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Applying Artificial Neural Network and Response Surface Method to Forecast the Rheological Behavior of Hybrid Nano-Antifreeze Containing Graphene Oxide and Copper Oxide Nanomaterials

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Abstract: In this study, the efficacy of loading graphene oxide and copper oxide nanoparticles into ethylene glycol-water on viscosity was assessed by applying two numerical techniques. The first technique employed the response surface methodology based on the design of experiments, while in the second technique, artificial intelligence algorithms were implemented to estimate the GO-CuO/water-EG hybrid nanofluid viscosity. The nanofluid sample's behavior at 0.1, 0.2, and 0.4 vol.% is in agreement with the Newtonian behavior of the base fluid, but loading more nanoparticles conforms with the behavior of the fluid with non-Newtonian classification. Considering the possibility of non-Newtonian behavior of nanofluid temperature, shear rate and volume fraction were effective on the target variable and were defined in the implementation of both techniques. Considering two constraints (i.e., the maximum R-square value and the minimum mean square error), the best neural network and suitable polynomial were selected. Finally, a comparison was made between the two techniques to evaluate their potential in viscosity estimation. Statistical considerations proved that the R-squared for ANN and RSM techniques could reach 0.995 and 0.944, respectively, which is an indication of the superiority of the ANN technique to the RSM one.

Keywords: hybrid nanofluid; viscosity; artificial neural network; response surface method

1. Introduction

Heat transfer, given its various applications, has long been the focus of researchers and engineers [1,2]. Heat transfer can be attributed to the movement of free molecules, lattice vibration, molecular diffusion, as well as molecular collisions. Thermal conductivity is the potential of matter in heat transfer, so that materials with high thermal conductivity have a greater heat transfer rate [3,4]. Many studies have shown that the presence of nanoscale solids with high thermal conductivity can amplify the fluid thermal conductivity [5,6] and, consequently, intensify the heat transfer rate [7–13]. Considering the fluid motion in the convection heat transfer, in addition to the thermal conductivity, the viscosity can

also be effective in determining the convective heat transfer coefficient [14,15]. In the convection heat transfer scope, the fluid velocity field is required. To know the fluid velocity field, momentum conservation (Navier–Stokes equation) must be solved [16]. In the Navier–Stokes equation, there is friction force, which is itself a function of viscosity and velocity gradient. Therefore, knowledge of the viscosity changes is essential for solving convection heat transfer problems. Adding nanoparticles can have two significant effects on the viscosity; that is, an increase in viscosity and a change from Newtonian behavior to non-Newtonian behaviour [17,18]. From one perspective, studies on graphene-containing nanofluid are divided into viscosity, thermal conductivity, and other properties [19]. The number of studies on thermal conductivity is greater than studies on viscosity [20]. Table 1 summarizes the studies using graphene nanoparticles.

Table 1. Summary of studies using graphene.

References	Nanoparticles	Concentration \ Temperature	Findings
Hu et al. [21]	<i>GN/EG – DW</i> 60 : 40 vol.%		Investigation of pool boiling heat transfer (PBT) Up to 0.02 wt.%, PBT enhanced
Yu et al. [22]	<i>GON/EG</i>	0–5 vol.% 10–60 °C	$\frac{k_{nf}}{k_{bf}} = 1.61$
Yu et al. [23]	<i>GON – DW</i> <i>GON – propyl glycol</i> <i>GON – liquid paraffin</i>	0–5 vol.%	Owing to incorporation GON nanosheets into DW, propyl glycol, and liquid paraffin, k intensified by 30.2, 62.3, and 76.8%
Yu et al. [24]	<i>GON/EG</i>	5.0 vol.%	Enhancement in $\frac{k_{nf}}{k_{bf}}$ is up to 61.0%. It was found that the maximum enhancement was independent of temperature
Yu et al. [25]	<i>GN/EG</i>	5 vol.%	$\frac{k_{nf}}{k_{bf}} = 1.86$
Moghaddam et al. [26]	<i>Graphene-glycerol</i>	0.25–2 wt.% 20–60 °C	μ_{nf} intensified up to 401.49% at 2 wt.% and 20 °C.
Ahammed et al. [27]	<i>Graphene-w</i>	0.05, 0.1, and 0.15 vol.% 10–50 °C	37.2% enhancement in thermal conductivity at 0.15 vol.%
Baby et al. [28]	<i>Graphene/EG – DI</i>	0.005–0.05 vol.% 25–50 °C	$\frac{k_{nf}}{k_{bf}} = 1.75$
Aravind and Ramaprabhua [29]	<i>GN/EG</i> <i>GN/DI water</i>	0.14 vol.% 25 °C	$\frac{k_{nf}}{k_{EG}} = 1.065$ $\frac{k_{nf}}{k_{DI\ water}} = 1.136$
Kole and day [30]	<i>GNP/EG – W</i>	0.041–0.395 vol.% 10–70 °C	viscosity increased by 100%

Given the relatively high thermal conductivity ($76.5 \frac{W}{m.K}$) of copper oxide (CuO), this nanoparticle has also been used in many studies. Table 2 summarizes the studies using copper oxide nanoparticles.

Hybrid nanofluids benefit from the advantages of two different nanoparticles, so many researchers have focused on this class of nanofluids [39–44]. The correlations used to estimate thermal conductivity or viscosity (for base fluids) are not accurate for nanofluids [45–48]. Conducting many experimental measurements on thermal conductivity or viscosity is costly as well as time-consuming [49,50]. Therefore, using predictive techniques can be helpful. In this way, algorithms based on artificial intelligence (ANN) and response surface methodology (RSM) have been widely used by researchers [51–53]. In Table 3, several studies including ANN/RSM approaches are summarized.

Table 2. Summary of studies using CuO.

