

Investigation of Thermal Conductivity of Nanofluids Containing Decorated Ag Nanorods with Cu Nanoparticles Using Statistical Method

Farid Nourivatan¹, Sedigheh Abbasi^{2,*}

¹ Department of Chemical Engineering, Sirjan Branch, Islamic Azad University, Sirjan, Iran

² Esfarayen University of Technology, Esfarayen, North Khorasan, Iran

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ABSTRACT

In this paper, the statistical analysis of thermal conductivity of nanofluids containing decorated Ag nanorods with Cu nanoparticles was performed. For this purpose, Ag-Cu hybrid was synthesized and characterized using transmission electron microscopy (TEM) and X-ray diffraction (XRD) analysis. TEM studies showed that Cu nanoparticles successfully decorated the outer surface of Ag nanorods. The XRD pattern of decorated Ag nanorods with Cu nanoparticles reveals that Ag-Cu hybrid displayed both of the peaks assigned to Ag and Cu, respectively. Investigation of the effect of temperature and mass fraction on thermal conductivity of nanofluids showed that thermal conductivity of all nanofluids increases with temperature and mass fraction. The results of the statistical analysis of thermal conductivity confirm that there is a significant difference among five temperatures and three tested weight fractions for thermal conductivity of all nanofluids. However, the influence of temperature is more significant than that of mass fraction.

1-Introduction

Conventional fluids such as water and oil have poor thermal properties that restrict the heat transfer performance compared to most of the solids [1]. The dispersion of nanoparticles (the sizes of which are less than 100 nm) with excellent thermo physical properties such as thermal conductivity in the conventional heat transfer fluids improves thermal conductivity, thermal diffusivity and convective heat transfer coefficients of the base fluids [2]. Among the various thermophysical properties of nanofluids, thermal conductivity is the most complex and for many applications the most important one [3]. Several parameters such as thermal conductivity of the base fluid and the nanoparticles, the volume fraction, the shape and kind of the nanoparticles, the surface area,

and the temperature of nanofluids affect the thermal conductivity of nanofluids [1]. The effective thermal conductivity (k_{eff}) of a mixture containing two components is given by the following equation [4].

$$k_{eff} = \frac{k_p \phi_p (dT/dx)_p + k_b \phi_b (dT/dx)_b}{\phi_p (dT/dx)_p + \phi_b (dT/dx)_b} \quad (1)$$

Where k_p is thermal conductivity of the particle, k_b is the thermal conductivity of the base fluid, and ϕ is the particle volume fraction in the suspension.

Thermal conductivity of solid particles is much higher than that of the conventional heat transfer fluids. Up to now, various types of nanoparticles such as metallic, non-metallic, polymeric and nanotubes have been suspended in the base fluids [5, 6]. Among various metallic nanoparticles as seen in Table 1, silver (Ag) and

* Corresponding author:

E-mail address: abasi_1362@yahoo.com , s.abbasi@esfarayen.ac.ir

copper (Cu) nanoparticles are a good choice for preparation of nanofluids due to their very excellent thermal conductivity [3].

Table 1. Thermal Conductivity Values of various metallic nanoparticles.

Metallic nanoparticles	Thermal conductivity/ $\text{W m}^{-1} \text{K}^{-1}$	Temperature/K
Aluminum (Al)	237	293
Copper (Cu)	401	273-373
Gold (Au)	318	273-373
Iron (Fe)	80.40	273-373
Silver (Ag)	429	300

Liu et al. [7] investigated the thermal conductivity of ethylene glycol (EG), water, and synthetic engine oil in the presence of copper (Cu), copper oxide (CuO), and multi-walled carbon nanotube (MWCNT). Their results demonstrated that nanoparticles loading plays a key role in the thermal conductivity enhancement of nanofluids, so that in Cu-water nanofluids thermal conductivity enhancement at 0.1 vol.% was 23.8%.

Jamal-Abadi et al. [8] reported the effect of concentration (500, 1000 and 2000 ppm) and type of nanoparticles such as Al and Cu on the thermal conductivity enhancement of nanofluid. Their results showed that thermal conductivity of nanofluids was higher than that of the base fluid and thermal conductivity of Cu/water nanofluid was more than that of Al/water nanofluid, because the thermal conductivity of Cu is higher in comparison with Al.

Fang et al. [9] reported the influence of silver (Ag) nanoparticles of various shapes (nanospheres, nanowires, and nanoflakes) and temperature in the range of 10 to 30°C on the thermal conductivity enhancement of ethylene glycol based suspensions. Their experiments revealed that the thermal conductivity of nanofluids increased with raising the temperature. Meanwhile, they reported that nanofluids containing Ag nanowires caused the greatest relative enhancement up to 15.6% at the highest loading of nearly 0.1 vol. %. It could be related to the high aspect ratio of Ag nanowires. Godson et al. [10] studied the effect of different factors such as temperature (between 50°C and 90°C) and concentration (0.3, 0.6, and 0.9%

volume concentration) on the thermal conductivity of Ag-deionized water nanofluid. Their results showed that the thermal conductivity increased with the increase in temperature and particle concentration.

Saterlie et al. [11] prepared the stable nanofluids of copper nanoparticles into water base fluid and studied the influence of Cu nanoparticles concentrations varying from 0.55 to 1 vol.%. Their experiments demonstrated that the thermal conductivity enhancement of 22% and 48% over water was observed for the 0.55 and 1 vol.% Cu nanofluids, respectively.

The shape of nanoparticles (spherical and cylindrical) is the key factor in the thermal conductivity enhancement of nanofluids. The previous investigations showed that due to a large length to diameter ratio, the nanofluids containing cylindrical (nanorod or tube) nanoparticles have higher thermal conductivity compared to nanofluids having spherical nanoparticles [9, 12].

