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# Synthesis, stability, transport properties, and surface wettability of reduced graphene oxide/water nanofluids

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ABSTRACT

In this study, reduced graphene oxide (rGO) is synthesized from graphite using modified Hummer and chemical reduction methods. Various characterizations are done using X-ray diffraction, Raman's spectra, Fourier transform infrared, scanning electron microscopy and atomic force microscopy. Different concentrations of 0.01, 0.1, and 0.3 g/l of rGO/water nanofluids are then prepared by ultra sonic homogenizer and probe sonicator. Dynamic light scattering technique is used to identify the size of rGO flakes in DI water. The thermal conductivity, viscosity, and surface tension of rGO/water nanofluids reveal their dependency on concentrations and temperature. Due to the improved dispersion stability as evident from zeta potential, the thermal conductivity of 0.01 and 0.1 g/l concentrations exhibits negligible change whereas 0.3 g/l shows a minimal change for a period of five days. The enhancement in thermal conductivity of 0.3 g/l of rGO/water nanofluid at 75 °C is 10%. The rGO/water nanofluids exhibit Newtonian behavior at higher shear rates due to the weakening of intermolecular interactions. The enhancement in surface tension is mainly due to the increase in surface energy by the accumulation of rGO flakes at the liquid–gas interface. Studies on wettability indicate an increase in contact angle with concentrations. Though it is not favorable as it reduces the contact between solid and liquid surface, many research works explain the enhanced boiling heat transfer mainly with porous layer rather than contact angle. Results show that the rGO/water nanofluid can be used as a suitable replacement for the conventional fluids in heat transfer applications.



## 1. Introduction

The performance of heat transfer systems like heat exchangers, solar heaters, nuclear reactors, refrigerators, and hydrogen storage devices mainly depends on the transport properties of conventional heat transfer fluids such as water, ethylene glycol, and oil. Since improvements in the properties of these fluids have become more critical, research on the innovative working fluid called nanofluid is a fascinating one for the last two decades owing to their enhanced properties. Although nanofluids have potential benefits than conventional heat transfer fluids, they are not widely used in heat transfer applications due to the difficulty in preparing stable nanofluids. The formulation of nanofluids with better stability is the difficult task for the researchers and scientists rather than conducting experimental studies with different volume/weight fractions till now. Various methods such as pH control, surfactants and surface functionalization are currently available to improve the dispersion stability. But these methods are still at research level. It is believed that the nanofluids are stable when the pH value is far away from the Isoelectric point (IEP) [1,2]. On the other hand, though the addition of surfactants improves the dispersion stability to a greater extent, it affects the transport properties of nanofluids. For instance, the presence of surfactants decreases the thermal conductivity of nanofluids [3].

As seen from the literature, most of the research work showed better thermal conductivity at lower temperatures and an increase in thermal conductivity with temperature and/or concentrations besides the effect of particle size [4,5]. Further, the enhancement in thermal conductivity is mainly due to the Brownian motion of nanoparticles, layering of base fluid molecules over nanoparticles, particle clustering and ballistic heat transport respectively [5]. On the other hand, a number of analytical models are being used to identify the enhancement in thermal conductivity of nanofluids.

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However, the Nan model [6] predicts the thermal conductivity closer to the experimental values of nanofluids than any other analytical models.

In the recent years, viscosity measurement has attracted researchers due to its role in many of the continuous fluid flow systems and heat transfer studies of nanofluids. Viscosity of nanofluids database shows an increase in concentration may lead to: (i) Newtonian/non-Newtonian behavior, and (ii) linear/non-linear variation. Also, the viscosity is dependent/independent of temperature [7]. Though several empirical correlations and analytical models are available to predict the viscosity of nanofluids with respect to volume fraction and temperature, no model can predict the viscosity of nanofluids precisely [7,8].

Surface tension plays a major role in boiling heat transfer applications, oil recovery efficiency, the capability of cleaning oil spills, etc. Unfortunately, only few studies have been reported the effect of concentrations and temperature on surface tension [9,10]. Khaleduzzaman et al. [9] reported in their concluding remarks that no empirical correlation or model is capable of predicting the surface tension of nanofluids with respect to concentrations. Further, they suggested that more studies would require on particle concentrations and temperature as there are ambiguities within the available results. It is also identified that the surface wettability plays a vital role in determining the critical heat flux (CHF) in boiling heat transfer. Most of the research on nanofluid boiling found an enhanced CHF due to the reduction in contact angle (improved surface wettability) by the deposition of nanoparticles on the heating surface. Further, the contact angle decreases with the increase of concentrations [1,11].

