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INVESTIGATION OF THERMAL CONDUCTIVITY PROPERTY OF PLASMONIC NANOFLUIDS BASED ON GOLD NANORODS PREPARED BY SEED-MEDIATED GROWTH METHOD

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Key words: Nanofluid, Gold nanorods, Thermal conductivity, Transient hot wire method, Aspect ratio, Volume fraction.

Abstract

In this paper, nanofluids were prepared based on gold nanorods in basic fluid, water, by single-stage chemical reduction and in different volume fractions and the used gold nanorods were synthesized by seed-mediated growth method in different dimensional ratios. The properties of the prepared nanoparticles, including crystalline size, aspect ratio, surface properties, nanoparticle purity, shape and morphology of nanostructures were investigated using x-ray diffraction, UV-vis spectroscopy, FT-IR, and transmitted electron microscopy. The effect of changing parameters of Nano rod dimensions, changes in Nano rod volume fraction in water and also the effect of temperature on the nanofluid thermal conductivity coefficient were investigated using transient hot wire method. The results showed that reducing the aspect ratio, increasing the volume fraction and increasing the temperature increase the thermal conductivity. In fact, results show that an increase

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in the nanorods aspect ratio with a constant volume fraction of 1:50 of gold in water nanorod and at room temperature leads to a decrease in the thermal conductivity of the nanofluid. Also, increasing the two parameters of volume fraction and temperature significantly increases the thermal conductivity coefficient.

Introduction

Heat transfer plays a very important role in several key engineering sectors including microelectronics, power generation, transportation, automotive, aerospace, and nuclear power plants. Due to bugs in the use of traditional fluids and even micro-fluids, including sedimentation and deposition of particles, erosion, fouling of tubes and increasing pressure drop of the flow channel, the researchers turned to nanofluids (Eastman et al. 2001). The idea of using a nanofluid was proposed by Choi and Eastman (1995), and a major evolution in fluid heat transfer was generated. In fact, a new look at solid suspensions with particles in nanoscale dimensions presented, in which the small amount of corrosion reduced the impurities and pressure loss problems, and improved fluidity stability over sedimentation.

The nanofluid consists of two main components: base fluid and nanoparticles. The base fluid is the fluid to which nanoparticles are added, and the common fluids contain water, ethylene glycol and engine oil. Nanoparticles are also divided into three groups which are metallic, metallic oxides and non-metallic oxides that are dispersed in the base fluid. In general, two methods are considered for the preparation of nanoparticles: a two-stage method and a single-stage method. In a two-step procedure, the nanoparticles are first synthesized and then dispersed in a base fluid. In a one-stage process the synthesis of nanoparticles (by methods such as chemical/physical deposition or chemical reduction) is occurred with the combination of it with the base fluid simultaneously (Yu, Xie 2012). One-stage method for nanofluid production showed better stability than the two-stage method (Li et al. 2009). For more than a decade, researchers have used a single-step method to study nanofluids (Yu, Xie 2012, Li et al. 2009). Nanoparticles of several precious metals such as gold, silver, palladium and platinum have been investigated for the manufacture of nanofluids and their use in a variety of engineering applications due to their unique catalytic, electrical, magnetic, optical and mechanical properties (Tseng et al. 2013).

The thermal conductivity coefficient is one of the most important factors in the study of heat transfer. An overview of existing research shows that adding a small amount of nanoparticles would significantly increase the thermal conductivity of the nanofluid relative to the base fluid (Wang, Fu 2011, Li et al. 2005). Also, the thermal conductivity of the nanofluid depends on the parameters such as the composition of the chemical percent of the nanoparticles and nanofluids, the volumetric percent of nanoparticles, the shape and size of the particles, the activated surface materials, and the temperature (PAUL et al. 2011, SURESH et al. 2011). Two mechanisms are considered to increase the thermal conductivity of nanoparticles. The first mechanism is Brownian motion of the nanoparticles inside the fluid, which results in increased mixing and, in fact, heat transfer is facilitated and the thermal conductivity is increased. The second mechanism is the coupling between particles, which increases the thermal conductivity coefficient. In this mechanism, nanoparticles stick together, and create chains through which heat transfer becomes faster (YU, XIE 2012, LI et al. 2009). Various theories, including Maxwell's theory, the Hamilton-Crosser model and Bragman model have been proposed to calculate the thermal conductivity of Nano fluid, which Bragman model has better predictions than other models (SURESH et al. 2011):

