

Thermal Conductivity Enhancement of Graphene Oxide Nanofluid Using an Improved Synthesis Method for Heat Transfer Applications

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Abstract: In this research work, graphene oxide (GO) nanomaterial was synthesized by using an improved synthesis method and deionized (DI) water is used as base fluid. The X-ray diffraction (XRD) analysis, Raman spectroscopy, scanning electron microscope (SEM) imaging, and Ultraviolet-visible (UV-Vis) spectroscopy were used to study characterization and morphology structure of GO. The three different nanofluid volumetric concentrations of 0.001%, 0.002% and 0.003% were prepared and the stability of nanofluid samples was evaluated by the zeta potential analysis method. The zeta potential analysis results revealed that the prepared nanofluid samples were stable for up to 30 days with no obvious sedimentation. This study experimentally investigates the effects of temperature variation and different nanofluid samples on the thermal conductivity GO/DI nanofluid. The findings of experimental study demonstrated that the thermal conductivity of GO/DI nanofluid increases with increase in the temperature and nanofluid concentrations. The maximum increment in thermal conductivity of nanofluid was obtained to be 25.15% at 40^o C for 0.003% volumetric concentration as compared to water.

Keywords: Graphene oxide; Nanofluid; Stability; Thermal conductivity.

1. Introduction

In any heat transfer process, the heat transfer fluid plays a crucial role. Water is most commonly used as a heat transfer fluid because of its availability, high specific heat capacity, and cheapness. However, different types of oils and ethylene glycol-based mixtures are also used as heat transfer fluids for specific applications but for evaluating these heat transfer fluids the estimation of thermo physical characteristics such as thermal conductivity measurement, specific heat capacity gain and viscosity of fluid becomes significant. The enhancement in thermo physical properties leads to an increase in thermal performance [1]. The thermal conductivity of water as heat transfer fluid is very low as compared to metal and metal oxides [2] but this can be improved by the addition of nano-sized particles (below 100 nm in size) into water [3]. The increase in thermal conductivity of nanofluid enhances thermal performance especially in solar collectors [4]. Therefore, most of the researchers have focused on finding nanofluid's thermal conductivity. The experimental work on Cu-H₂O nanofluid is carried out by [5] evaluates that the nanofluid's thermal conductivity depends on the weight concentration of nanoparticles, pH values, and sodium dodecyl benzene sulphonate (SDBS) concentrations. In this research work, the thermal conductivity of nanofluid was enhanced up to 10.7% at a weight fraction of 0.1%. The diamond has inherent characteristics of high thermal transport [6] which enhances the thermal conductivity of diamond nanofluid (DNF) in

water as base fluid up to 18% at a volume fraction of 0.02%.

In recent years, usage of carbon-based materials has grown in research such as carbon nanotubes (CNTs), graphene, graphene oxide, and graphene nanosheet. These carbon-based materials have higher thermal conductivity as compared to other nanoparticles due to low density and strong C-C covalent bonds [7]. The study of adjustable thermal conductivity in multi walled carbon nanotubes (MWCNTs) reported by [8] explored that the ratio of thermal conductivity enhances with an increase in volumetric concentration and temperature. The experiment carried out by [9] on graphite nanoparticles revealed that the maximum increment in thermal conductivity ratio is 34% at 2.0% volumetric concentration. The experimentation presented on CNTs, with polyalphaolefin (PAO) oil as base fluid showed that there is an enhancement of 161% in thermal conductivity by using CNTs-PAO oil mixture [10]. This result indicates carbon nanoparticles can be used to increase thermal conductivity as compared to other nanoparticles. However, experiment performed by [11] on CNTs like single walled, double walled and multi walled in water as base fluid identifies the significant effects on thermal conductivity improvement. The results showed that single walled nanotubes (SWNTs) have more thermal conductivity when compared to double walled nanotubes (DWNTs) and multi walled nanotubes (MWNTs). The carbon nanotubes with smaller diameters enhances thermal conductivity due to the liquid-solid interface and thermal conductivity for SWNTs with water is linearly dependent on temperatures lower than 40⁰ C as reported [12].

