

Investigation of Heat Transfer Coefficient of Ethylene Glycol/ Graphenenanofluid in Turbulent Flow Regime

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Abstract:

In the present work, graphene was synthesized by chemical vapor deposition (CVD) method. The structure of graphene was then confirmed by X-Ray diffraction (XRD) and scanning electron microscope (TEM) images. After that, mixed acid method (H_2SO_4/HNO_3) was used to make the structure of synthesized graphenehydrophilic. In this method, carboxylate and hydroxide groups were linked to the edges of graphenenano sheets. The hydrophilic graphene was added to ethylene glycol (EG) with the concentrations of 0.100, 0.125, and 0.150 wt%, and the mixtures were sonicated at 60°C for 3hours to prepare the ethylene glycol/ graphenenano fluid. The thermal conductivity of samples was measured by KD2, while thermophysical properties of were estimated by valid theoretical equations. Then heat transfer coefficient of the samples was measured by using a straight pipe under constant heat flux and a turbulent flow regime. According to the obtained results, thermal conductivity and heat transfer coefficient increased by 21.2% and 42.4%, respectively, only by the addition of 0.15wt % of graphene in to ethylene glycol. Also thermal conductivity and heat transfer coefficient were improved by increasing the temperature and concentration of graphene.

Keywords: Graphene, nanofluid, Ethylene glycol, Thermal conductivity, Heat transfer coefficient, Turbulent flow.

1. INTRODUCTION

Nanofluids (NFs) are stable suspensions, which are made by uniform dispersion of nanoparticles (NPs) in fluids [1]. The thermal conductivity of NFs is significantly higher than conventional fluids due to the stability, particle size, and higher conductivity [2]. Therefore, application of NFs in heat exchangers is so suitable [3]. In chemical processes, one of the most important devices related to energy and heat transfer is heat exchanger. The poor heat transfer properties of employed fluids (such as water, mineral oil, and

ethylene glycol (EG)) are obstacles for using different types of heat exchangers. Efforts have been done to enhance heat transfer, reduce the heat transfer time, minimize size of heat exchangers, finally increase energy and fuel efficiencies. EG has been used widely as the cooling fluid and anti-freezing agent in heat exchangers and thus improving its thermal properties that are of a great importance [4]. Since nano particles (metallic, nonmetallic, and carbon structures) have thermal conductivity higher than that of fluids, when they

are dispersed in the fluids (NFs) result in higher heat transfer characteristics and modify heat transfer performance of fluids.

Graphene, a sheet with thickness of one carbon atom, is made from carbon atoms in a hexagonal lattice structure. The thermal conductivity of graphene is about 5000 W/mK, which is much higher than that of diamond (1800 W/mK) and carbon nanotubes (CNT) (3000 W/mK) [5]. All experimental results have indicated the enhancement of thermal conductivity by addition of NPs [6-14] also there are several published studies on the forced convective heat transfer coefficient of NFs, and most of them are under the constant heat flux or constant temperature boundary conditions at wall of tubes and channels. The experimental results for forced convection inside a channel show that convective heat transfer coefficient of NFs is enhanced compared to base fluid [15-21]. The objective of the present study is to investigate the heat transfer characteristics (such as overall and convective heat transfer coefficients, and Nusselt number) graphene /EG NFs for turbulent flow in a horizontal stainless steel shell and tube heat exchanger.

2. EXPERIMENT

2.1. Synthesis and Characterization of Graphene

In this research, graphene was grown over copper foils by the CVD method. A mixture of methane and hydrogen gas was used under atmospheric pressure as the feed. First, copper foils were inserted inside the reactor, and the reactor was heated up to 1050°C by using hydrogen flow with flowrate of 910 sccm. After 40 min, methane was introduced to the system with flowrate of 335 sccm, and hydrogen flow was decreased to 665 sccm. The furnace, which applied to synthesize of graphene, is represented in Figure 1.