$$\frac{k_{nf}}{k} = \frac{1}{4}[(3\varphi - 1)k_s + (2 - 3\varphi)K] + \frac{k}{4}\sqrt{\Delta} \quad (1)$$

$$\Delta = \left[(3\varphi - 1)^2\left(\frac{k_s}{k}\right)^2 + (2 - 3\varphi)^2 + 2(2 + 9\varphi - 9\varphi^2)\left(\frac{k_s}{k}\right)\right] \quad (2)$$

In the above equations, K_{nf}, k, k_s, and φ are the thermal conductivity of the nanofluid, the thermal conductivity of the base fluid, the thermal conductivity of the solid and the volume fraction of the nanoparticles (According to reference papers, the thermal conductivity of water and solids of gold were considered to be k=0.6 W/(m·k) and k_s=318 W/(m·k), respectively). It should be noted that these theories are not complete. SURESH et al. (2011) concluded that these predictions show less value than the measurements, and the reason for these observations is that the effects of particle size and intermolecular forces are not applied in these models. The researchers have proposed several laboratory methods for measuring thermal conductivity, the most common of which are: THW Transient hot- wire technique, Steady-state parallel-plate method, cylindrical cell method, and omega-3 method. Surface plasmon oscillations caused by free electron oscillations in the surface of metal nanoparticles are activated by appropriate wavelength and its rate depends on the permeation coefficient and particle geometry (RAETHER et al. 1988, ZAYATS et al. 2005). When surface plasmon oscillations are induced, the input photon energy is transmitted oscillatingly, and results in a significant amount of heat in the particles (RAETHER et al. 1988, GOVOROV et al. 2006). If the nanoparticle is placed in an environment such as water, it absorbs heat generated in the environment and increases its temperature (RICHARDSON et al. 2006). In other words, the absorption of light increases by the environment with using of nanoparticles which have surface plasmon oscillations (BOHREN et al. 2007). Therefore, the main goal is to improve the performance of nanofluids, and many researchers have studied various nanofluids to achieve this goal.

Sani et al. (2010, 2011) showed that single-wall carbon nanohorns could improve the nanoparticle optical properties. In another study, Tyagi et al. (2009) reported an increase of 10% in the efficiency of flat plate solar collectors when water fluid was used with aluminum nanoparticles instead of pure water. Also, Khullar et al. (2012) showed that aluminum nanofluid can be used to concentrate solar collectors. Lee et al. (2012) showed that it is possible to achieve a broadband absorption plasmonic nanofluid with the dispersion of gold nanocages in water fluid. Due to the solar radiation is includes all spectrum sizes, range from ultraviolet to infrared, broadband absorption is very desirable for solar energy applications. Taylor et al. (2013) reported that the shell thickness of gold should be less than 10 nm in order to stimulate surface plasmon oscillations of nanocages in the visible and near-infrared region, and making this shell is very difficult. A promising option to solve this problem is the use of gold nanorods. Gold nanorods can be easily prepared compared to gold nanocages, and their optical properties can be controlled simply by adjusting their aspect ratios. Nanoparticles of several noble metals like gold, silver, palladium and platinum have extensively been studied because of their unique catalytic, electrical, magnetic, optical, and mechanical properties that are different from the coarse grained counterparts of the same materials (Lo et al. 2007). Therefore, nanofluid optical properties optimization when used with gold nanorods can be very convenient, cost effective and functional. Therefore, it is very important to study the thermal conductivity of nanofluids on the basis of gold nanorods for use in industrial and medical fields in the future.