However, a stable mixture of graphene oxide and ethylene glycol was prepared [13] which remains stable for seven days, and the thermal conductivity enhanced up to 61% at 5.0% volume concentration. The research work on finding the effect of functionalization over graphene's thermal conductivity revealed that the thermal conductivity increment was more in the alkali functionalization method than in the acid functionalization method. The thermal conductivity ratio was increased to 24.4% at 20⁰ C and 39.9% at 60⁰ C [14]. The greatest increase in heat conductivity was found to be 33% at a composite weight concentration of 0.1% by using TiO₂ over reduced graphene oxide as reported in [15].

It is important to note that, graphene has a higher thermal conductivity of 5000 W/mk and carbon nanotube has 3000 W/mk [16] which attracted researchers to use graphene-based material in the heat transfer process. Hence in this work graphene oxide is used as a nanomaterial. The graphene oxide was synthesized using an improved synthesis method because it does not produce toxic gases.

2. Experimental methods

2.1 Nanofluid preparation

In this research work, the graphene oxide is prepared in the laboratory with an improved synthesis method. It consists of the insertion of graphite powder in the mixture of Sulphuric acid (H₂SO₄) and Phosphoric acid (H₃PO₄). The mixture is then stirred with the addition of 18 gm. of potassium permanganate (KMNO₄). After the reaction is cooled, it was poured onto the ice with hydrogen peroxide (H₂O₂) followed by a decantation process with DI water and Hydrogen chloride (HCl) to maintain the pH value of 7.

After the preparation of graphene oxide, the dry GO was directly dissolved in deionized (DI) water as the base fluid because graphene oxide is hydrophilic and has good water compatibility [17]. The three different volumetric concentrations of 0.001%, 0.002%, and 0.003% graphene oxide and DI water samples were prepared, and then it is sonicated for up to 1 hr. to prepare a stable solution. Figure 1 (a & b) shows prepared GO and prepared different samples of GO/DI water.

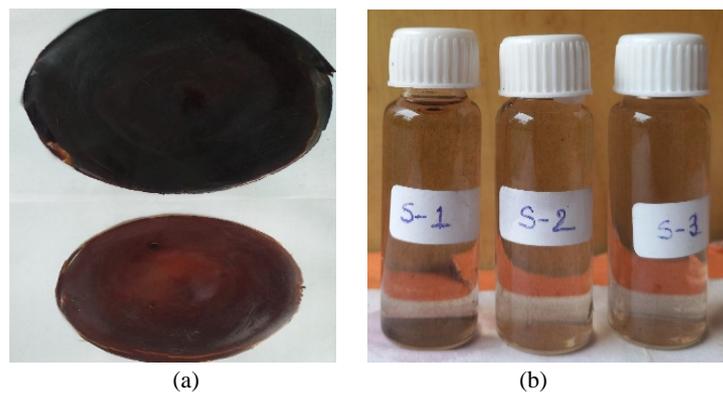


Figure 1- Preparation of GO a) GO sample, b) GO/DI samples

2.2 Characterization of GO

The prepared GO was characterized by using X-ray diffraction (Bruker D8 Advance) which was operated at 40 kV and 45 mA to study the crystalline structure of graphene oxide with a scanning speed of $20^\circ/\text{min}$. The Raman spectroscopy is also carried out by using Horiba Japan Xplora Plus which is a widely used technique to investigate and structurally characterize graphene-based materials [18]. The morphology structure of GO is examined with a scanning electron microscope image by using Hitachi, S 3400N Microscope. Finally, the optical analysis of GO is carried out by using UV-visible spectroscopy (LAMDA 750, perkin elmer, UV-vis NIR spectrophotometer) between 200 to 800/nm wavelength range.