Jha et al. [13] reported the effect of decorated carbon nanotubes with various nanoparticles such as Ag, Au, and Pd on the thermal conductivity of water and ethylene glycol based nanofluids. Their experiments demonstrated that nanofluids maintain the same sequence of thermal conductivity as metal nanoparticles of Ag-MWNTs > Au-MWNTs > Pd-MWNTs.

Amiri et al. [14] investigated the dispersion stability and thermal conductivity of multi-walled carbon nanotubes nanofluids in the presence of gum arabic (MWCNT-GA) as well as functionalized MWCNT with cysteine (MWCNT-Cys) and silver (MWCNT-Ag). The effect of temperature and mass concentration on the enhancement of thermal conductivity revealed that the covalent functionalization by Ag is more effective than noncovalent functionalization.

Although thermal conductivity of nanofluids containing various nanoparticles such as Cu and Ag has been investigated by many researchers, the effect of decorated Ag nanorods with Cu nanoparticles on the thermal behavior has never been reported so far. Meanwhile, up to now, no paper has reported the statistical analysis of the thermal conductivity of these nanofluids. Therefore, in this study, we aim to report for the first time the effect of modified Ag nanorods with Cu nanoparticles on the thermal

conductivity of nanofluids and statistical analysis of the thermal conductivity of nanofluid containing Ag and Cu nanorods and decorated Ag nanorods with Cu nanoparticles.

2- Materials and methods

Silver nitrate (AgNO_3 , $M=169.87$, Merck Co., Germany), polyvinyl pyrrolidone (PVP, $(\text{C}_6\text{H}_9\text{NO})_n$, Merck Co., Germany), ethylene glycol (EG, $\text{C}_2\text{H}_6\text{O}_2$, $M=62.07$, Merck Co., Germany), sodium hydroxide (NaOH , $M=40$, Merck Co., Germany), copper chloride (CuCl_2 , $M=134.42$, Merck Co., Germany), cetyl trimethyl ammonium bromide (CTAB, $\text{C}_{19}\text{H}_{42}\text{BrN}$, $M=364.46$, Merck Co., Germany), hydrazine hydrate ($\text{N}_2\text{H}_4\cdot\text{H}_2\text{O}$, $M=50.06$, Merck Co., Germany), ethanol (96%, $\text{C}_2\text{H}_6\text{O}$, $M=46.07$, Merck Co., Germany) and sodium chloride (NaCl , $M=58.44$, Merck Co., Germany) were used for the synthesis of Ag nanorods and decorated Ag nanorods with Cu nanoparticles.

The Ag nanorods used in this study have been prepared by a simple hydrothermal method. Firstly, Ag nanorods were synthesized according to the Liu et al. [15] method. Typically, 0.67 g of PVP and 0.007 g of NaCl were added to 42 Cm^3 of EG, and formed the clear solution A. 0.67 g of AgNO_3 was dissolved in 42 Cm^3 of EG to form solution B, and then solution B was added dropwise to solution A and stirred for 10 min. The reaction mixture solution was then transferred into a Teflon-lined stainless-steel autoclave (200 Cm^3). The autoclave was sealed and treated at 160 °C for 90 min in an oven. Finally, the autoclave was cooled down to room temperature. The resulting precipitates were centrifuged and dried at 80°C for 12 h in order to remove water molecules.

For decoration of Ag nanorods with Cu nanoparticles, firstly 1.2 g of NaOH was dissolved in 10 Cm^3 of distilled water. Then, 0.021 g of synthesized Ag nanorods was added to the NaOH solution. The mixture was sonicated at room temperature for 5 min in an ultrasound bath. Subsequently, 0.0107 g of CuCl_2 was dispersed in this solution. Then, 0.218 g of CTAB was added and agitated for 30 minutes at 50 °C by a magnetic stirrer at a high speed to ensure the complete dissolution of CTAB. Finally, 0.1 Cm^3 of hydrazine hydrate was added to the mixture and the resulting mixtures were maintained at room temperature

for 1 h. The obtained precipitates were centrifuged, washed with water and ethanol and dried at 80°C for 12h.

X-ray powder diffraction was performed to characterize the phase composition and crystal structure of the samples, using a PHILIPS-binary equipment as well as a Bruker D8 Advance (40 kV/30 mA) with Cu-K α (1.542Å) radiation. The scanning velocity was 0.02°s⁻¹, and the 2 θ range was 2° to 90°. Transmission electron microscopy (TEM) micrograph of decorated Ag nanorods with Cu nanoparticles was obtained using a LEO 912 AB system operating at 120kV.

KD2 Pro thermal property analyzer purchased from Decagon Devices Inc. was used to measure the thermal conductivity of nanofluids. The transient hot wire technique was used for the measurement of thermal conductivity of nanofluids. In this study, we used the single-needle (KS-1) with 60 mm length and 1.3 mm diameter and an accuracy of $\pm 0.01 \text{ W}/(\text{m}\cdot\text{K})$ from 0.02 - 0.2 $\text{W}/(\text{m}\cdot\text{K})$. The measurements were carried out in the temperature and mass fraction ranges of 20 to 60 °C and 0.25 wt% to 0.5 wt%, respectively. A certain amount of each nanoparticle was dispersed in water and the mixture was put for 2 h in an ultra-sound bath. In order to change and control the temperature, the KD2 Pro device was connected to a constant temperature bath (Thermo Haake K10 TT4310) which had a circulator and was able to maintain temperature uniformity. For more accuracy, we maintained the sample and probe in a double walled cylindrical container having liquid circulating facility at a constant temperature and waited for about 15 min between the readings. Fig. 1 illustrates the use of KD2 Pro device. A number of measurements were taken for each sample and only those measurements resulted with the mean correlation coefficient $r^2 > 0.9998$ were considered. All experiments and measurements were carried out in triplicate, and the data were subjected to analysis of variance (ANOVA). Two factor completely randomized designs were used for statistical analysis of the thermal conductivity using MSTATC software (Ver 1.42). Duncan's multiple range test was applied to the study of significant differences between the means.