According to studies, no experimental article has ever investigated the thermal conductivity of nanofluids based on gold nanorods. The objectives of this study is to prepare nanofluids based on gold nanorods, to detect gold nanostructures by UV-vis, FT-IR spectroscopy, X-ray diffraction and transmitted electron microscopy, as well as measuring the thermal conductivity coefficient and evaluating its variation as a function of the nanorods' dimension and their volume fraction in water and temperature. It is likely that the level of thermal conductivity improvement will be significantly higher than all reported studies.

Materials and methods

Materials

Tetrachloroauric acid ($HAuCl_4.3H_2O$, 99.95%), Ascorbic acid (99%), Cetyltrimethylammonium bromide (CTAB, 99%), Sodium borohydride ($NaBH_4$, 99%) and Silver nitrate ($AgNO_3$) has been purchased from Merck company. Deionized water was used in the preparation of all aqueous solutions, as well as the washing of test dishes.

Preparation

In order to perform the experiment, gold nanorods was first prepared from seed-mediated growth method in three different proportions by changing the growth volume of silver nitrate growth controller (JANA et al. 2001a, b). The seed solution was prepared by adding $HAuCl_4$ (0.5 ml, 0.005 M) and $NaBH_4$ (0.6 ml, 0.01 M) to a continuously stirred CTAB (5 ml, 0.2 M) solution. Then stirring was continued for 2 minutes and the solution was kept at room temperature for 2 hours. In the preparation of the growth solution, a solution of CTAB (5 ml, 0.2 M) was separately mixed with 0.05 ml, 0.2 ml and 0.4 ml of $AgNO_3$ (0.004 M) and $HAuCl_4$ (1 ml, 0.005 M) solution. After gentle stirring, 70 µl of ascorbic acid solution (0.08 M) was added to the reaction containers. Ascorbic acid, as a weak reducing agent, changes the color of the growth solution from dark yellow to pale color. In the final step, 12 µl of the seed solution prepared in the previous step was added to the growth solution at 27-30°C. The color of the solvent in the stirring mode has gradually changed in 10 to 15 minutes. The order of the volumes of silver nitrate in the growth solution, respectively, produce gold nanorods with a short, medium and long dimension, with the color of blue, violet and red solutions. Nanofluids of gold-water nanorods with the different volume fractions were prepared with the same method. In this case, three volume fractions of nanofluids of gold-water nanorods, 1:50, 2:50 and 3:50 were prepared, while the aspect ratio of short gold nanorods were kept constant. The color of the solution turned out from violet to light pink. To investigate the effect of temperature on the thermal conductivity coefficient, the Nano fuids prepared with gold nanorods with a short aspect ratio and diluted in water with a volume fraction of 1:50, was incubated in baths with different temperatures of 25, 35, 45 and 55°C for 30 minutes and was sampled to measure thermal conductivity.

Properties Determination

The gold nanoscale absorption spectra were recorded using, the UV-1800 model of the UV-vis spectrophotometer manufactured by Japan's Shimadzu Corporation, and the aspect ratio of the prepared nanorods were obtained according to the linear relationship between peak position and the aspect ratio of the prepared nanorods (LINK, EL-SAYED 1999, HUTTER, FENDLER 2004, MOHAMMADI et al. 2009, TAKAHASHI et al. 2008, PALIK 1998, WANG et al. 2009, JAIN et al. 2006). Then, the properties of the surface of the gold nanorods were measured by FT-IR spectroscopy using the MagnaIR -550 model of FT-IR, manufactured by Nicolet Corporation, USA. The structure of synthesized gold nanorods was recorded using a XRD diffraction pattern, which was recorded using the X-ray diffraction device model X' Pert Pro MPD manufactured by Philips Corporation. Also, to investigate the morphology of the synthesized gold nanostructures, Hitachi 2010's TEM has been used. The samples were deposited on copper networks for analysis of TEM.

