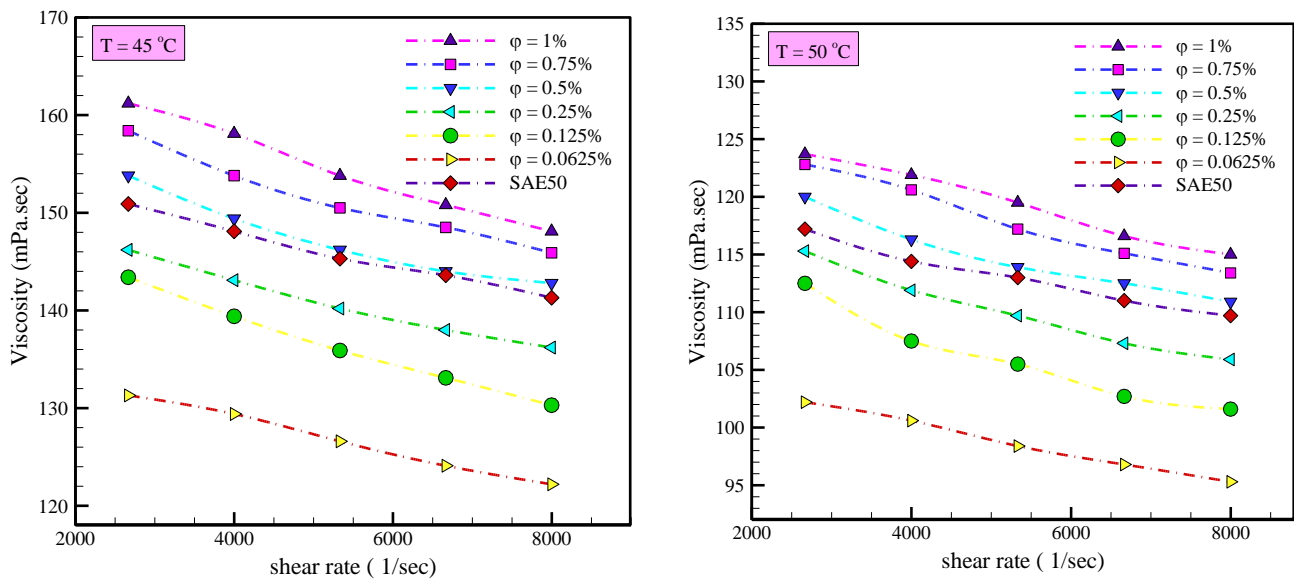


Fig.3. Viscosity versus Shear rate at different temperatures

Figure 4 shows the viscosity of MWCNT-CuO(30%-70%)/SAE50 nanofluid as a function of volume fraction and temperature at the shear rate of 2666(1/s). With the volume fraction of solid nanoparticles increasing, the viscosity of nanofluids increases among nanomolecules due to the van der Waals forces and results in the formation of a cluster combination in non-lubricating concentrations. After cluster formation, the friction among oil layers increases its viscosity as compared with a pure lubricant. At the volume fractions higher than 0.5%, nanofluid viscosity increases gradually with the volume fraction increasing. The effect of the presence of nanoparticles in the base fluid on the nanofluid viscosity is higher at lower temperatures, as viscosity increased by 12% at 25°C. However, the increase reaches 5% at 50°C. Moreover, the results show that nanofluid viscosity reduces with the temperature increasing in a similar process. At the volume fraction of 1% and 0.0625%, nanofluid viscosity reduces by 76% and 75% with the temperature increasing.