References	Nanoparticles	Concentration\Temperature	References
Águila et al. [31]	CuO/Octadecane	30–55 °C 2.5, 5, and 10 wt.%	$\frac{\mu_{\text{CuO/Octadecane}}}{\mu_{\text{Octadecane}}} = 1.6$ non-Newtonian
Sahoo and Kumar [32]	Al ₂ O ₃ -CuO-TiO ₂ /water	0.01–0.1% 35 °C to 50 °C	$\frac{\mu_{\text{Al}_2\text{O}_3\text{-CuO-TiO}_2/\text{w}}}{\mu_{\text{Al}_2\text{O}_3\text{-CuO/w}}} = 1.1725$ $\frac{\mu_{\text{Al}_2\text{O}_3\text{-CuO-TiO}_2/\text{w}}}{\mu_{\text{Al}_2\text{O}_3\text{-TiO}_2/\text{w}}} = 1.5541$
Esfe et al. [33]	CuO-MWCNT/10W40	5–55 °C Up to 10 vol.%	$\frac{\mu_{\text{MWCNT-CuO/10W40}}}{\mu_{\text{10W40}}} = 1.22$ Non-Newtonian
Akilu et al. [34]	TiO ₂ -CuO-C/EG	Up to 2 vol.% 30–60 °C	$\frac{\mu_{\text{TiO}_2\text{-CuO-C/EG}}}{\mu_{\text{EG}}} = 1.80$ Newtonian
Asadi et al. [35]	CuO-TiO ₂ /water	0.1 to 1 vol.% 25–55 °C	Newtonian behavior
Shah et al. [36]	CuO/EG-water (70;30)	0.3 vol.%	Newtonian
Priya et al. [37]	CuO-water	Up to 0.016 vol.% 28–55 °C	$\frac{\mu_{\text{CuO/water}}}{\mu_{\text{water}}} \sim 1.10$
Alawi et al. [38]	CuO/R134a	1 to 5 vol.% 27–47 °C	$\frac{\mu_{\text{CuO/R134}}}{\mu_{\text{R134}}} \sim 3.47$

Table 3. Summary of studies including ANN/RSM approaches.

Reference	Nanofluid	Input Data/Output Variable	Temperature/Concentration	Accuracy	Methods
Zhao et al. [54]	Al ₂ O ₃ -water CuO-water	T, φ, ρ_{np} d_{np} and μ_{bf} output: μ_{nf}	10–50 °C up to 12 vol.%	$(R^2)_{\text{Al}_2\text{O}_3/\text{water}} = 0.9966$ $(R^2)_{\text{CuO/water}} = 0.9998$	ANN
Shahsavari and Bahraei [55]	Fe ₃ O ₄ - CNTs/water	$T, \varphi_{\text{Fe}_3\text{O}_4}$ $\dot{\gamma}$ and φ_{CNTs} output: μ_{nf}	25 and 55 °C $0.1 < \varphi_{\text{Fe}_3\text{O}_4} < 0.9$ $0 < \varphi_{\text{CNTs}} < 1.35$	$R^2 = 0.999$	ANN
Ghaffarkhah et al. [56]	MWCNT- Al ₂ O ₃ /SAE40 MWCNT- MgO/SAE40 MWCNT- ZnO/SAE40 MWCNT- SiO ₂ /SAE40	T, φ μ_{nf}	25 and 50 ° 0.05, 0.25, 0.50, 0.75, and 1 vol.%	Not reported	ANN
Alirezaie et al. [57]	MWCNT- MgO/Engine oil	$T, \varphi, \dot{\gamma}/\mu_{nf}$	0.0625, 0.125, 0.25, 0.5, 0.75, and 1 vol.% 25–50 °C	$R^2 = 0.9973$	ANN
Yan et al. [58]	Graphene nanosheets/ethylene glycol	T, φ σ_{nf}	25–70 °C 0–5 wt.%	$R^2 = 0.981$	RSM
Tian et al. [59]	Al ₂ O ₃ - MWCNT/10W40	T, φ k_{nf}	25–65 °C Up to 1 vol.%	$R^2 = 0.9948$	RSM
Yan et al. [40]	MWCNTs- TiO ₂ /EG	T, φ μ_{nf}	25–55 °C up to 1 vol.%	$R^2 = 0.995$	RSM
Tian et al. [60]	CuO/MWCNTs into water/EG	T, φ μ_{nf}	20–60 °C Up to 1 vol.%	$R^2 = 0.9987$	RSM

Table 3 reveals that the methods of estimating GO-CuO/water-EG viscosity have not been investigated. In this study, two different techniques are utilized. The first technique employed the response surface methodology based on the design of experiments. In the second technique, artificial intelligence algorithms are implemented to estimate the GO-CuO/water-EG hybrid nanofluid viscosity. Considering the possibility of non-Newtonian behavior of nanofluid, temperature, shear rate, and volume fraction were effective on the target variable and were defined in the implementation of both techniques. Considering two constraints (i.e, the maximum R-square value and the minimum root mean square error), the best neural network and suitable polynomial were selected. Finally, a comparison was made between the two techniques to evaluate their potential in viscosity estimation.

2. Input Data

In this paper, the GO-CuO/water-EG hybrid nanofluid viscosity is predicted. The nanofluid is prepared from GO as well as CuO and water-EG. μ_{nf} was measured at 25–50 °C (with 5 °C interval) and 0.1, 0.2, 0.4, 0.8, and 1.6 vol%. [61] (Figure 1). In the paper conducted by Rostami et al. [61], the details of morphology, XRD and TEM/SEM images, preparation, and stability were explained. By carefully studying the viscosity changes in terms of shear rates, it is found that the behavior of the hybrid nanofluids is strongly dependent on the amount of added nanoparticles. For $\varphi \leq 0.4$ vol.%, GO-CuO/water-EG hybrid nanofluids are treated like a fluid with Newtonian behavior, while with a further increase in φ , the nanofluids exhibit non-Newtonian behavior [61]. Note that, for a fluid with Newtonian behavior, viscosity is influenced by T as well as φ , while for non-Newtonian behavior, shear rate should be added to the effective parameters.