Measuring the Thermal Conductivity Coefficient

In this study, the thermal conductivity of nanofluids was measured using the KD2 thermal analysis device based on the principles of the transient hot wire method (Decagon, online). In this device, a platinum wire with a diameter of 25 micrometers operates both as a heat sink and a thermometer. For induction of heat input (q), the thermal conductivity coefficient (k) is calculated from Equation 3 (this equation is derived from a solution of the appropriate solution of a Fourier-Kirchoff transient heat conduction problem solution in cylidrical coordinates. The erroneous interpretation is copied from PAUL 2011):

$$k = \left[\frac{q}{4\pi(T_2 - T_1)}\right] \ln\left(\frac{t_2}{t_1}\right) \tag{3}$$

In this equation T_1 and T_2 are, respectively, the temperatures at t_1 and t_2 times. To normalize the possible variation due to human and instrumental errors, the data is expressed as the ratio between the thermal conductivity of the nanofluid with respect to that of the base fluid.

Discussion and Results

Identification of properties of nanofluids based on gold nanorods

Figure 1 shows the XRD pattern of gold particles prepared by the growth method using seeds. The X-ray source used was Cu-Kα radiation at 40 kV and 20 mA, and diffraction was analyzed using the X-ray diffraction device model X' Pert Pro MPD. All the conventional Bragg reflections of this face centered cubic (FCC) metal exist. The amount of the fairly wide peaks is evidence of the crystalline nature of gold nanorods in a nanometer range (ZHANG et al. 2009, JIA et al. 2014). The results of the FT-IR spectroscopy (Fig. 2) show that the surface of the prepared nanorods is completely coated with CTAB (GENTILI et al. 2009).

Figure 3 shows the spectrum of UV-vis nanofluids of gold nanorods diluted in water at a volume ratio of 1 to 50. CTAB absorption peaks and deionized water are indistinguishable and can be ignored. Thus, we can see have two absorption peaks in each nanofluid of gold nanorods: One is in the range of 520 nm and