2.3 Stability of GO/DI sample

The stability of nanofluid samples was evaluated by using zeta potential analysis (Malvern Instruments, UK). To know the stability period of samples, the zeta potential analysis is carried out just after the preparation of samples and after the 30 days of preparation of samples.

3. Results and discussion

3.1 Characterization techniques

The X-ray diffraction pattern of GO is presented in Figure 2, which shows purity and degree of oxidation [19]. A clear visible characteristic diffraction peak of 11.54° was observed at 2θ which was attributed to the crystalline plane of GO. It also shows that oxygen-based functional groups are present in between graphene nanosheets of GO [20].

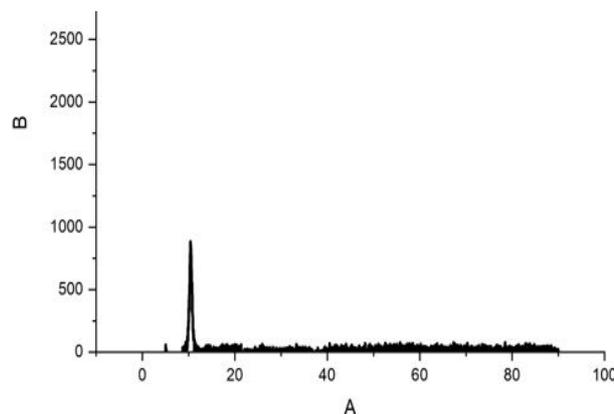


Figure 2- X-ray diffractogram of GO

In order to determine the thickness and structural flaws in GO, Raman spectroscopy was used. Figure 3 shows Raman spectra of GO. The D and G bands peak intensities are obtained at $1384/\text{cm}$ and $1581/\text{cm}$ respectively. The resulting peak of D

band is attributed to the edges, disordered carbon atoms, and various flaws whereas the G band obtained is attributed to the ordered sp^2 bonded carbon atoms. The ration of D band and G band is greater than one which shows that oxygen presence on the surface of GO [21].

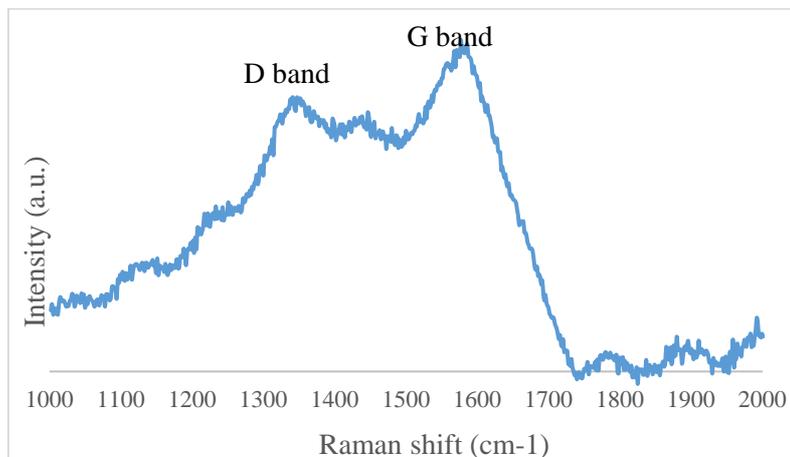


Figure 3- Raman spectroscopy graph of synthesized GO

The high magnification and resolution image are obtained by focusing the beam of electrons that scans the sample. The scanning electron microscopic image is shown in Figure 4 (a & b) which is used to study surface morphology [22] and particle characterization of GO. From SEM images it is clear that GO has two-dimensional structure and multiple lamellar layers. This layer formed are one above the other with wrinkled shape. The GO sheets have higher thickness at the edge because of oxygen containing functional groups.