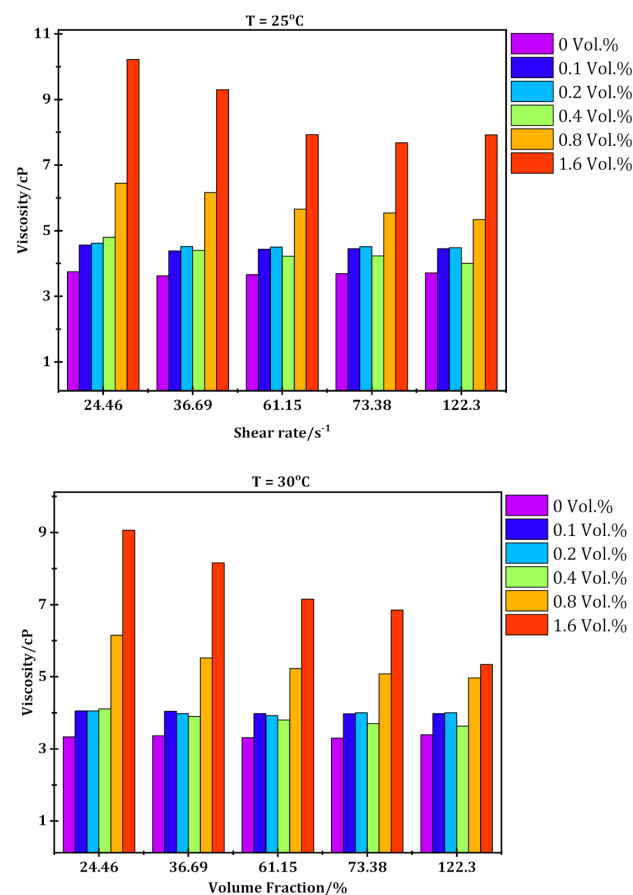


Figure 1. Cont.

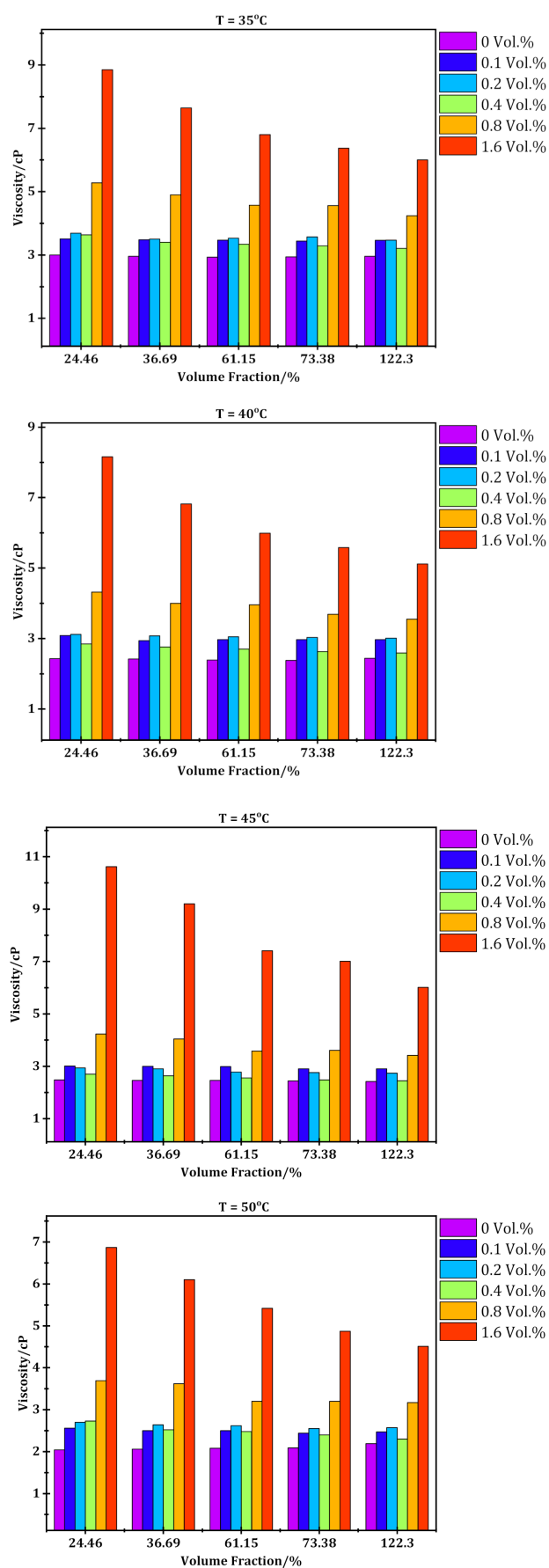


Figure 1. The experimental viscosity.

3. Artificial Neural Network

Artificial neural networks are inspired by the human brain structure and utilized in viscosity prediction by many researchers [62–65]. ANNs consist of three layers: input, hidden, and output layers [66,67]. Each layer contains a group of neurons that are generally associated with all other layer neurons. Note that the neurons of each layer do not interact with other neurons of the same layer. The neuron is the smallest unit performing information processing. It is considered as a basis for ANN. The neuron is considered as a function with nonlinear behavior, so ANNs formed by the assembly of nonlinear functions are completely complex. The number of neurons in the first layer (i.e., input layer) is determined by the number of input variables. Input variables in this study are temperature, viscosity, and shear rate. Hence, the number of input neurons is three (Figure 2). In neural networks, each input (X_i) has a weight that is represented by (W_i). As can be seen, each input is connected to a single weight. Each input must be multiplied by its weight. In the neural network, the result of ($X_i W_i$) is summed by the sigma operator ($\sum X_i W_i$). The sigma operator output enters the activation function.

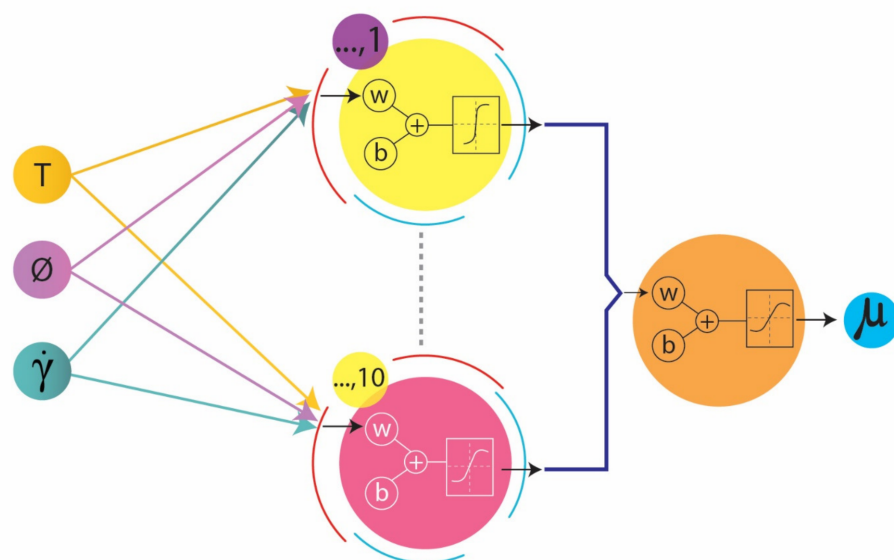


Figure 2. ANN structure.