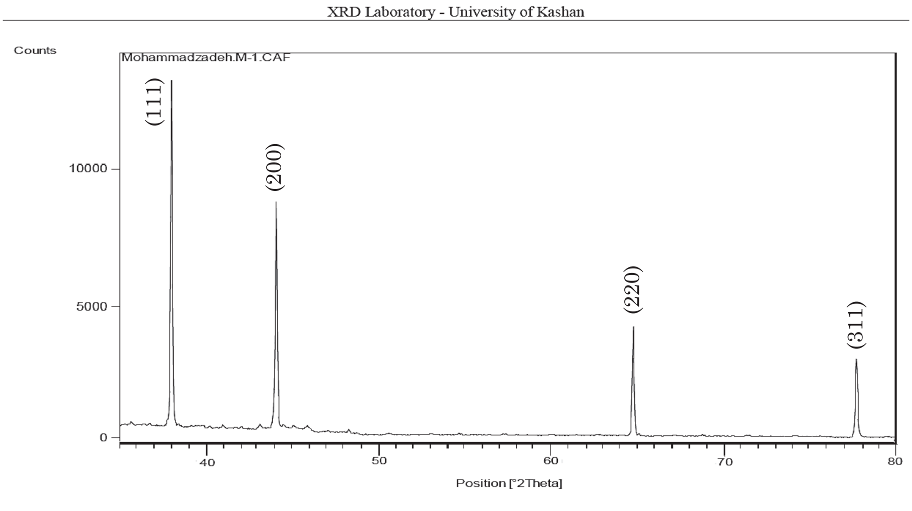

Fig. 1. Gold Nanorods XRD Pattern

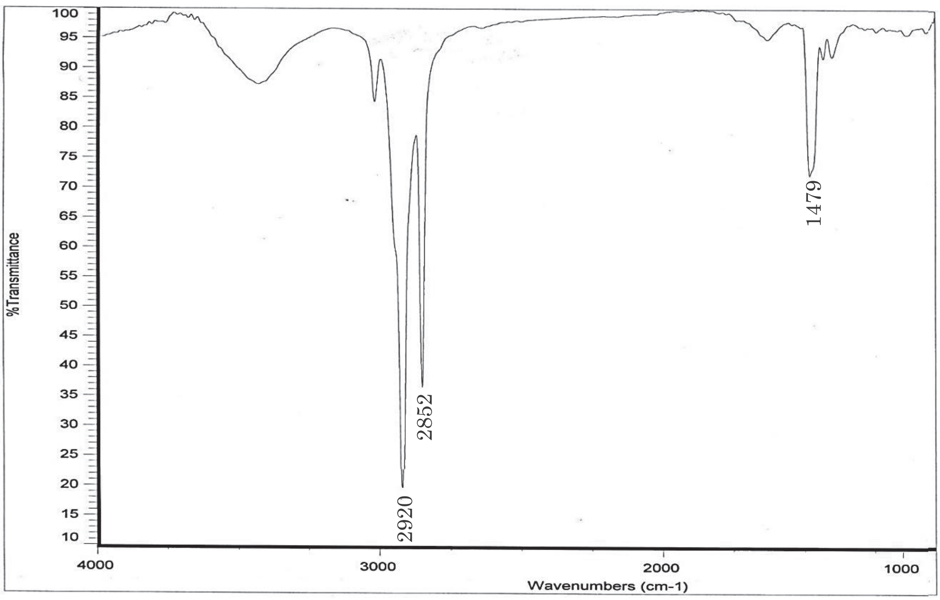

Fig. 2. FT-IR Spectrum of Synthesized Gold Nanorods Using Growth Method of Seeds

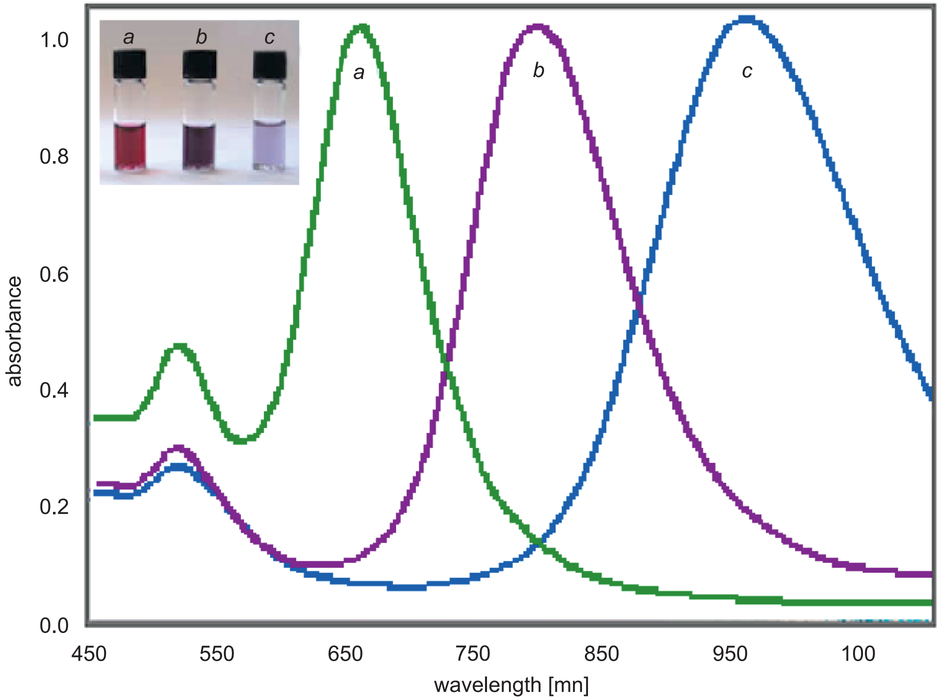

Fig. 3. The UV-vis spectrum of gold nanorods diluted in water with a ratio of 1 to 50 (1/50)

the other occurred at a higher wavelength depending on the aspect ratio of nanorods (BURDA et al. 2005). The longitudinal resonance wavelength in each sample of gold nanorods is 660 nm, 780 nm and 950 nm, respectively, for short, medium and high nanorods. Also, Figure 4 shows the transmitted electron microscopy (TEM) images of the prepared samples, confirming the results obtained from the absorption spectra of the samples and samples are called "short", "medium", and "high" based on their aspect ratios. From the LSPR absorption peak (Fig. 3),