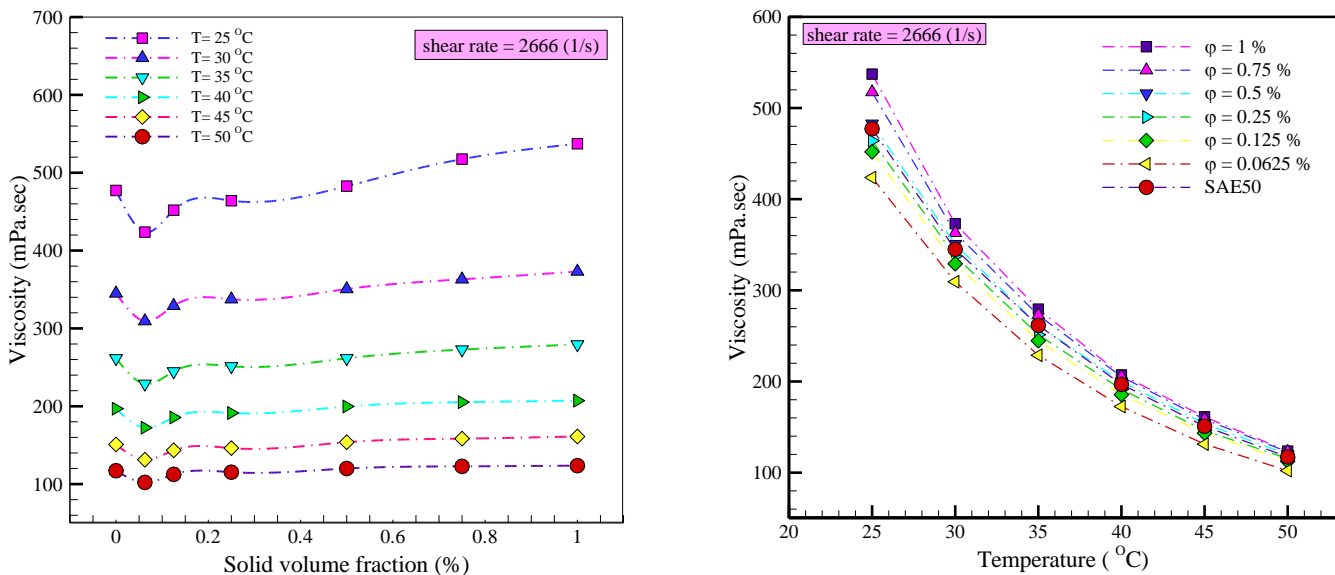


Figure 4. Viscosity values as a function of temperature and solid volume fraction at shear rate of 2666 (1/s)

Figure 5 shows the percentage increase of nanofluid viscosity with respect to the volume fraction at different temperatures. Nanofluid viscosity reduces after adding a small amount of nanoparticles (the volume fractions lower than 0.25%) to the base fluid. It could be because of an unknown phenomena which play a significant role at very low concentrations of nanoparticles which relates to the interactions between some nanoparticles and the host fluid. The solvent, within several molecular diameters of whatever we have added to it can have dynamic and hydrodynamic properties that are quite different from the bulk, so adding nano-objects can lower the viscosity because of hydrodynamic effects. This phenomena is observed in other research results like [44]. The more the content of the nanoparticles is (the volume fractions more than 0.5%), the more the collisions will be and the harder the layers will move. Finally, the increase of collisions raises nanofluid viscosity. With respect to the reduction of nanofluid viscosity (maximum increase of 15%) at low volume fractions, and increase in nanofluid viscosity at high volume fraction (maximum increase of 12%), the nanofluid can have industrial use for heat transfer. As the range of nanofluid viscosity variation is low, the nanofluid thermal properties can be investigated.