As a result of methane decomposition, hexagonal structures of graphene were grown over copper foils. The reactor was cooled after 15 min with temperature slop of 180°C/min [22]. After cooling procedure, purification treatment was performed. X-ray diffraction analysis of

synthesized graphene sheets has been represented in Figure 2.



Figure 1. The furnace which applied to synthesize of graphene

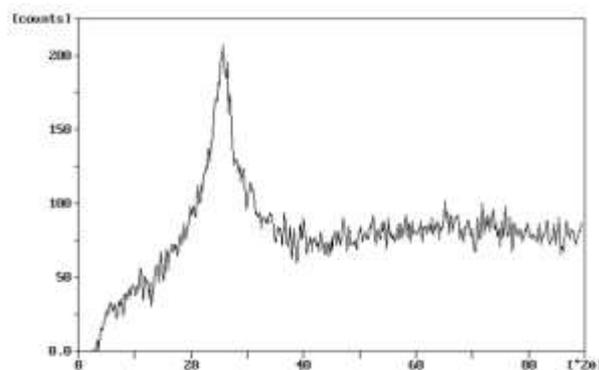


Figure 2. XRD of graphene synthesized by CVD method

According to this figure, there is a short and wide peak at 26.5°, which is consistent with the data in the literature [23]. It is known that copper has a peak at 46° and rolling of carbon nanotubes is attributed to 42° and 76.8° [24]. As it can be seen from the figure, there is not any copper in the system, and the graphene sheets have not been rolled. Therefore, it can be concluded that the obtained graphene is pure. By using Scherrer equation [25], the crystalline size of graphene is calculated as 2.1 nm, and when it is divided by the distance of graphene layers (3.4 Å) [26], the number of graphene sheets is calculated as 6.1 layers [23].

For studying the morphology of graphene, TEM analysis was used (Figure 3). As indicated from this figure, the synthesized graphene has a disordered structure similar to a folded paper, which is attributed to the nature of pure graphene and its natural stability [27].



Figure 3. TEM image of graphene synthesized by CVD method

2.2. Hydrophilic of graphene and nanofluid preparation

Since the synthesized graphene is hydrophobic, it precipitates rapidly in polar fluids. Thus for increasing the stability, graphene is converted to hydrophilic graphene oxide by using an acidic method (a mixture of H_2SO_4 and HNO_3 with the ratio of 3–1 for 3 hours at 60 °C) [28]. For checking the presence of carboxylic groups over graphene after acidic oxidation, FTIR spectra of GO sample was compared with that of graphene sample.

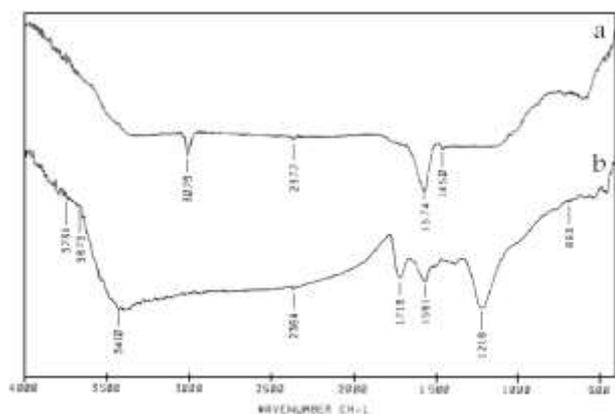


Figure 4. FTIR spectra of a) Graphene b) Graphene oxide

As shown in part b of Figure 4, peaks at 1218 cm^{-1} and 3410 cm^{-1} are attributed to C-O bond and hydroxide group (OH), confirming the presence of carboxylic group (-COOH) [29]. The existence of peak at 1717 cm^{-1} indicates that during the oxidation of graphene, some C=O group are bonded to the edges of graphene sheets [30] and the shift from 1735 to 1718 cm^{-1} reveals high number of hydroxide groups which are bonded to graphene [31]. Also the shift of 1574 cm^{-1} which is attributed to C=O bond, to the right hand side of 1581 cm^{-1} indicates the presence of carboxylate group over graphene structure [32] and as a result it can be concluded that a good oxidation has been performed. After making graphene sheets, hydrophilic nanofluid samples (GO/EG) were prepared. Graphene powder was added with volumetric percentages of 0.05, 0.075, and 9.1% to EG as the base fluid and the mixture was sonicated for 45 min at ambient temperature. Characteristics of samples have been represented in Table 1.