These functions are a part of neural networks whose input is a number (small or large in arbitrary range), and their output is usually a number between 0 and 1, or -1 and $+1$. In fact, these functions convert an input number to a specified interval (e.g., -1 to $+1$). Transfer functions are also called activation functions. The non-linear sigmoid activation function is used in this simulation. In general, the neural network can be considered as a transformation function from a space with n dimension (in the input) to a space with m dimension (in the output). In this study, the neural network converts a three-dimensional input space into a one-dimensional space output. Input space consists of temperature, mass fraction, and shear rate (3D), and output space is only viscosity (1D).

4. RSM

Mathematical methods have many applications in many fields [68–72]. One of the methods for predicting the objective function (e.g., viscosity and thermal conductivity) is the use of response surface methodology [73–77]. In this technique, a polynomial is derived using mathematical and statistical methods to estimate the objective function. In these polynomials, there are either independent variables with the power of 1, 2, and 3 (e.g., T^2) or the combination of multiple independent variables (e.g., $T\dot{\gamma}$). The significance of each independent parameter can be estimated by applying ANOVA. Then, by applying

the concept of regression and considering the values of R-square, the best polynomial correlation is proposed.

5. Results and Discussion

Temperature (T), shear rate ($\dot{\gamma}$), and mass fraction (φ) are identified as input variables and viscosity is the output (objective or target) variable. Using the RSM technique, the following polynomials are suggested for visibility estimation.

$$\begin{aligned} \mu_{nf} = & a_0 + a_1 T + a_2 \varphi + a_3 \dot{\gamma} + a_4 T\varphi + a_5 T\dot{\gamma} + a_6 \varphi\dot{\gamma} + a_7 T^2 + a_8 \varphi^2 + a_9 \dot{\gamma}^2 \\ & + a_{10} T\varphi\dot{\gamma} + a_{11} T^2\varphi + a_{12} T\dot{\gamma}^2 + a_{13} T\varphi^2 + a_{14} T\dot{\gamma}^2 \\ & + a_{15} \varphi^2\dot{\gamma} + a_{16} \dot{\gamma}^2\varphi + a_{17} T^3 + a_{18} \varphi^3 + a_{19} \dot{\gamma}^3 \end{aligned} \tag{1}$$

Table 4 reports the results of ANOVA. If the p -value for any parameter is less than 0.05, then it can be said that any variations in that parameter have a substantial result on the target variable, while values higher than 0.1 show that the variation in the parameters has no considerable effect [78]. According to Table 4, the significant parameters are shown in bold.

Table 4. ANOVA results.

p -Value	F-Value	Parameter	p -Value	F-Value	Parameter
0.98	0.00058	$T\dot{\gamma}$	< 0.0001	32.95	T
0.153	2.0577	$T\varphi$	< 0.0001	55.026	φ
< 0.0001	123.39	$\dot{\gamma}\varphi$	0.2777	1.1862	$\dot{\gamma}$
0.711	0.1374	$T\dot{\gamma}\varphi$	0.0075	7.313	T^2
0.618	0.25	$T^2\dot{\gamma}$	< 0.0001	22.77	$\dot{\gamma}^2$
0.863	0.0296	$T^2\varphi$	< 0.0001	46.368	φ^2
0.741	0.109	$T\dot{\gamma}^2$	0.055	3.737	T^3
0.0166	5.855	$T\varphi^2$	0.395	0.726	$\dot{\gamma}^3$
< 0.0001	20.611	$\dot{\gamma}^2\varphi$	0.778	0.0794	φ^3
0.0224	5.312	$\dot{\gamma}\varphi^2$			

Table 5 presents the values of $a_0 - a_{19}$. Two criteria of mean square error (MSE) [79,80] and R-square [74,81] can be utilized to assess the proposed correlation performance (Equation (1)):

$$MSE = \frac{1}{N} \sum_{i=1}^N (\mu_{Pred} - \mu_{Exp})^2 \tag{2}$$

$$-square = \left(\frac{\sum_{i=1}^N (\mu_{Exp} - \bar{\mu}_{exp})(\mu_{Pred} - \bar{\mu}_{Pred})}{\sqrt{\sum_{i=1}^N (\mu_{Exp} - \bar{\mu}_{exp})^2} \sqrt{\sum_{i=1}^N (\mu_{Pred} - \bar{\mu}_{Pred})^2}} \right)^2 \tag{3}$$

$$correlationcoefficient(R) = \left(\frac{\sum_{i=1}^N (\mu_{Exp} - \bar{\mu}_{exp})(\mu_{Pred} - \bar{\mu}_{Pred})}{\sqrt{\sum_{i=1}^N (\mu_{Exp} - \bar{\mu}_{exp})^2} \sqrt{\sum_{i=1}^N (\mu_{Pred} - \bar{\mu}_{Pred})^2}} \right) \tag{4}$$

Table 5. Coefficients’ value of the proposed correlation.