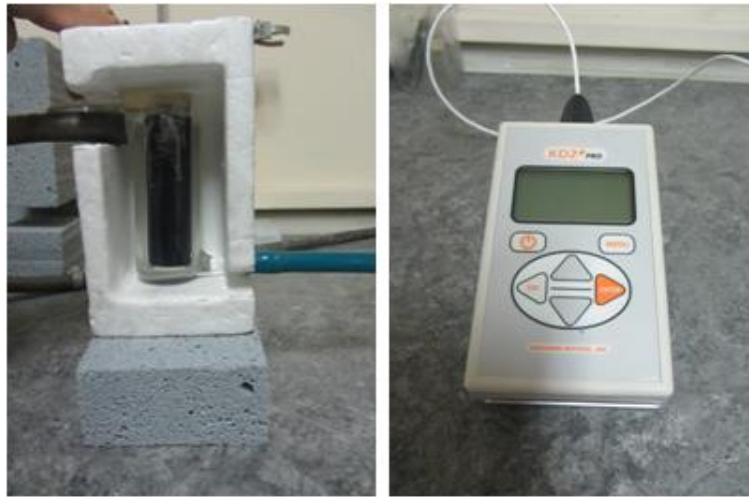


Fig. 1. Experimental set up (KD2 Pro device) for thermal conductivity measurements.

The level of statistical significance was determined at 95%. Therefore, P values less than 0.05 were considered as statistically significant. If the significant difference was found, the treatments were compared by using Duncan's multiple comparison test. For the investigation of the combined effects of temperature and weight fraction on the thermal conductivity of nanofluids, response surface method was carried out using Minitab Release software (Ver 11.12). If the significant difference was found, the treatments were compared by using Duncan's multiple comparison test.

3- Results and discussion

3-1- X-Ray diffraction

Fig. 2 shows the XRD pattern of decorated Ag nanorods with Cu nanoparticles. The observed diffractions in the Ag-Cu hybrid display both of the peaks assigned to the Ag and Cu, respectively. The XRD pattern reveals that the characteristic peaks at 38.11° , 44.27° and 64.42° are corresponding to the (111), (200) and (220) Bragg reflection planes of Ag, respectively. Meanwhile, the XRD results reveal that the Ag nanorods had cubic crystal system, with lattice constants a, b and c = 4.08°A , which are in good agreement with other literatures [15].

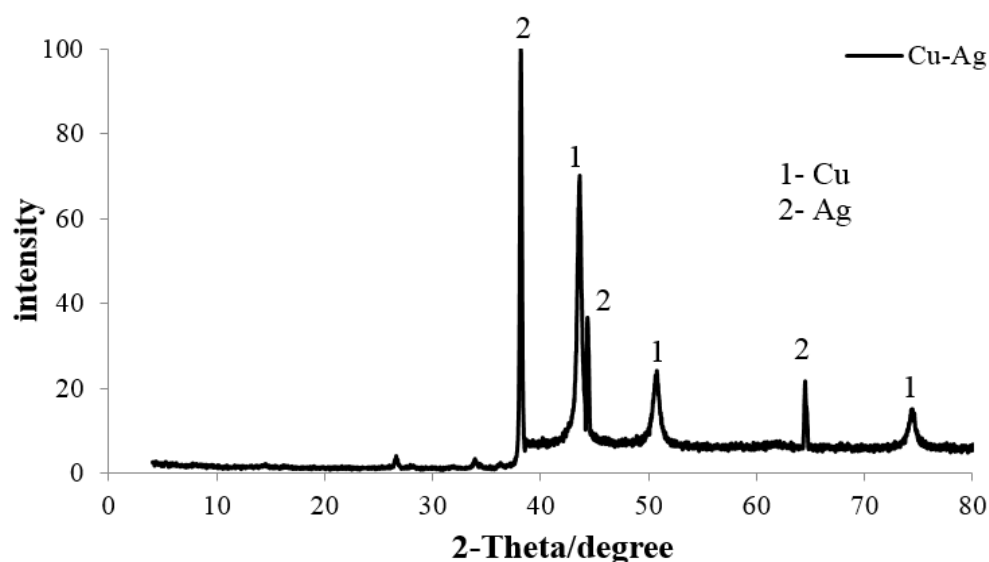


Fig. 2. XRD pattern of the synthesized Ag-Cu hybrid.

The XRD pattern of the Ag-Cu hybrid demonstrate that the peaks at the 2θ values of 43.47° , 50.67° and 74.67° can be associated with (1 1 1), (200) and (220) Bragg reflection planes of Cu nanoparticles, respectively. Meanwhile, the XRD results reveal that the Cu nanoparticles had cubic structure with lattice constants a , b and $c = 3.59 \text{ \AA}$

3-2- TEM studies

Fig. 3 shows TEM images of Ag-Cu hybrid, which reveals that the outer surface of Ag nanorods is successfully coated with Cu nanoparticles. It is found that the dominant mechanism in decoration of outer surface of Ag nanorods with Cu nanoparticles is absorption of copper ions in the solution to the surfaces of Ag nanorods due to the electrostatic attraction and then decoration of outer surface of Ag nanorods with Cu nanoparticles [16].