Table 1. Profile samples Nano fluids graphene oxide / ethylene glycol

No	Sample	Grapheneoxide (wt. %)
1	EG/ARG-1	0.100
2	EG/ARG-2	0.125
3	EG/ARG-3	0.150

2.3. Calibration of the system

The experimental set up includes test section, pump and fluid cycling system, a shell and tube heat exchanger, and a circulator. The test section consists of a thin and smooth copper pipe with length of 1 meter, and it is covered by electrical element for providing constant heat flux. Thermal insulator has been used over the element for isolating the test section from the surrounding. Five thermocouples were used with equal distances along the copper pipe for measuring the wall temperatures and two additional thermocouples were used for measuring the fluid temperature in inlet and outlet sections.

The working fluid is pumped into the system and after heating inside the copper pipe, it flows through the heat exchanger. The exchanger is connected to the circulator, and thus, it makes the

temperature of the fluid to decrease. After that, the fluid is collected in a reservoir for the removal of its disturbance, and it is then pumped again into the system, and the cycle is repeated. Figure 5 represents the schematics of the experimental setup.



Figure 5. Experimental setup of shell & tube heat exchanger

Calibration of the system was performed by calculating one of base fluid properties (EG). The aim of calibration of the system is to ensure the correctness of the results, repeatability of the tests, and error evaluation. Therefore, Nusselt (Nu) number was measured and was compared with the theoretical relations [27]. EG was flowed inside the system by a flowrate of 0.23lit/min and Reynolds (Re) number of 2840. The element created a heat flux of 352.1W over the outlet surface of copper pipe. After some time, the steady state was reached and the inlet and outlet temperatures were recorded. Thermophysical properties of EG was calculated from standard tables at average temperature ($\bar{T}_f = (T_{in} + T_{out})/2$). The results are represented in Table 2 [33].

The heat transferred from hot wall to the fluid was calculated from $q_2 = \dot{m} \cdot C_p (T_{out} - T_{in})$; $\dot{m} = \rho \cdot \dot{Q}$ as 339.4W, which is almost the same as the heat flux produced by the element. The constant flux is calculated from $q'' = \bar{q} / \pi D_o L$ as $9368.7W/m^2$. The average temperatures of wall and fluids were calculated from $\bar{T}_w = (\sum_{i=1}^5 T_{w_i})/5$; $\bar{T}_f = (T_{in} + T_{out})/2$.

The calculated values for average wall and

fluid temperatures are as 35.7 and 32.2°C. The average heat transfer coefficient is calculated from $\bar{h} = q'' / (\bar{T}_w - \bar{T}_f)$ as $2602.4W/m^2K$ and the Nu number is obtained as 103.51. In turbulent flow inside pipe, if $4.4 * Re^{1/6} < L/D$, then the flow is fully developed [34], and the Nu number is calculated from below equation [35] as $Nu = 0.023 Re^{0.8} * Pr^{0.44}$. By replacing the values in this formula, the theoretical Nu number is calculated as 115.35 which is just 10.2% different from experimental value. This difference is acceptable as a low error, and it confirms that the system is calibrated in turbulent flow regime.