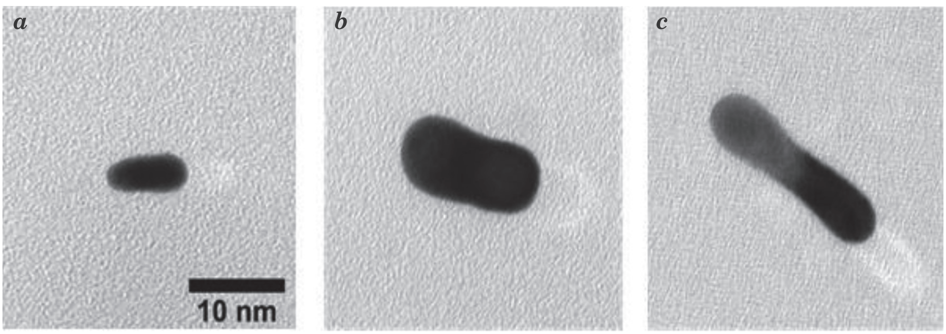

Fig. 4. TEM images of gold nanorods samples: *a* – "short", *b* – "medium", and *c* – "high"

we can obtain comprehensive information of the prepared gold nanorods. Because of the calculations done DRAINE and FLATAU (1994), using a discrete dipole approximation (DDA), an exact relationship is established between the longitudinal LSPR peak wavelength and the aspect ratio of the gold nanorods (equation 4). Therefore, we can calculate even the nanorods aspect ratio with the UV-vis spectrum.

$$\mathrm{AR} = \frac{\text{peak position [nm]}}{99.3} - 4.6 \tag{4}$$

In this regard, by determining the longitudinal LSPR peak wavelength, the nanorods aspect ratio (AR) is calculated. It should be noted that there is a linear relationship between the peak position and the nanorods aspect ratio in other works (NIKOOBAKHT, El-Saye 2003, JAIN et al. 2012). In this equation the least squares method for the data set was used to declare the aspect ratio and position of the corresponding peak, so that the shown linear equation was gained. The aspect ratio of the synthesized gold nanorods with regards to the longitudinal LSPR peak wavelength is given in Table 1.

Table 1

The average aspect ratio of prepared gold nanorods

Resonance [nm]	Aspect Ratio
660	2.1
780	3.2
950	4.9

Thermal conductivity of nanofluids

In this research, the nanofluids' thermal conductivity coefficient of gold/water nanorods was investigated in different aspect ratios, volume fractions and temperatures. Figure 5 shows variations in the thermal conductivity of the nanofluid in different aspect ratios of the gold nanorods. As shown in the figure, the thermal conductivity of the nanofluid decreases with the increase in the nanorods' aspect ratio in a 1:50 constant volume fraction of gold nanorods in water at room temperature. Many studies have shown that rod-shaped nanoparticles impact on the nanofluid effective thermal conductivity is greater than spherical nanoparticles, due to the larger aspect ratio and the larger ratio of surface area of the particle to its volume (LEI et al. 2015). But according to the results, the smaller the aspect ratio of the gold nanorods, the higher the thermal conductivity, which is due to their Brownian motion (CHOPKAR et al. 2008). On the contrary, another model (PATEL et al. 2008), taking into account

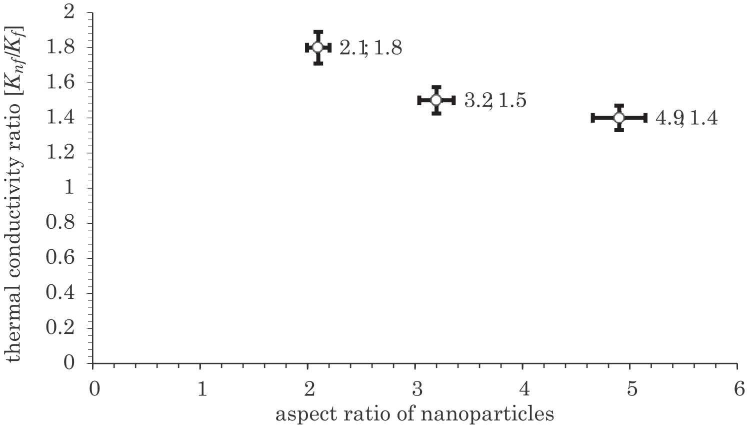

Fig. 5. Nanofluid thermal conductivity coefficient of gold/water Nano rod in various aspect ratios (volume fraction: 1:50, temperature: 25°C)