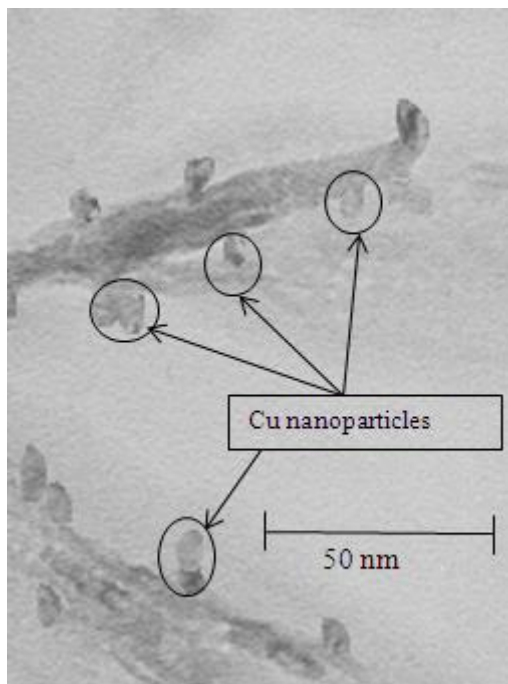


Fig. 3. TEM image of the synthesized Ag-Cu hybrid.

3-3- Thermal conductivity studies

The calibration of experimental apparatus was done with glycerol. The standard thermal conductivity of glycerol is $0.285 \text{ W/(m}\cdot\text{K)}$ at 20°C . To improve the accuracy of experimental results, the needle of KD2 pro thermal property analyzer was maintained in the nanofluids and

15 min of equilibration time was allowed. In addition, the insulation of the vial and needle is a key factor for minimization of error during the measurements. Therefore, the temperature of nanofluids and needle should be constant and prevent from free convection.

Fig. 4 depicts the measured thermal conductivity of Ag nanorod/water nanofluids as a function of temperature at two different mass fractions (0.25 and 0.5 % wt).

As we know, the temperature of nanofluids is a key factor in the thermal conductivity of nanofluids. Therefore, in this study we investigate the thermal conductivity of nanofluid as a function of temperature in the range varying from 20 to 60°C . In Fig. 4, it is clear that the thermal conductivity increases with increasing the temperature and Ag nanorod concentration. Meanwhile, it can be deduced that temperature has a more significant effect than the mass concentration. In the previous studies, it has been reported that the thermal conductivity of nanofluids containing spherical metal or metal oxide nanoparticles was affected by temperature due to the Brownian motion of nanoparticles. In the Brownian motion, nanoparticles which are suspended in the base fluid had a random motion as a result of their collision with atoms or molecules of the base fluid. Therefore, it can be deduced that the increasing of fluid temperature leads to the enhancement of collision between nanoparticles and molecules of the base fluid [1, 17].

Fig. 5 depicts the thermal conductivity of nanofluids of decorated Ag nanorods with Cu nanoparticles as a function of temperatures. The mass fraction of the Ag-Cu hybrid in water is 0.25, 0.5 and 1 % wt. Meanwhile, from Fig. 5 it can be inferred that at 60°C , by increasing the concentration of Ag-Cu hybrid from 0.25 to 1 % wt., thermal conductivity of nanofluids increased from $2.74 \text{ W/(m}\cdot\text{K)}$ to $3.79 \text{ W/(m}\cdot\text{K)}$. Therefore, the augmentation of thermal conductivity is equal to 38.3 %. Thus, it can be inferred that the effect of concentration is not significant and can be negligible. However, from Fig. 5 it can be inferred that by increasing the temperature from 20 to 60°C , the thermal conductivity of nanofluids containing 0.5 % wt. of Ag-Cu hybrid increased 110%.

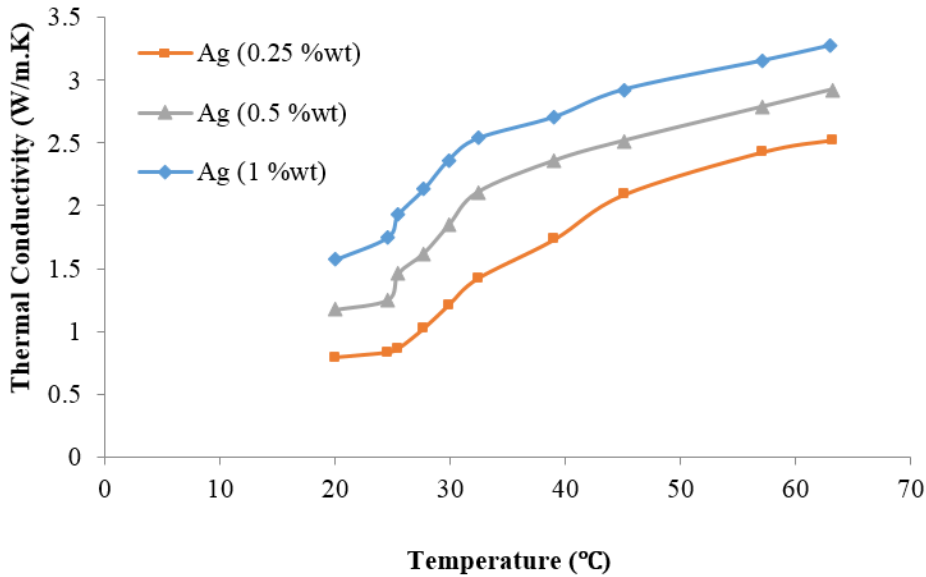


Fig. 4. Dependence of the thermal conductivity of Ag nanorod / water nanofluids on temperature at different mass fractions.