3. RESULTS AND DISCUSSION

3.1. Calculation of thermophysical properties of EG/ graphene nanofluid

The thermophysical properties of nanofluids including the density, viscosity, and heat capacity are usually different from the base fluid, and they have an important impact in heat transfer coefficient. These properties are used in the measurement of heat transfer coefficient and therefore they are assessed before calculating the heat transfer coefficient. For evaluation of thermophysical properties of nanofluids usually theoretical relations are used which are based on the single phase fluid. This assumption is correct by reducing the concentration of nanoparticles and due to very low concentration of graphene in this work, it can be used. The viscosity of suspension with concentrations lower than 4% can be obtained from Drew and Passma relationship: $\mu_{nf} = \mu_f (1 + 2.5\phi)$ [36]. The density of stable nanofluid can be obtained by Pak & Cho formula: $\rho_{nf} = \phi \rho_p + \rho_f (1 - \phi)$ [37]. Also the average heat capacity of nanofluids can be obtained from Xuan & Roetzel equation: $(\rho C_p)_{nf} = \phi (\rho C_p)_p + (1 - \phi) (\rho C_p)_f$ [38]. The thermal conductivity of nanofluids were measured by hot-wire method by using KD2 [39]. The thermophysical properties of samples have been represented in Table 3.

Table 2. Thermo physical properties of EG- 32.2°C

Thermal conductivity(W/m.K)	Specific Heat(J/Kg.K)	Viscosity (Kg/m.s)	Density (Kg/m ³)
0.269	2391.4	0.0152	1110.1

Table 3. Thermo physical properties of EG/G NFs- 32.2°C

sample	Graphene (wt%)	Thermal conductivity(W/m.K)	Density (Kg/m ³)	Viscosity (cp)	Specific Heat(J/Kg.K)
EG	0	0.253	1109.1	14.51	2427.7
Graphene	1	5000	2200	-	790.1
EG/ARG-1	0.1	0.312	1218.2	18.14	2131.9
EG/ARG-2	0.125	0.324	1250.3	19.24	2051.8
EG/ARG-3	0.15	0.338	1283.9	20.21	1969.5

According to this table, it can be concluded that the presence of graphene in EG leads to an increase in density, viscosity and thermal conductivity, and a decrease in specific heat capacity. Moreover, density and viscosity of nanofluids are enhanced by increasing the graphene concentration, while the heat capacity decreases. For maximum concentration of graphene (0.15 wt%), the density and viscosity of base fluid increased by 15.76% and 39.28%, respectively, and the heat capacity decreased by 18.9%.

3.2. Measurement of local heat transfer coefficient for EG/graphene nanofluid in turbulent flow

Since the viscosity of graphene nanofluids is higher than EG, for making a turbulent flow under a constant Reynolds number, it is necessary to use higher flowrates of nanofluid in comparison with EG. For comparison, at first a constant Re number was set for both of EG fluid and EG/graphenenanofluid. This was reached by changing the flowrate of fluid. By setting the flowrate of samples, a constant Re number of 2840 under constant heat flux of 352.1 W was reached.

3.2.1. Measurement of average heat transfer coefficient of EG/graphenenanofluid in turbulent flow

Since it is common to use average heat transfer coefficient for designing heat exchangers, evaluation of this coefficient would be so useful. The experimental values of average heat transfer coefficient for EG/graphenenanofluid at different temperatures are represented in Table 4.

According to Table 4, it can be seen that by

increasing the temperature and concentration, the average heat transfer coefficient improves. For instance at average temperature of 30°C, just by adding 0.1 wt% of graphene, heat transfer coefficient increases by 27.9% in turbulent flow. One reason of such increase can be related to the 21.2% increase in thermal conductivity of nanofluid. Furthermore, it can be observed that an increase in temperature at higher concentrations of graphene in EG, enhances the heat transfer rate, because at graphene concentrations of three times of initial concentration. As a result, the trend of heat transfer coefficient enhancement in EG/graphene nanofluid will be a function of conditions, and it will increase by temperature increase. The maximum heat transfer coefficient is related to sample EG/ARG-3 with 2990.8W/m²K at 40°C.