Parameter	Value	Parameter	Value
a ₀	14.914	a ₁₀	−7.438 E−5
a ₁	−0.78439	a ₁₁	−1.620 E−4
a ₂	4.26804	a ₁₂	7.576 E−6
a ₃	−0.0209	a ₁₃	0.0383
a ₄	−0.054	a ₁₄	−1.2028 E−6
a ₅	−3.288 E−4	a ₁₅	−9.1588 E−3
a ₆	−0.039446	a ₁₆	2.57 E−4
a ₇	0.018264	a ₁₇	−1.517 E−4
a ₈	0.55451	a ₁₈	−0.13075
a ₉	4.33736 E−4	a ₁₉	−1.908 E−6

Applying Equations (2) and (3), the values of MSE and R-square are 0.166 and 0.944, respectively. The R-square value of 0.944 affirms that the accuracy of the proposed correlation (Equation (1)) is very high and, therefore, the viscosity can be predicted with high accuracy. Another criterion of the suitability of the proposed correlation (Equation (1)) can be obtained by calculating the margin of deviation (MOD) [58,79].

$$MOD = \frac{\mu_{Exp} - \mu_{Pred}}{\mu_{Exp}} \times 100 \tag{5}$$

By focusing on Figure 3, it is found that the MOD varies in the range of 0 to 24.48%. Thereby, the maximum margin of deviation is 24.48%.

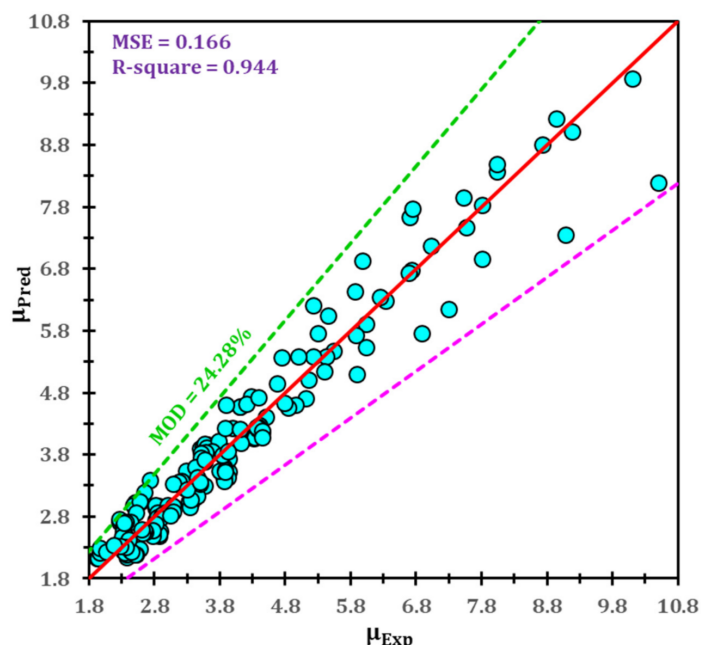


Figure 3. Margin of deviation of proposed correlation (Equation (1)).

However, many researchers also require the maximum amount of residual. The maximum value of the residual can be seen in Figure 4.

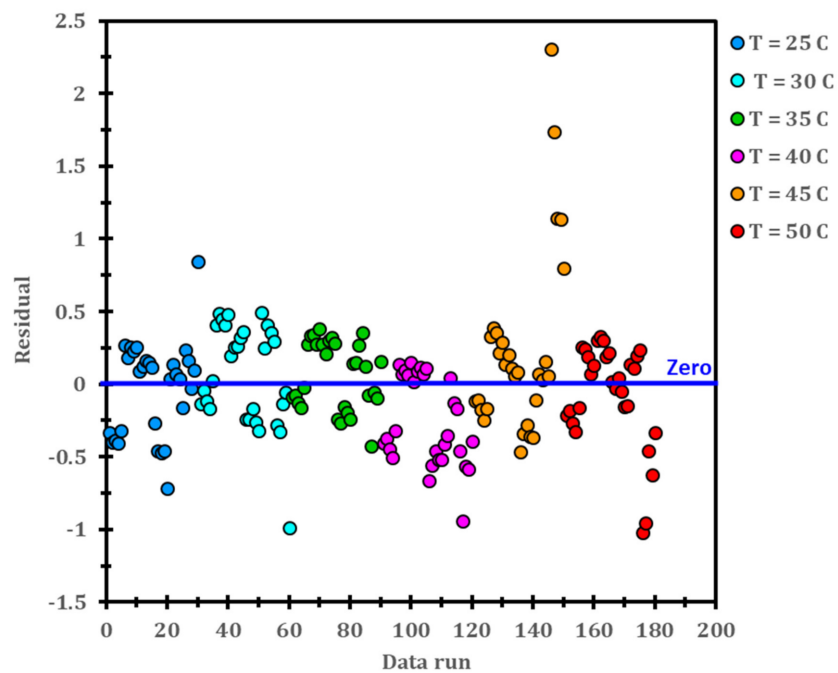


Figure 4. Residual value for the proposed correlation.

The most important criterion for evaluating the accuracy of a correlation is illustrating the predicted value against the laboratory value. In Figure 5, all experimental viscosity (at 180 points) are compared with the predicted viscosity value. At low temperatures, accuracy seems to be poor. Therefore, a comparison was made for each temperature separately. Figure 6 shows the compared μ_{Exp} and μ_{Pred} at each temperature.

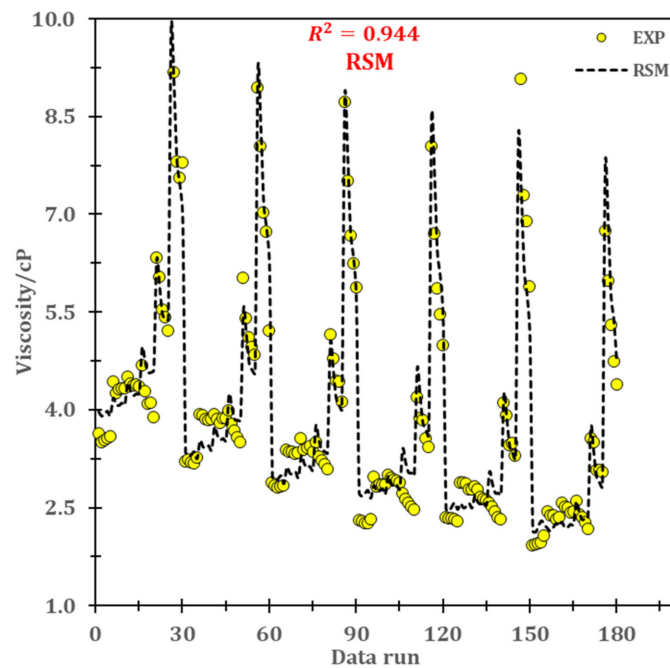


Figure 5. Comparison between μ_{Exp} and μ_{RSM} .