The enhancement of thermal conductivity can be attributed to the effect of temperature on the destruction of the hydrogen bond of water. The hydrogen bond of water was weakened due to the augmentation of temperature and this leads to the destruction of the water molecules structure. Therefore, by increasing the temperature, the produced free water molecules will increase. These free water molecules can be

arranged around the Ag-Cu hybrid surface and form the liquid layer. This liquid layer has a higher thermal conductivity than the bulk liquid [6, 18]. Therefore, in the water-based nanofluids, in addition to the Brownian motion, the chemical functionalized groups have a key effect on the amount of energy which is transferred into the nanofluids by changing the temperature.

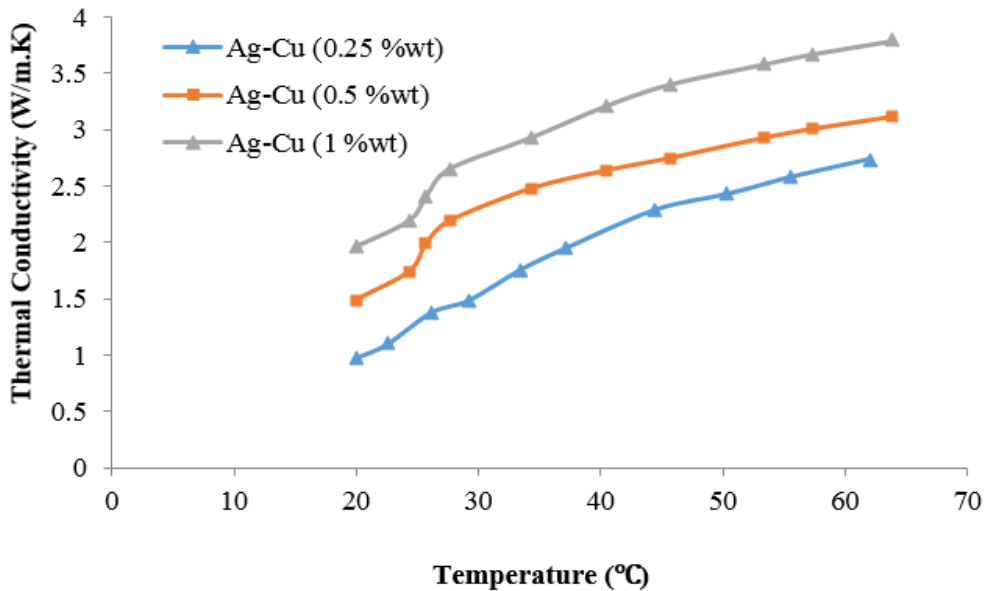


Fig. 5. Dependence of the thermal conductivity of Ag-Cu hybrid/ water nanofluids on temperature at different mass fractions.

Table 2. Analysis of variance of thermal conductivity of Ag nanofluids.

* Significant at 5% level of probability

Source	Degrees of Freedom	Sum of Squares	Mean Square	F Value	Prob
Temperature	4	16.535	4.134	5654.108	0.000*
Weight fraction	2	5.938	2.969	4061.061	0.000*
Temperature and Weight fraction	8	0.253	0.032	43.2644	0.000*
Error	30	0.022	0.001	----	----
Total	44	22.748	----	----	----

3-4-Data analysis and statistical studies

3-4-1-Thermal conductivity of nanofluids containing Ag nanorods

The ANOVA table of Ag nanofluids is shown in Table 2. From this table it can be inferred that temperature, weight fraction and their combined effect have a reasonable impact on the thermal conductivity of Ag nanofluids.

The effect of temperature on the thermal conductivity of Ag nanofluids is demonstrated in Fig. 6. It is clear that thermal conductivity of Ag nanofluids increased with respect to temperature. The index was increased from 20°C (1.178 W/m. K) to 30°C (1.83 W/m. K) equal to 55.34%, from 30°C to 40°C (2.308W/m. K) equal to 26.12%, from 40°C to 50°C (2.659 W/m. K) equal to 15.20%, and from 50°C to 60°C (2.862 W/m. K) equal to 7.63%. Meanwhile, it

can be inferred that there was a completely significant difference among different temperatures in terms of thermal conductivity.

Fig. 7 illustrates the effect of weight fraction on the thermal conductivity of nanofluids containing Ag nanorods. It is clear that there is a significant difference among three weight fractions for thermal conductivity of Ag nanorods nanofluid ($\alpha=0.05$) and the thermal conductivity of nanofluids increases with respect to the weight fraction. From Fig. 7 it can be inferred that the least thermal conductivity was recorded for the weight fraction of 0.25 %wt. equal to 1.71W/m. K. In addition, maximum thermal conductivity was related to the weight fraction of 1 %wt. equal to 2.599 W/m. K.