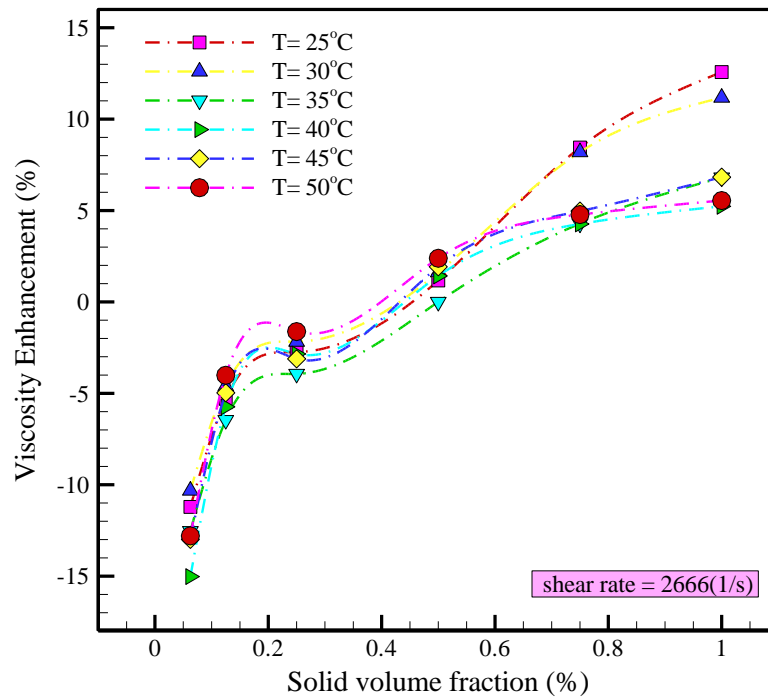


Figure 5. Viscosity enhancement versus solid volume fraction at different temperatures

### 3.2. Comparison with Other Nanofluids

Addition of nanoparticles to the base fluid changes nanofluid thermophysical properties. One of the properties includes nanofluid viscosity changes. Presence of different nanoparticles at varied volume ratios leaves various effects on nanofluid viscosity. To find the best performance of the nanofluid, the results of viscosity test for MWCNT-Cu(30%-70%)/SAE50 nanofluid were compared with the results obtained by Hemmat *et al.* [45-50] using various nanofluids. The comparison was made at 25°C and the shear rate of 2666 (1/s) at different volume fractions. Figure 6 shows that the study nanofluid and MWCNT-ZnO(10%-90%)/SAE40, as compared with other nanofluids, expose to lower viscosity increase at all volume fractions. Therefore, nanofluid MWCNT-ZnO(10%-90%)/SAE40 uses other than lubrication can be considered for them.