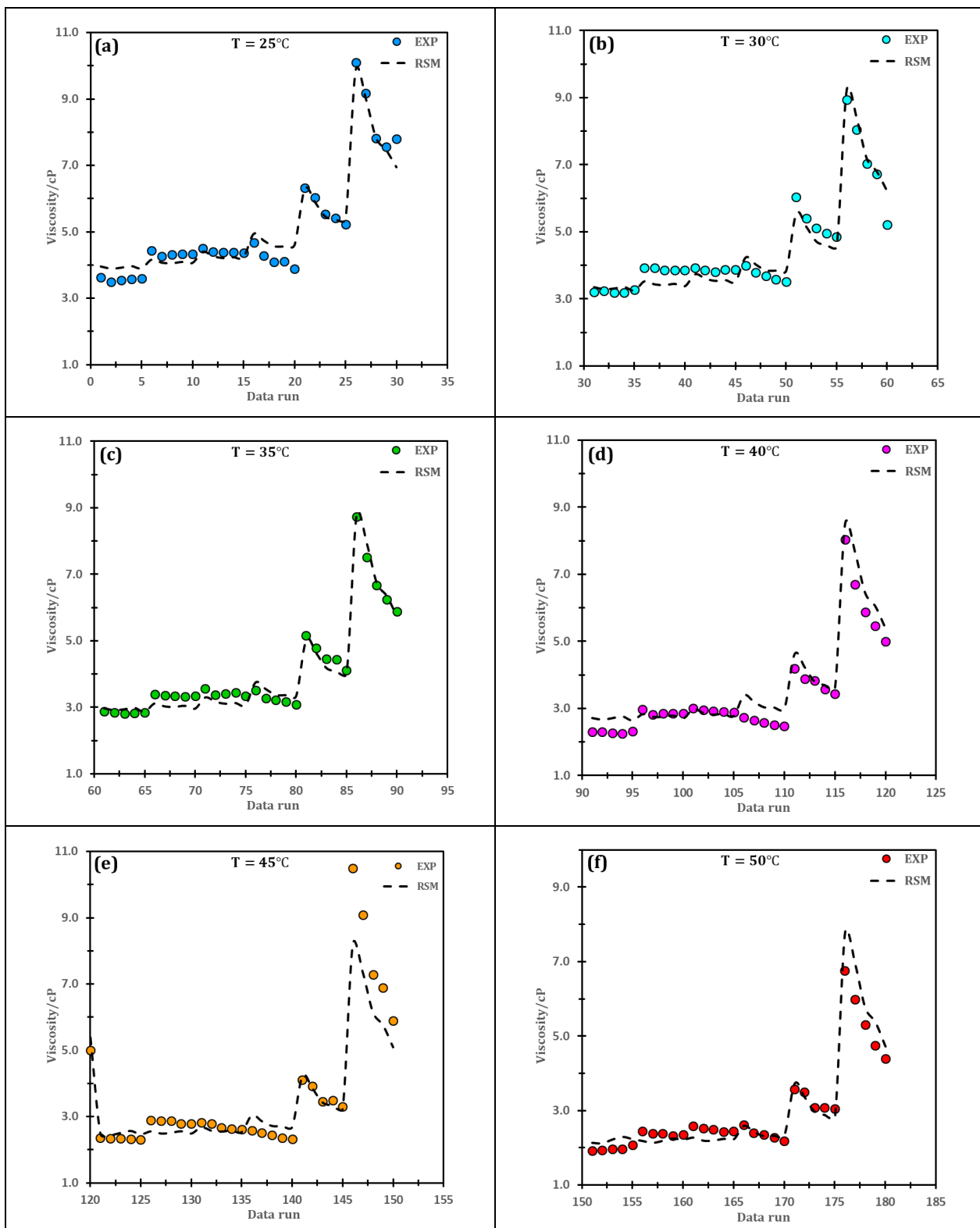


Figure 6. Correlation versus experimental data at different temperatures (RSM method).

As can be seen, for each temperature, some points are not fitted by the correlation. In the following, the results obtained from the ANN method are presented. As mentioned, the ANN method consists of three layers, and Figure 7 shows the amount of correlation coefficient value in all three layers.