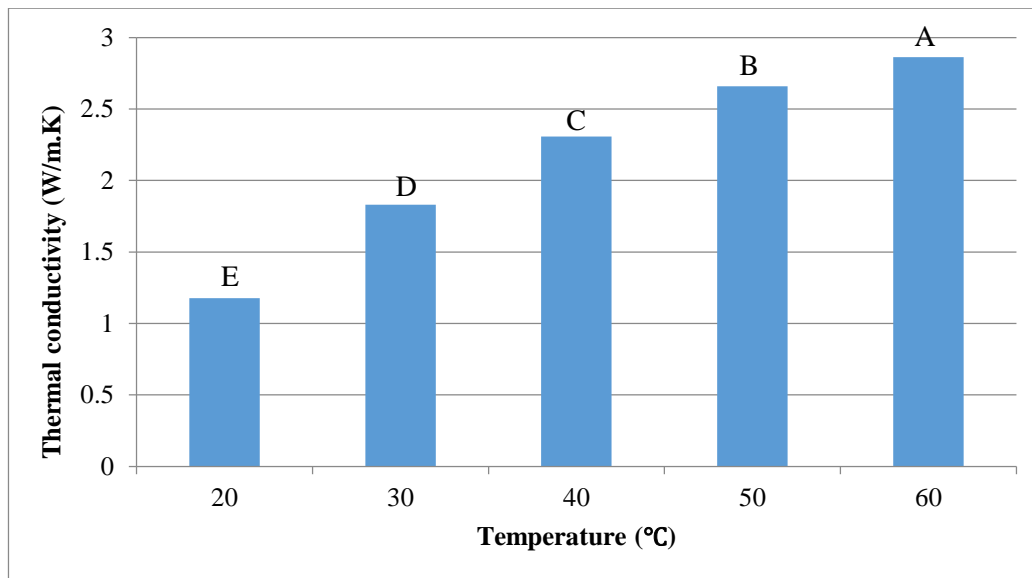


Fig. 6. Effect of temperature on thermal conductivity of Ag nanofluid. Means with different letters are significantly different based on Duncan's multiple range test ($\alpha=0.05$).

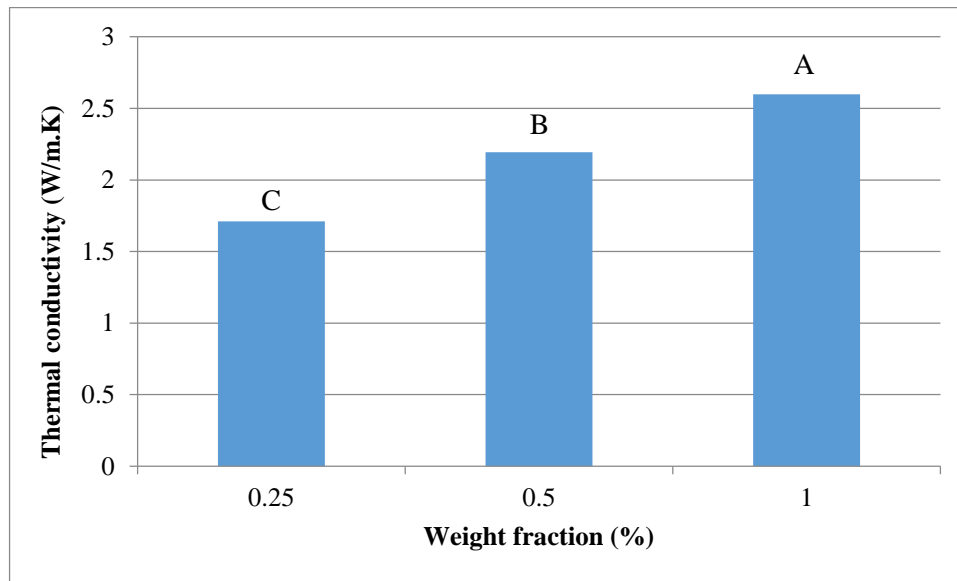


Fig. 7. Effect of weight fraction on thermal conductivity of Ag nanofluid. Means with different letters are significantly different based on Duncan's multiple range test ($\alpha=0.05$).

The response surface fitting method was used in order to study the combined effect of temperature and weight fraction on the thermal conductivity of nanofluids. Fig. 8, which shows the surface response of thermal conductivity of Ag nanofluid, implies that the thermal conductivity of Ag nanofluid increases with increasing temperature and weight fraction, although the weight fraction variations of the thermal conductivity is less compared to temperature. It can be related to the destruction of the structure of water molecules due to the augmentation of temperature. Because by increasing the temperature, the hydrogen bond of water was weakened, the structure of water molecules was destroyed and the number of free water molecules increased. These free water molecules can be arranged around the Ag nanorods surface. This liquid layer which was produced due to the chemical surfaces of Ag nanorods and van der Waals force between the water molecules has a higher thermal conductivity than the bulk liquid [1, 18]. Fig. 9 shows the contour lines of the thermal conductivity of Ag nanofluid at different temperatures and weight fractions of Ag nanorods. It is obvious that weight fraction decreases with increasing temperature.

3-4-2-Thermal conductivity of nanofluids containing decorated Ag nanorods with Cu nanoparticles

Table 3 shows the ANOVA table of Ag-Cu nanofluids. As can be seen, the thermal conductivity of Ag-Cu nanofluids significantly changed with respect to the temperature and weight fraction. Meanwhile, it can be observed that the combined effect of temperature and weight fraction on the thermal conductivity of Ag-Cu nanofluids is completely significant at 5% level of probability.

Figs. 10 and 11 depict the effect of temperature and weight fraction on the thermal conductivity value of nanofluids containing decorated Ag nanorods with Cu nanoparticles, respectively. It is clear that the thermal conductivity of Ag-Cu nanofluids increased with respect to both temperature and weight fraction. According to these figures, it can be inferred that at 5% level of probability temperature and weight fraction have a significant effect on the thermal conductivity of Ag-Cu nanofluids. By increasing the temperature from 20 to 60°C the thermal conductivity of Ag-Cu nanofluids increased from 1.48W/m.K to 3.143 W/m. K equal to 112.36%.