Moreover as it can be seen from Figure 6, although the trend of heat transfer coefficient enhancement is similar to EG, but the enhancement rate increases by graphene concentration. It should be noted that the presence of graphene in EG has more effect in enhancement of thermal conductivity in comparison with heat transfer coefficient, and this effect increases by increasing graphene concentration. Also since the distance between curves increases by temperature, thus temperature has more significant effect in heat transfer enhancement, which makes the application of EG/graphenenanofluid more interesting for heat exchangers for cooling fluids with higher temperatures, manufactured nanoparticles, automotive, electronic, micro scale fluidic, biomedical and photo catalysts.

Table 4. Heat transfer coefficient of EG/graphenenanofluid- $Re=2840$

T_{ave} (°C)	Average heat transfer coefficient ($W/m^2.K$)				Improvement than EG(%)		
	EG	EG/ARG-1	EG/ARG-2	EG/ARG-3	EG/ARG-1	EG/ARG-2	EG/ARG-3
30	1608.4	2058.8	2190.6	2290.4	28	36.2	42.4
35	1723.2	2321.2	2407.3	2503.8	34.7	39.7	45.3
40	2033.3	2631.1	2871.0	2991.0	29.4	41.2	47.1

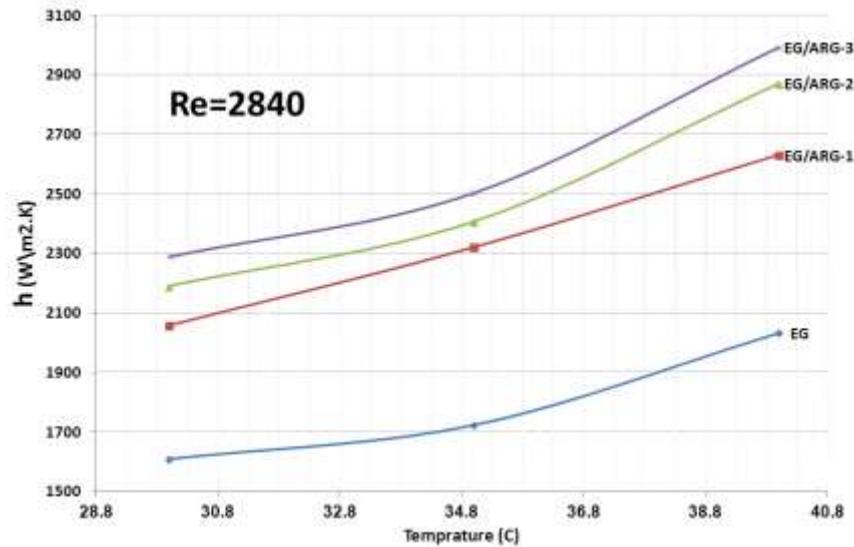


Figure 6. Heat transfer coefficient of EG/graphenenanofluid in various temperature

4. CONCLUSION

In this research, heat exchange coefficient of EG/graphenenanofluid was investigated in fully developed region of turbulent flow. Some thermophysical properties of EG/graphenenanofluid as well as heat transfer coefficient was measured at different concentrations. Thermal conductivities of graphenenanofluid were measured by KD2 at three concentrations of graphene. Also it was observed that at lower temperatures, the presence of graphene has more effect in thermal conductivity in comparison with heat transfer coefficient, while by increasing temperature, increase in heat transfer coefficient is higher than thermal conductivity enhancement. For instance by adding 0.1wt % graphene to EG at 30°C, thermal conductivity and heat transfer coefficient improve by 21.2 and 42.4%, respectively. Also increasing graphene

concentration affects some thermophysical properties of EG (enhancement of density, viscosity, and reduction of heat capacity), but generally it improves the thermal behavior of nanofluids. Heat transfer coefficient of graphenenanofluid in turbulent flow and under constant heat flux is an ascendant function of concentration and temperature.

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