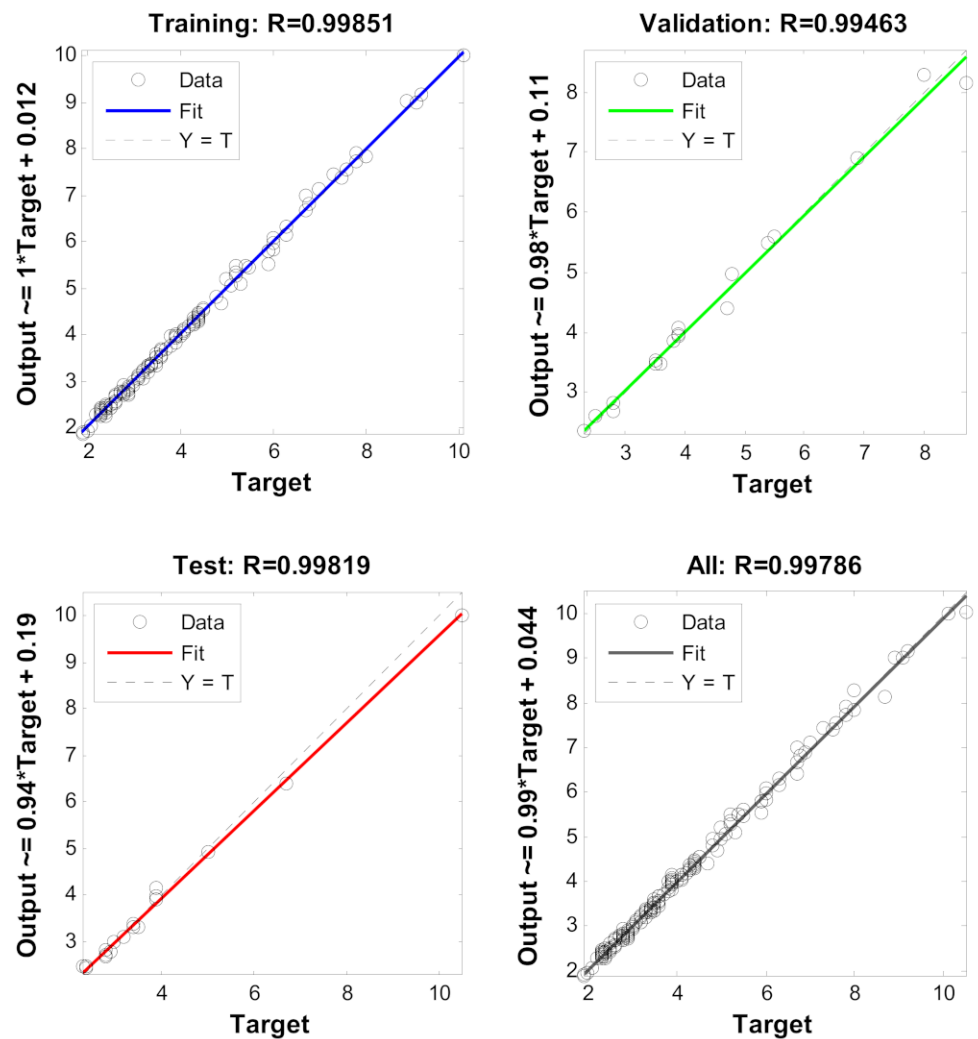


Figure 7. Investigation of accuracy in training, validation, and test sections of neural network algorithm and correlation coefficient values (R).

Note that the R-square for ANN, according to Figure 7, is $0.99786^2 \sim 0.995$. The accuracy of the neural network can be obtained by drawing the output data. In Figure 8, the viscosity value (output from the neural network) is compared with its experimental value.

It is found that the developed ANN can well estimate the viscosity. However, the values of the MOD are reported in Figure 9. As shown, the MOD_{maximum} for the developed neural network is approximately 6.7%. Figure 10 compares the results of ANN and RSM. At all temperatures, ANN accuracy outweighs the accuracy of the RSM method because it fits more points.

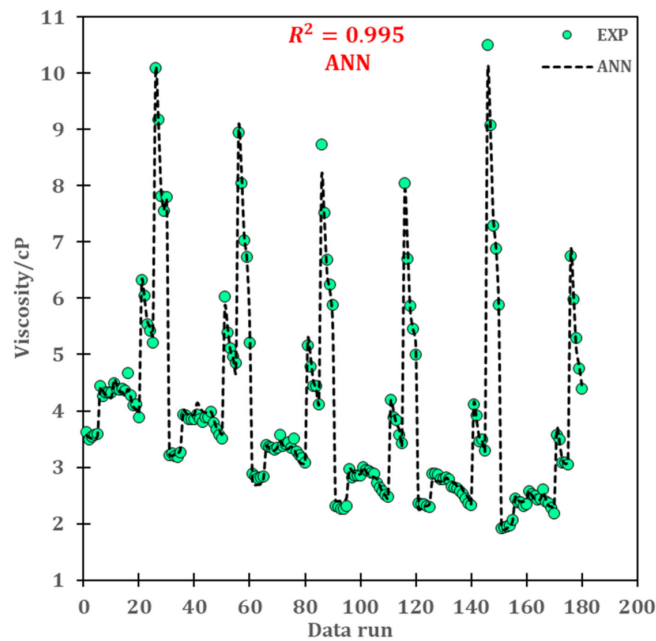


Figure 8. Investigating the ANN accuracy.

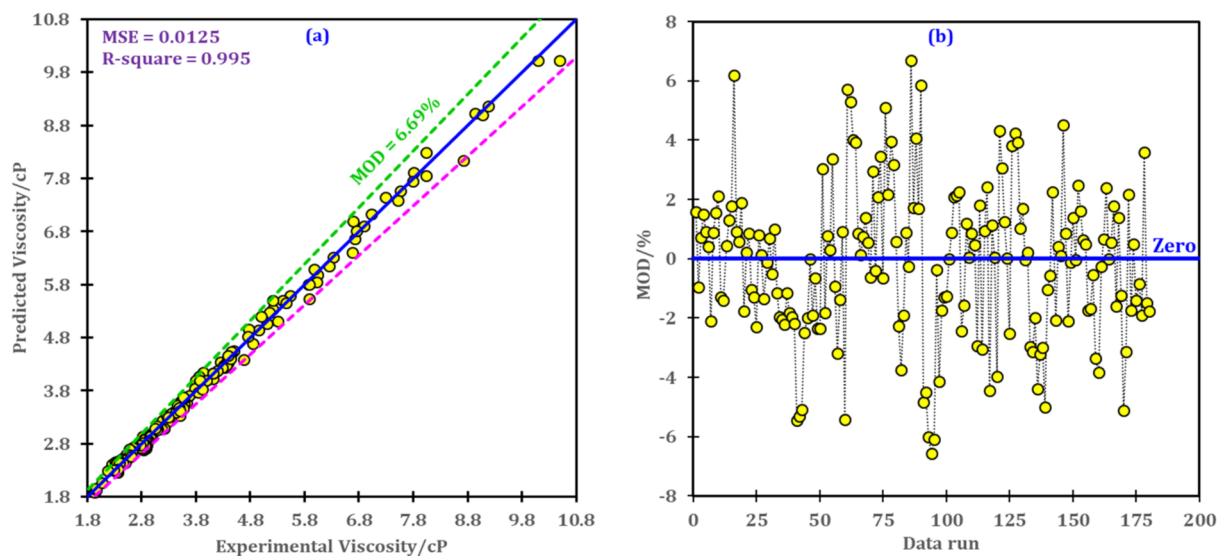


Figure 9. Margin of deviation for the ANN technique.