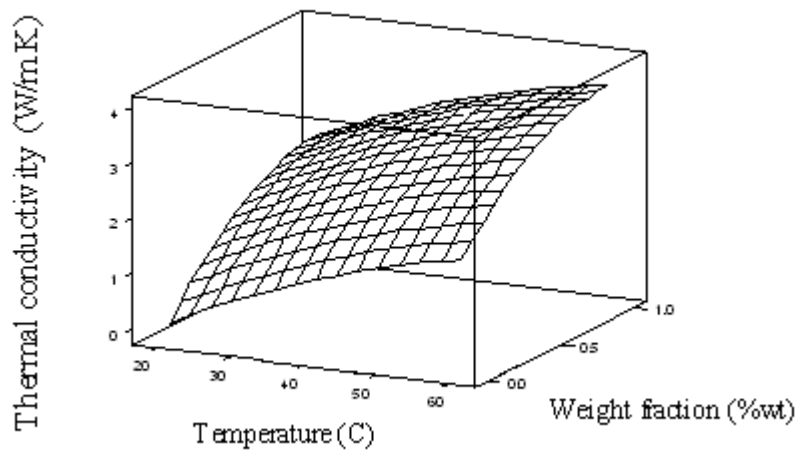


Fig. 8. Response surface of the thermal conductivity of Ag nanofluid versus temperature and weight fraction.

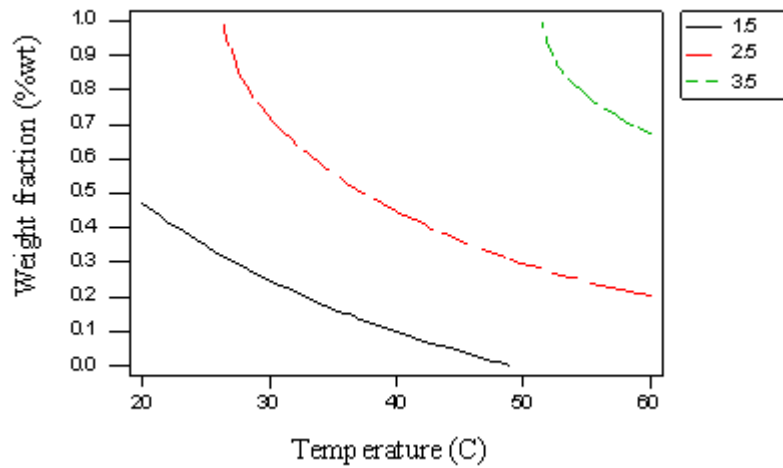


Fig. 9. Contour lines of thermal conductivity of Ag nanofluid at different temperatures and weight fraction.

Table 3. Analysis of variance of thermal conductivity of Ag-Cu nanofluids.

Source	Degrees of Freedom	Sum of Squares	Mean Square	F Value	Prob
Temperature	4	15.556	3.889	10057.55	0.000*
Weight fraction	2	8.709	4.354	11261.455	0.000*
Temperature and weight fraction	8	0.202	0.025	65.392	0.000*
Error	30	0.012	0.000	----	----
Total	44	24.478	----	----	----

* Significant at 5% level of probability

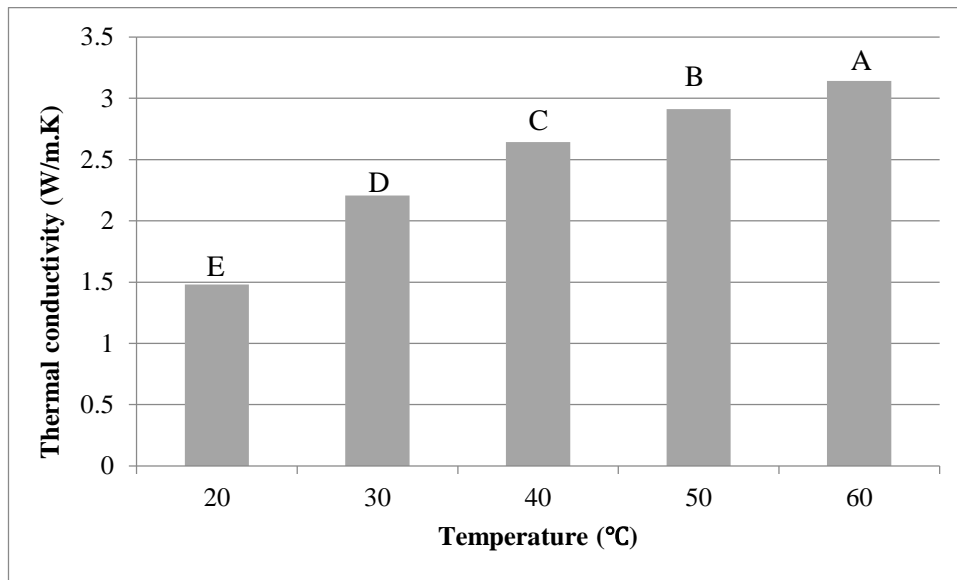


Fig. 10. Effect of temperature on thermal conductivity of Ag-Cu nanofluid. Means with different letters are significantly different based on Duncan's multiple range test ($\alpha=0.05$).