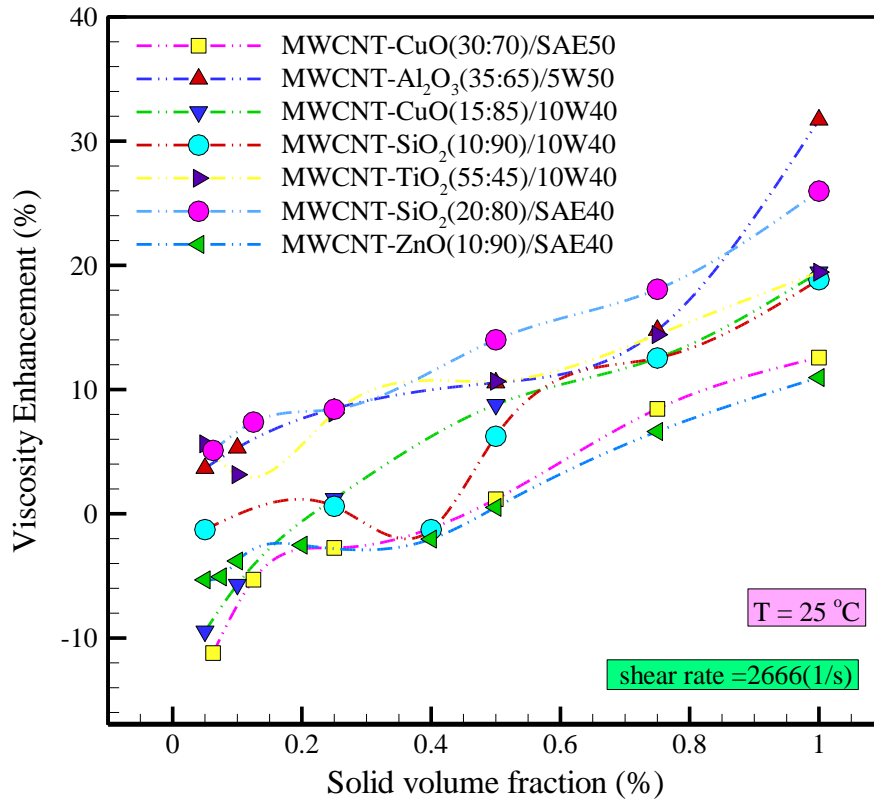


Figure 6. Comparison between viscosity enhancements

### 3.3. New Proposed correlation

The theoretical equations are unable to predict the viscosity of hybrid nanofluids at any volume fraction or temperature. Therefore, a new quadratic correlation using curve-fitting technique on the experimental data was extracted. In this case, repetition of many tests is avoided and it is possible to predict many results with an acceptable accuracy. A mathematical correlation is needed in this study to predict relative viscosity with volume fraction, temperature, and shear rate variables. To achieve this, the curve fitting was carried out on the experimental data.  $R^2$  shows an appropriate compatibility between the results of the proposed correlation and the experimental results. For the predicted correlation of the nanofluid,  $R^2$  equals 0.9453. Equation 5 represents the proposed correlation and table 4 show the analysis of proposed correlations' parameters

$$\frac{\mu_{nf}}{\mu_{bf}} = 0.99 + 0.49\varphi - 0.0077T + 6.81 \times 10^{-6}\dot{\gamma} - 0.00249\varphi T - 2.19 \times 10^{-7}T\dot{\gamma} - 0.19\varphi^2 + 0.00011T^2 \quad (5)$$

where  $\mu$  is the viscosity,  $\phi$  denotes to the volume fraction,  $T$  is the temperature, and  $\dot{\gamma}$  denotes to the shear rate,  $nf$  subscript is related to the nanofluid, and  $bf$  subscript is related to the base fluid.

The accuracy and reliability of regression models were examined by ANOVA. Table 5 shows the importance of each parameter of Equation 4. The importance of the regression model was shown with the high  $F$  value (409.8665). Values  $Prob>F$  show less than 0.05 of the statistical importance of the model parameters. It shows the effects of low-impact parameters of the model with  $Prob>F$  values greater than 0.05. All the insignificant parameters were deleted from the model.

Table 5. Analysis of proposed correlations' parameters

Source	Sum of Squares	df	Mean Square	F Value	p-value Prob > F	
Model	0.847879	7	0.121126	409.8665	< 0.0001	significant
A-phi	0.717597	1	0.717597	2428.217	< 0.0001	
B-T	0.014984	1	0.014984	50.70164	< 0.0001	
C-shear rate	0.001169	1	0.001169	3.955746	0.0484	
AB	0.008494	1	0.008494	28.7407	< 0.0001	
BC	0.001754	1	0.001754	5.936149	0.0159	
A <sup>2</sup>	0.04734	1	0.04734	160.189	< 0.0001	
B <sup>2</sup>	0.006508	1	0.006508	22.02194	< 0.0001	
Residual	0.049057	166	0.000296			
Cor Total	0.896936	173				

The percentage of the deviation of data from the experimental values was calculated by Eq. (6) to compare the experimental data with the results obtained from the proposed correlation for viscosity estimation.

$$MOD = 1 - \frac{\mu_{rel} |_{Pred}}{\mu_{rel} |_{Exp}} \quad (6)$$

Figure 7 reports the deviation margin in terms of volume fraction at various shear rates and temperatures. If the data are close to the zero line, it is possible to achieve the high accuracy of the proposed relation. The studies show that the data estimation is appropriate for the nanofluid because most data were within the range of -6% to +7%