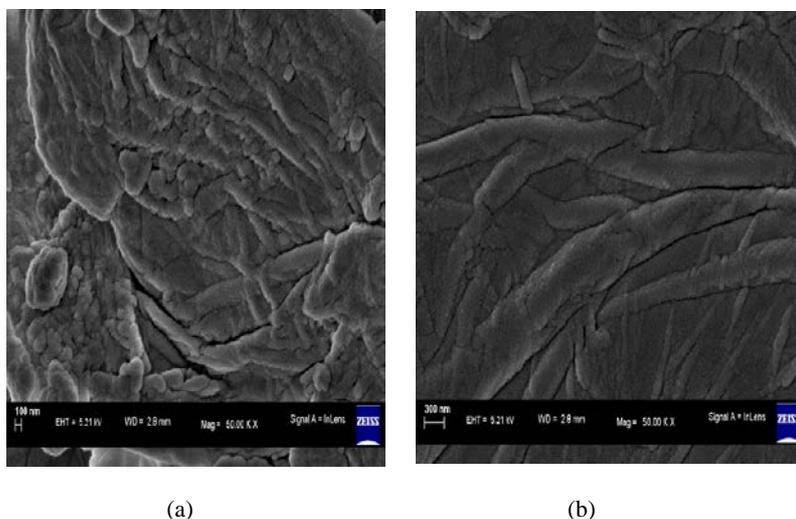


Figure 4- a, b) Scanning electron microscopic images

Figure 5 shows UV-visible absorption graph which is captured to study absorbance characteristics of GO. The primary absorption peak is visible in spectrum at a wavelength of 237 nm for GO which corresponds to functional and carbonyl groups. These carbonyl groups improve the water solubility and enhances dispersibility of GO into base fluid [23].

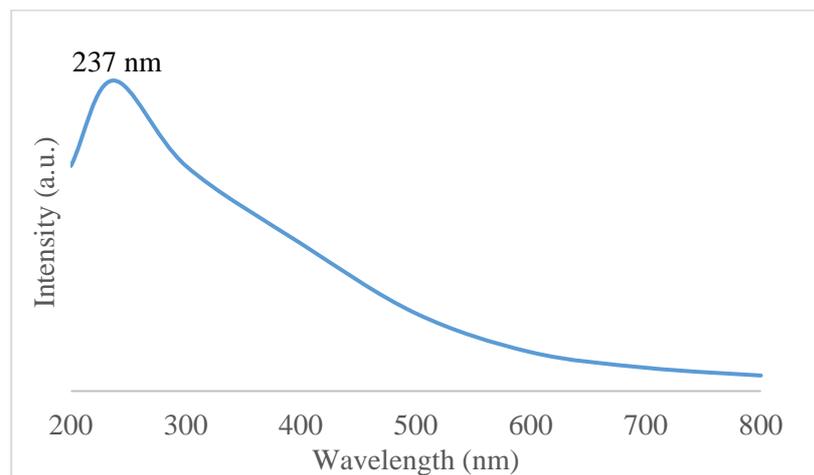


Figure 5- UV-visible spectrum of GO

3.2 Stability of nanofluid samples

To find the stability of prepared nanofluid samples, the prepared GO/DI samples were kept under observation for 30 days just after the preparation. After 30 days, samples were observed with no obvious sedimentation under atmospheric conditions. From Figure 6, it can be concluded that the prepared samples can remain stable for up to 30 days. Similarly, the zeta potential analysis is carried out just after the preparation of nanofluid samples and after 30 days of preparation. The results show that the prepared nanofluid samples have a zeta potential value of 39.3 mV at the time of preparation and it decreases up to 31.3 mV for a volumetric concentration of 0.003% which confirms that prepared nanofluid samples are stable [24].