the combined effects of high specific surface area of the particles, liquid layering at the solid–liquid interface and convective heat transfer enhancement associated with the Brownian motion of the particles quite accurately predicts the thermal conductivity enhancement of the present experimental data as a function of both concentration and particle size. Thus a cumulative effect of several mechanisms such as liquid layering, high nanoparticle specific surface area, and Brownian motion of the particles can be considered to be the possible contributing factors for the phenomenal enhancement of thermal conductivity in nanofluids. Also, smaller nanoparticles have high resistance to sedimentation and precipitation, which is one of the greatest technical challenges in nanofluids (PRASHER et al. 2006). It may be pointed out that the maximum uncertainty limit of thermal conductivity data recorded by the hot-wire device is ±5%, which is consistent with the error bar for the experimental data on thermal conductivity enhancement observed for nano-gold dispersed water based nanofluids (Figs. 5, 6, 7).

Figure 6 shows an increase in the thermal conductivity of the nanofluid with the increase of the volume fraction of the gold nanorods in the base water, with a ratio of 2.1 and in room temperature. Probably, the fundamental factor in increasing thermal conductivity is the degree of dispersion of nanoparticles in the base fluid. In spite of the dramatic increase in thermal conductivity due to the increase of the volume fraction, it should be noted that the volumetric fraction of nanorods in the base fluid is high and it is not appropriate to call the Nano fluid the Nano suspension. Also, this will result in a pressure drop in the flow of fluid. For this reason, it is more common to use the nanofluid in lower volume fractions (KARTHIKEYAN et al. 2008).

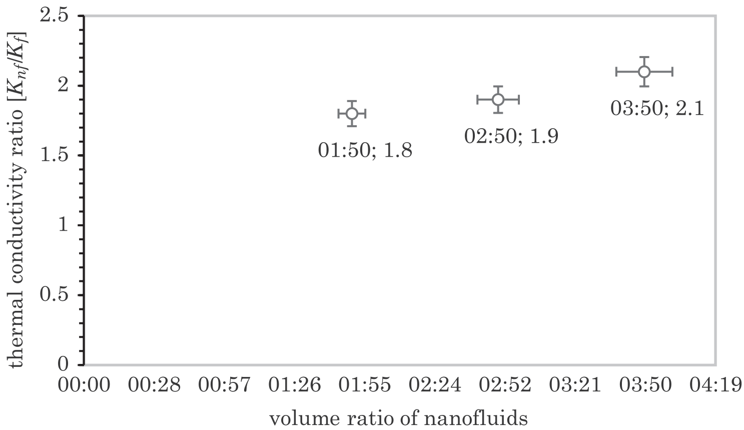

Fig. 6. Nanofluid thermal conductivity coefficient of gold/water nanorods in different volume fractions (aspect ratio: 2.1, temperature: 25°C)

Also, the increase in the thermal conductivity coefficient of nanofluid at various temperatures is shown in Figure 7. The thermal conductivity of the nanofluid is increased by increasing their Brownian motion with increasing temperature, while the thermal conductivity of the fluid without nanoparticles does not change with temperature change (TAHA-TIJERINA et al. 2012). On the contrary, it changes for about 10% with the temperature increase of about 50 C degree from the room temperature (cf. ROHSENOW et al. 1998, table 2.16). An increase of 66% was observed in thermal conductivity at 55°C with a 1:50 volume fraction of nanorods with an aspect ratio of 2.1, relative to the base fluid.