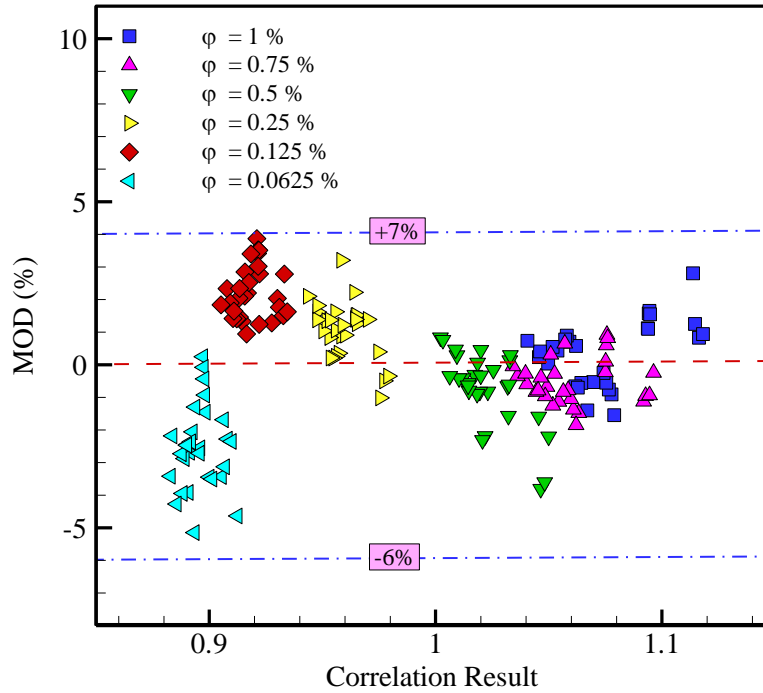


Figure 7. Deviation percentage of the values obtained from the proposed relation using the experimental data

Figure 8 compares the results obtained from the proposed model and the experimental results for different volume fractions. In case of a low deviation from the bisector, it is possible to use “proper” and “correct” expressions for the test results. Figure 10 shows an appropriate agreement between the experimental data and the data obtained from the correlation. They are also balanced relative to the bisector.

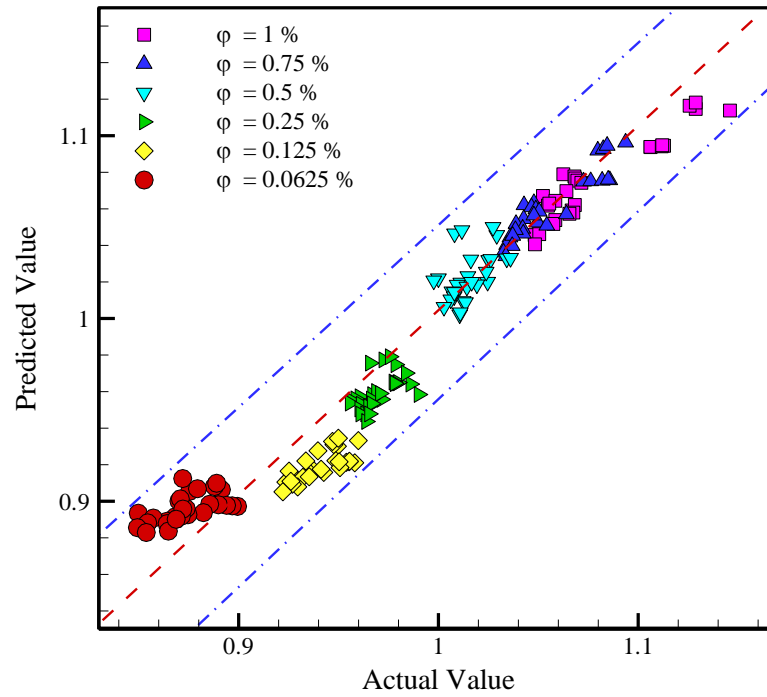


Figure 8. The experimental results of the proportion of dynamic viscosity and the results obtained from the mathematical relation extracted at different volume fractions

Figure 9 shows the consistency of the experimental data and the experimental results and the values of residuals. The difference between the experimental data and the values of the modeling (+0.04,-0.05) indicates the high accuracy of the proposed correlation.