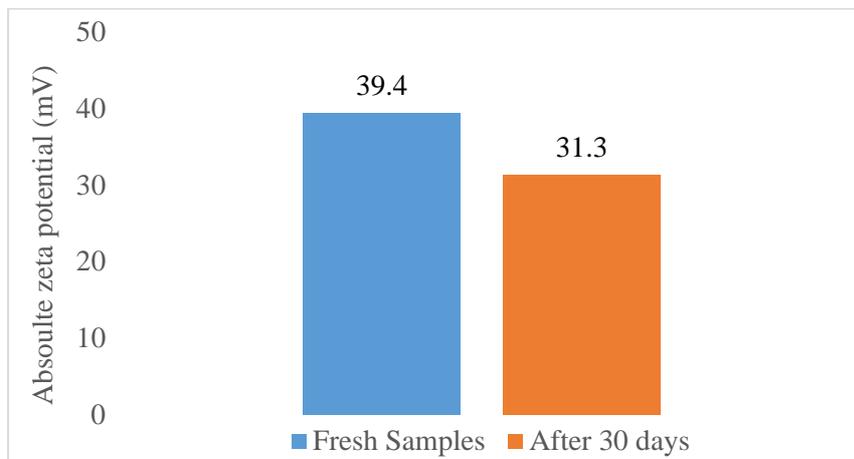


Figure 6- Zeta potential analysis for 0.003% vol. conc. sample

3.3 Thermal conductivity

There are several parameters that effects on thermal conductivity of GO nanofluids such as preparation process, base fluid type, size of nanoparticles, and temperature of medium [25]. The three different samples of 0.001%, 0.002%, and 0.003% volumetric concentrations for GO/DI water nanofluid were prepared and tested over the temperature range from 20⁰ C TO 40⁰C. The influence of volumetric concentrations and temperature variation on the thermal conductivity of GO/DI water nanofluid is discussed in this section.

3.3.1 Temperature variation effect

The influence of temperature variation on the thermal conductivity of nanofluid

samples was investigated and discussed in this section. According to Figure 7, as temperature increases, the thermal conductivity of each sample increases. For water, thermal conductivity values increase from 0.62 W/mk to 0.64 W/mk at a temperature of 20⁰ C and 40⁰ C respectively. The thermal conductivity enhances from 0.664 W/mk to 0.747 W/mk for volume concentration of 0.001% at 20⁰ C and 40⁰ C respectively. Further, increase in volumetric concentration from 0.002% to 0.003%, the thermal conductivity also increases.

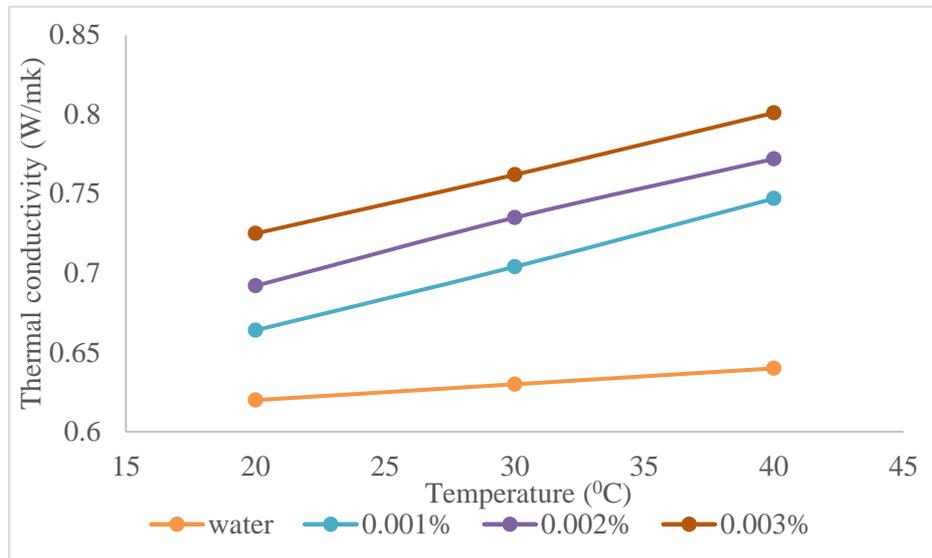


Figure 7- Thermal conductivity graph of GO/DI versus temperature

This improvement of thermal conductivity with temperature variation is due to the fact that, an increase in temperature, increases collision between nanoparticles in the fluid due to an increase of Brownian motion which can lead to having a higher thermal conductivity. Similarly, rise in temperature, increases collision between molecules of base liquid and nanoparticles and high thermal motion velocity which are attributed to the higher nanofluids thermal conductivity [26].