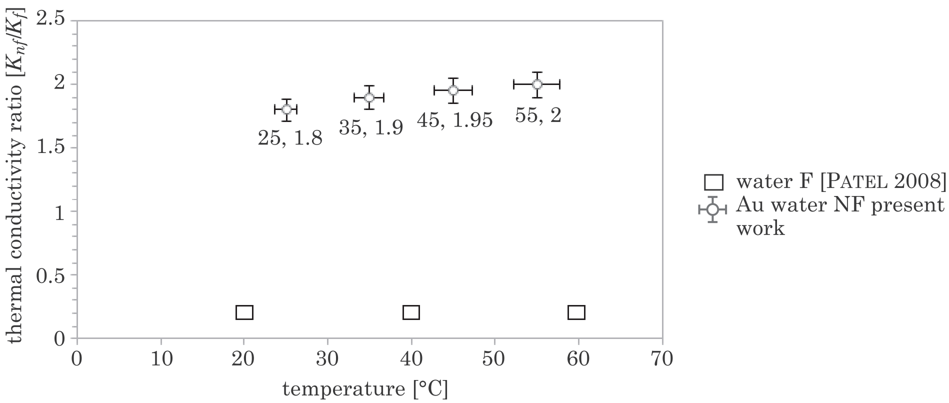

Fig. 7. The effect of temperature on the thermal conductivity of nanofluids in gold/water nanorods (aspect ratio: 2.1, volume fraction: 1:50)

Among the limited number of studies reported in the literature on thermal conductivity of pure gold dispersed water based nanofluids, JHA and RAMPRABHU (2009) have recently reported about 28% enhancement in thermal conductivity for gold and carbon-nanotube (multi wall) composite nanoparticle dispersed water based nanofluids as compared to only 15% enhancement for similar water based nanofluid with only carbon nanotube dispersion. Since thermal diffusivity is directly proportional to thermal conductivity for the same component or system, it is logical to anticipate thermal conductivity may also follow a similar trend in case of gold nanoparticle dispersed nanofluids. Among the proposed mechanisms for increasing the thermal conductivity of nanofluids, the presence of a nanolayer of fluid molecules in a solid-liquid joint-phase is one of the strong ideas put forth by researchers all over the world (YU, CHOI 2003, 2004, YAN et al. 2007, XIE et al. 2005, TILLMAN, HILL 2006, MURSHED et al. 2008). According to the studies and theories proposed by YU and CHUI (2003), XIE et al. (2005) and TILLMAN and HILL (2006), it can be concluded that the presence of a liquid layer in the solid-liquid interface cannot singly increase the thermal conductivity of nanofluids. As a result, in addition to increasing the surface-to-volume ratio of nanofluids based on gold nanorods and increasing the nanolayer formed on the solid-liquid interface along with the increase of Brownian motion of particles, increases the thermal conductivity, which can be considered as a function of the aspect ratio, volume fraction and temperature. Therefore, a theory consisting of several mechanisms, such as liquid layering in the solid-liquid interface, the high surface-to-volume ratio of nanoparticles, and the Brownian motion of particles, can be considered as effective factors in increasing the thermal conductivity of nanofluids.

Conclusion

In this study, nanofluids based on gold nanorods were prepared by a one-stage seed-mediated growth method. Then, the nanofluid thermal conductivity coefficient of gold/water nanorods was investigated experimentally in different aspect ratios, volume fractions and temperatures. In order to determine the thermal conductivity coefficient, the KD2 transient hot wire method was used. The results show that an increase in the nanorods aspect ratio with a constant volume fraction of 1:50 of gold in water nanorod and at room temperature leads to a decrease in the thermal conductivity of the nanofluid, while in general, the increase in this coefficient in the Nano fluid based on Nano rod is more than nanofluid based on measured spherical gold nanoparticles. Also, increasing the two parameters of volume fraction and temperature significantly increases the thermal conductivity coefficient. On the other hand, based on the laboratory findings, new and useful models for estimating the thermal conductivity of the nanofluid were presented. And a theory consisting of several mechanisms, such as liquid layering in the solid-liquid interface, the high surface-to-volume ratio of nanoparticles, and the Brownian motion of particles, can be considered as effective factors for increasing thermal conductivity in nanofluids.

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