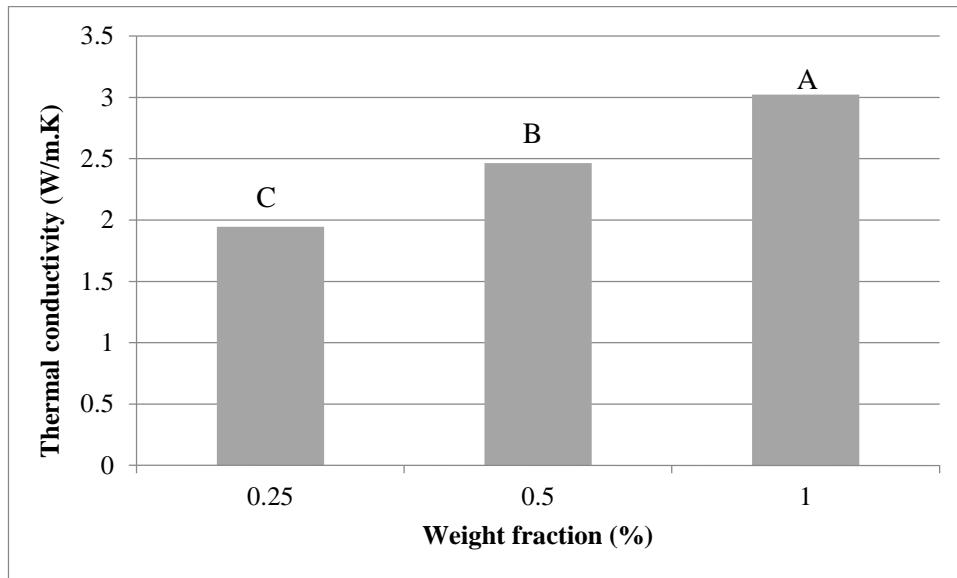


Fig. 11. Effect of weight fraction on thermal conductivity of Ag-Cu nanofluid. Means with different letters are significantly different based on Duncan's multiple range test ($\alpha=0.05$).

Whereas, increasing the weight fraction from 0.25 to 1 %wt. leads to the augmentation of the thermal conductivity of Ag-Cu nanofluids from 1.94 W/m. K to 3.02 W/m. K equal to 55.37%. Therefore, it can be inferred that the effect of temperature on the thermal conductivity of Ag-Cu nanofluids is more significant than that of weight fraction which is consistent with a previous work [6]. As mentioned before, it can be related to the effect of augmentation of

temperature on the destruction of water molecules. Therefore, by increasing the temperature, the number of free water molecules increased and arranged on the outer surface of Ag-Cu hybrid. This liquid layer has a higher thermal conductivity than the bulk liquid [1, 6]. The surface response and contour lines of the thermal conductivity of Ag-Cu nanofluids are depicted in Figs. 12 and 13, respectively.

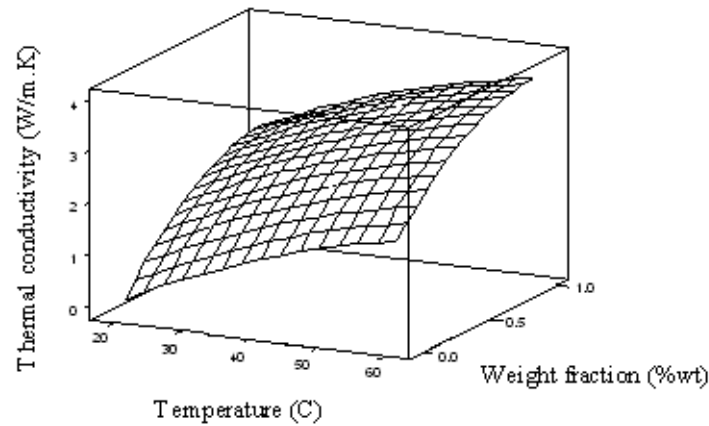


Fig. 12. Response surface of thermal conductivity of Ag-Cu nanofluid versus temperature and weight fraction.

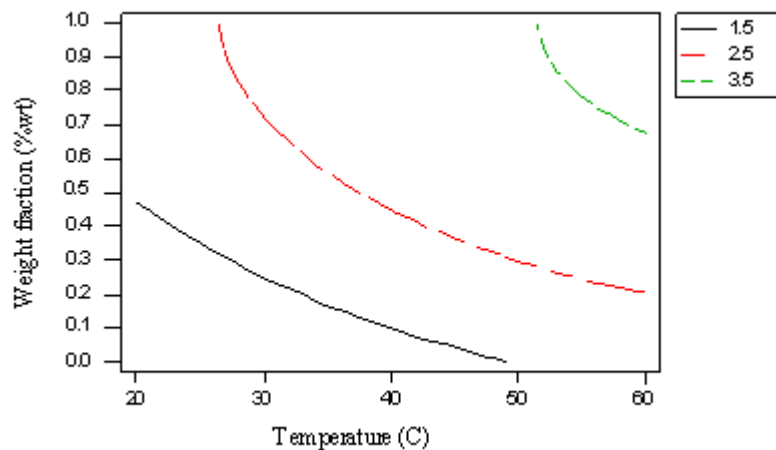


Fig. 13. Contour lines of thermal conductivity of Ag-Cu nanofluid at different temperatures and weight fraction.

From Fig. 12 it can be inferred that the thermal conductivity of nanofluid increases with respect to the temperature and weight fraction. However, the influence of temperature is more considerable than that of weight fraction. Fig. 13 shows the contour lines of thermal conductivity value of Ag-Cu nanofluids at different temperatures and weight fractions. The required temperature for reaching a certain value of thermal conductivity increases with decreasing the weight fraction.

4-Conclusions

In the current research, we synthesized decorated Ag nanorods with Cu nanoparticles and investigated the thermal conductivity

behavior of Ag and Cu nanofluids in water as a base fluid. The XRD results revealed that Ag nanorods and Cu nanoparticles in the synthesized hybrid possess cubic structure. TEM image of Ag-Cu hybrid reveals that the outer surface of Ag nanorods is successfully coated with Cu nanoparticles. The results show that the influence of temperature on the thermal conductivity of all studied nanofluids is more significant than the mass fraction. The statistical analysis of thermal conductivity shows that temperature, weight fraction, and their combined effect have a significant effect on the thermal conductivity of all nanofluids. Meanwhile, the results confirm that the thermal conductivity increases with respect to the temperature and weight fraction.

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