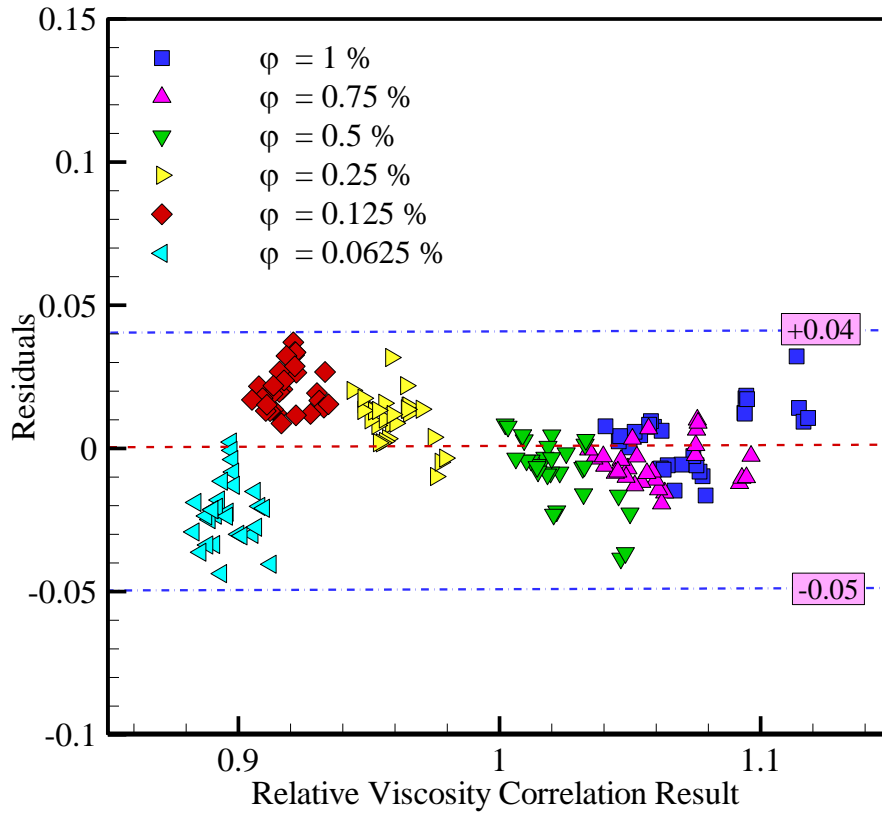


Fig. 9 . Residuals versus correlation results

### 3.4. Sensitivity Analysis

Figure 10 reports the changes of sensitivity of MWCNT-CuO(30%-70%)/SAE50 nanofluid viscosity with respect to the volume fraction and temperature at the shear rate of  $2666 \frac{1}{s}$ . The sensitivity percentage of each parameter is achieved separately by the changes of  $\pm 5\%$ ,  $\pm 10\%$ ,  $\pm 15\%$ ,  $\pm 20\%$  in terms of the temperature of  $30^\circ C$  and the volume fraction of  $0.75\%$ . The relationship between temperature sensitivity and volume fraction analysis and the viscosity of nanofluids were proposed by Eqs 7 and 8:

$$\text{Sensitivity \%} = \left[ \frac{\frac{\mu_{nf}}{\mu_{bf}}(\varphi=1\% \pm 5\%, \pm 10\% \pm 15\%, \pm 20\%; T=30^\circ C)}{\frac{\mu_{nf}}{\mu_{bf}}(\varphi=0.75\%, T=30^\circ C)} - 1 \right] \times 100 \quad (7)$$

$$\text{Sensitivity \%} = \left[ \frac{\frac{\mu_{nf}}{\mu_{bf}}(\varphi=0.75\%; T=30^\circ C \pm 5\%, \pm 10\% \pm 15\%, \pm 20\%)}{\frac{\mu_{nf}}{\mu_{bf}}(\varphi=0.75\%, T=30^\circ C)} - 1 \right] \times 100 \quad (8)$$

Figure 10 shows that at the temperature of 30°C and the volume fraction of 0.75%, the effect of temperature on the viscosity of nanofluids exceeds the impact of volume fraction on the viscosity of nanofluids.