It should be noted that, for data run within the range of 146–150 in Figure 7e, the fluid has a non-newtonian behavior. Fluid behavior at volume fractions of 0.1, 0.2, and 0.4 is Newtonian and, for 0.8 and 1.6 vol.%, the behavior changed to non-Newtonian [61]. In other words, up to a volume fraction of 0.4, viscosity is not sensitive to one of the input parameters (shear rate), but it is quite sensitive to the shear rate at 0.8 and 1.6 vol.%. Therefore, it is logical that the accuracy of RSM is not high in this range. Therefore, using ANN for nanofluids with changing behavior from Newtonian to non-Newtonian is much better than the RSM method.

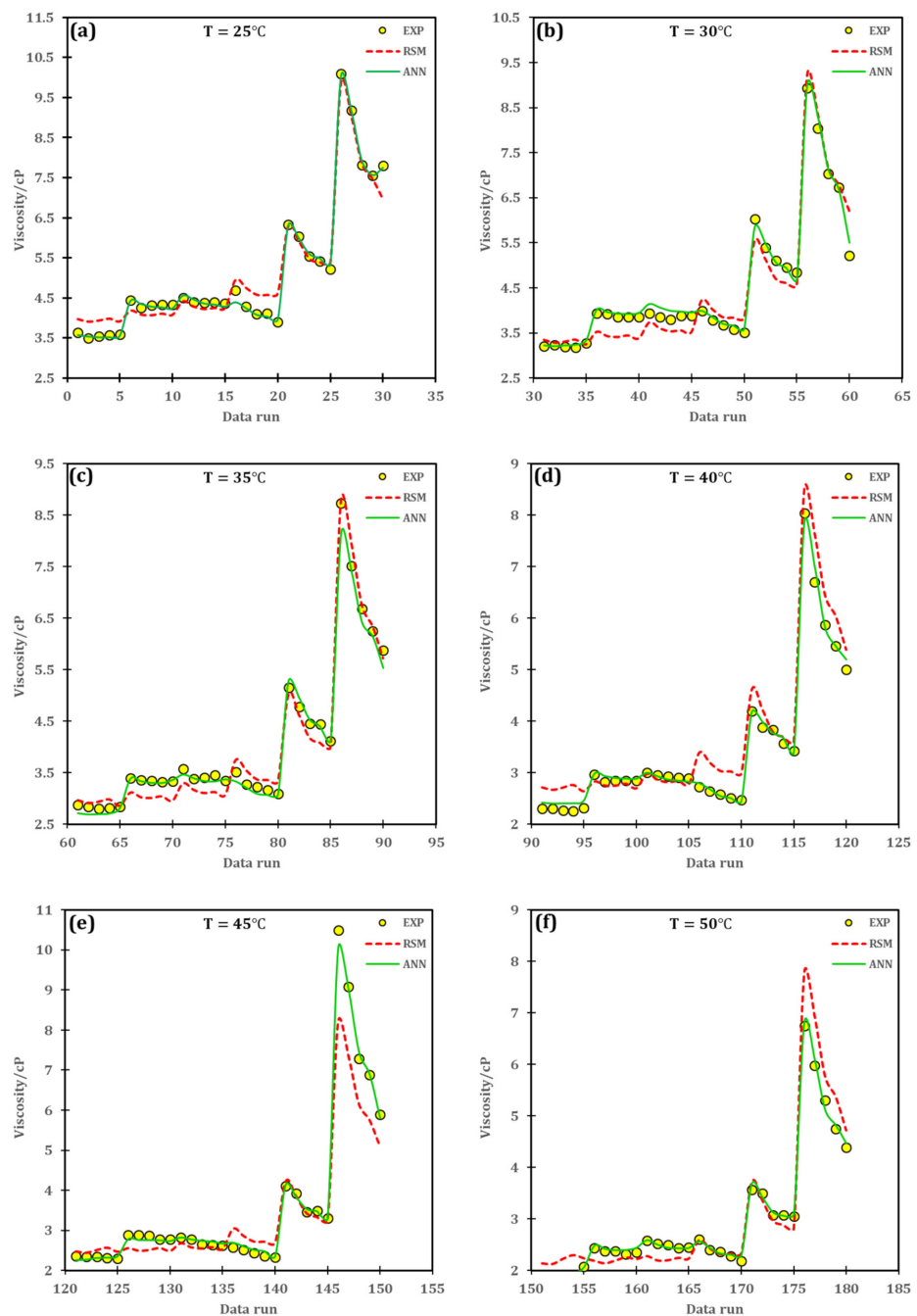


Figure 10. Comparison between ANN and RSM techniques.

6. Conclusions

In this study, the efficacy of loading GO and CuO nanoparticles into EG-water on viscosity was evaluated by applying two numerical techniques. The first technique employed the response surface methodology based on the design of experiments, while in the second technique, artificial intelligence algorithms are implemented to estimate the GO-CuO/water-EG hybrid nanofluid viscosity. Considering the possibility of non-Newtonian behavior of nanofluid, temperature, shear rate, and volume fraction were effective on the target variable and were defined in the implementation of both techniques. Considering two constraints maximizing the R-square as well as minimizing the mean square error, the best neural network and suitable polynomial were approved. It was found that the R-squared for ANN and RSM techniques can reach 0.995 and 0.944, respectively,

and, simultaneously, the mean square error for the approved ANN and RSM techniques was 0.0125 and 0.166, respectively, which indicate that using ANN is prioritized over the RSM technique.

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Conflicts of Interest: The authors declare no conflict of interest.

Nomenclature

ANOVA	Analysis of variance
ANN	Artificial neural network
EG	Ethylene glycol
GO	Graphene oxide
GON	Graphene oxide nanosheets
GN	Graphene nanosheets
k_{nf}	Nanofluid thermal conductivity / $Wm^{-1}K^{-1}$
k_{bf}	Base fluid thermal conductivity / $Wm^{-1}K^{-1}$
MSE	Mean square error
MOD	Margin of deviation
MWCNTs	Multi-walled carbon nanotubes
RSM	Response surface methodology
T	Temperature / $^{\circ}C$
TCR	Thermal conductivity ratio
Greek letter	
φ	Volume fraction / %
$\dot{\gamma}$	Shear rate / s^{-1}
μ	Viscosity / cP

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