Table 1- Percentage enhancement of thermal conductivities in volume fraction.

Temp (°C)	S-1 0.001%	S-2 0.002%	S-3 0.003%
20 °C	7.09	11.61	16.93
30 °C	11.74	16.66	20.95
40 °C	16.71	20.62	25.15

From Table 1, it is concluded that the maximum enhancement in thermal conductivity is obtained to be 25.15% at 40⁰C for 0.003% volumetric concentration as compared to water.

3.3.2 Effect of particle concentration

Figure 8 indicates the effect of volumetric concentration on thermal conductivity of GO/DI water nanofluid. The results show that as volumetric concentration of nanofluid sample increases, the thermal conductivity also increases. This can be due to the fact that, the addition of high thermal conductive GO into the DI water increases the thermal conductivity of samples. Also, the increase in the volumetric concentration of nanofluid

samples means the insertion of more nanoparticles into a base fluid which increases the area of contact between particles and base fluid and improves the thermal conductivity of nanofluid samples [27].

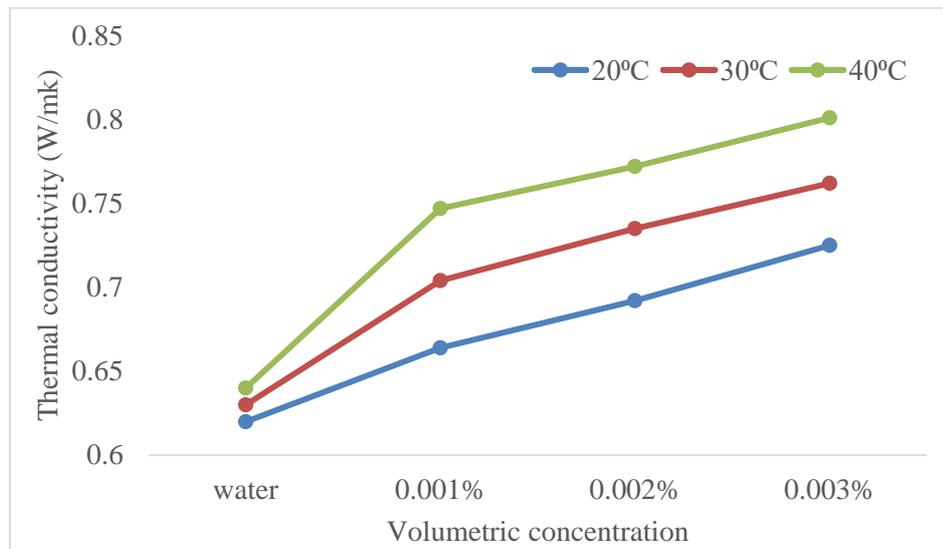


Figure 8- Thermal conductivity of GO/DI versus volume fraction

One more reason behind this behavior is that adding nanoparticles of graphene oxide into DI water forms a nanolayered structure which attracted liquid molecules to come close to the solid particle surface, and thermal conductivity increases.

4. Conclusions

In this study, graphene oxide and DI water were used as nanofluids with three different volumetric concentrations of 0.001%, 0.002%, and 0.003% samples. The GO was prepared in laboratory using an improved synthesis method and it is characterized by different techniques. The prepared nanofluid samples were found to be stable for up to 30 days without sedimentation which shows good stability of GO/DI water nanofluid. The nanofluid's thermal conductivity was measured in the temperature range of 20^o C to 40^o C. The results showed that there is a significant enhancement in the thermal conductivity of nanofluid as compared to water. The maximum enhancement in thermal conductivity was found to be 25.15% at 40^o C for a volumetric concentration of 0.003% as compared to water.

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