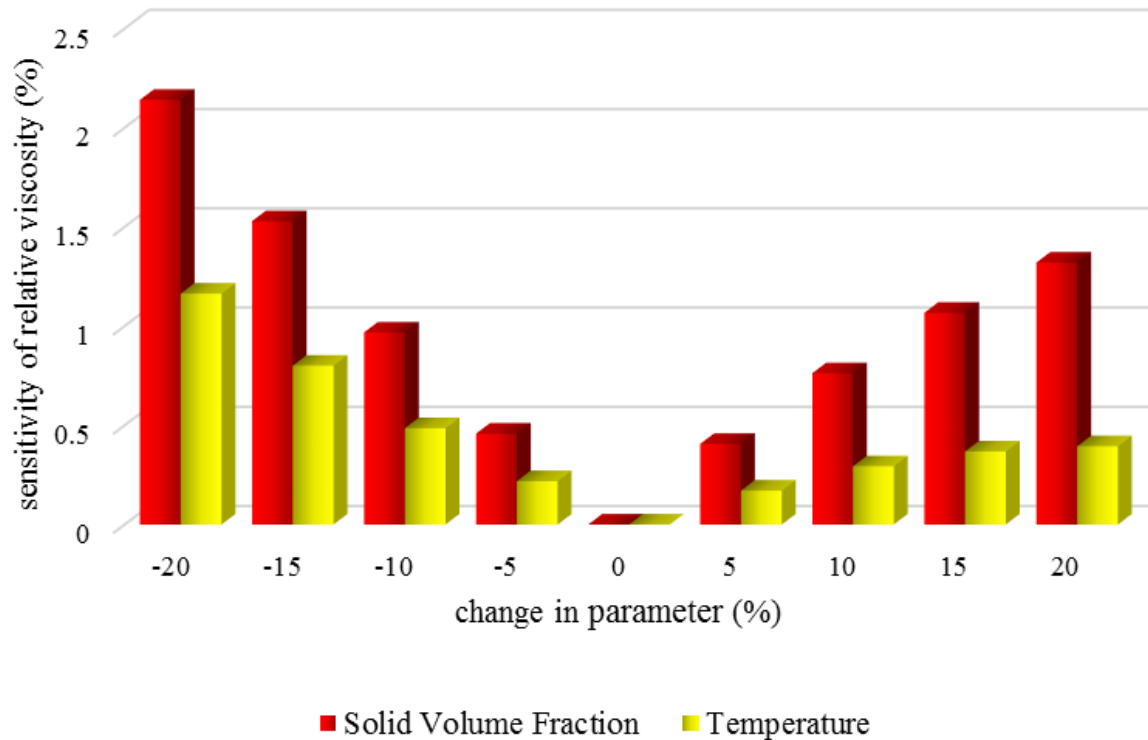


Figure 10. Viscosity sensitivity analysis at different temperatures and shear rates in terms of volume fractions

#### 4. Conclusion

This study discussed the viscosity of MWCNT-CuO(30%-70%)/SAE50 nanofluid at the volume fractions of 0%, 0.0625%, 0.125%, 0.25%, 0.5%, 0.75% and 1% at temperature of 25, 30, 35, 40, 45, and 50°C.

The following results were obtained:

- The nanofluid rheological behavior showed that the nanofluid exhibits a behavior close to a non-Newtonian quasi-plastic behavior.
- The viscosity reduction in 0.25%, 0.1% and 0.0625% solid volume fractions was found to be lower than base oils. This phenomenon reduces the power required for pumping, which is very important for application in various industries.

- Sensitivity analysis results showed that viscosity is more sensitive to variations of solid volume fraction compared to temperature variations.
- A new three-variable mathematical correlation was presented in terms of temperature, solid volume fraction and shear rate to predict the viscosity of nano-lubricant under different operating conditions. In addition, the significance of each of the variables of the presented correlation was investigated using the RSM method.
- It was found that, in high volume fractions (e.g. 1%), viscosity increased by 10%, which is small compared to the reported amounts for increase in the viscosity of other nanofluids. This result provides a substrate for improving the thermal properties of the oils, because in addition to improving the thermal properties of the oil, the power required for pumping also does not increase significantly due to the lack of increased viscosity in large quantities.

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