Therefore, the dispersion stability and transport properties of nanofluids are the important ones for analyzing the heat transfer performance of many thermal engineering systems. Since most of the studies devoted to the one dimensional nanomaterials and there are inconsistencies in the available results, it is of our interest to measure the transport properties of two dimensional material, i.e. reduced graphene oxide (rGO) based nanofluids. Graphene – a new allotrope of carbon in 2D form, $sp^2$-hybridized and one atom layer thick structure has dominated advances in Nanoscience and nanotechnology recently. It is found to be an ideal material for solar batteries, capacitors, liquid crystals, nano-electro-mechanical systems, etc. due to its high specific surface area, excellent thermal conductivity, and unique transport properties [12,13]. However, the usage of pristine graphene is very limited in heat transfer applications due to its hydrophobic nature and hence stable suspension of graphene in water and other base fluids is an important issue [14]. Alternately, rGO is a promising one in the recent years. Though rGO becomes hydrophobic after oxygen removal, oxygenated functionalities have not been eliminated completely during chemical reduction process, which is the advantage of adding rGO flakes in water [13]. By keeping these points, the main objective of this work is to measure the transport properties of rGO/water nanofluids at different temperatures and weight fractions along with its stability analysis and also to study the wetting characteristics of rGO flakes on different heating surfaces for the benefit of heat transfer community.

In the present work, rGO is synthesized from commercially available graphite using modified Hummer and chemical reduction methods. Various characterizations have been carried out to confirm the rGO thus prepared. The different concentrations of 0.01, 0.1, and 0.3 g/l of rGO/water nanofluids are then prepared by dispersing rGO flakes in DI water. The dispersion stability, transport properties, and surface wettability are evaluated along with supportive evidence to determine the feasibility of using this in heat transfer applications.

## 2. Experimental methods

### 2.1. Synthesis of rGO

The graphite oxide (GO) is prepared from graphite using modified Hummers method [15]. Typically, 2 g of graphite powder, 1 g of sodium nitrate, and 96 ml of concentrated sulfuric acid are stirred in a beaker for about 10 min and the mixture is then kept in an ice bath for 40 min duration. After this, 6 g of potassium permanganate is slowly added into the mixture by keeping its temperature below 20 °C for about 30 min. The beaker is now removed from the ice bath and the mixture is stirred at room temperature for 18 h. The resulting thick paste or brown slurry is then added into 150 ml of DI water and stirred for another 10 min. Then, 240 ml of DI water is added followed by a slow addition of 5 ml of hydrogen peroxide to the mixture and the color of the solution is changed to brilliant yellow along with bubbling. After continuously stirring for about 2 h, the mixture is filtered and washed with 250 ml of 10% HCl aqueous solution followed by DI water and ethanol to remove other ions. Finally, the resulting filtrate is dried by vacuum process and thus the GO is obtained.

The reduction of graphite oxide into rGO is as follows: 2 g of GO is dispersed in DI water and the solution is then sonicated for about 2 h to obtain a uniform dispersion of exfoliated GO. Next, 16 g of sodium borohydride is added to this GO aqueous solution under constant magnetic stirring. The reduction process is carried out for 12 h. The pH of the solution is maintained in the range of 9–10. Then the product is filtered and washed thoroughly with DI water. Finally, it is dried in vacuum for 24 h and crushed into fine powder to obtain the rGO.

### 2.2. Preparation of rGO/water nanofluids

In this study, two-step method is followed to prepare the rGO/water nanofluids. First, the required quantities of rGO dry flakes are weighed using an Acculab – VI – 1 mg precision balance. It is then dispersed in the known amount of DI water to obtain 0.01, 0.1, and 0.3 g/l concentrations of rGO/water nanofluids. The prepared rGO/water nanofluids are kept in an ultrasonic homogenizer and probe sonicator for about 12 h. After that, various analyses have been carried out to evaluate the dispersion stability of rGO/water nanofluids and discussed in Section 3.2.

## 3. Results and discussion

### 3.1. Characterization of rGO

The prepared rGO is subjected to various characterization studies (Fig. 1) such as X-ray diffraction (XRD), Raman spectroscopy, Fourier transform infrared (FT-IR), scanning electron microscopy (SEM), and atomic force microscopy (AFM). The XRD studies are carried out by Rigaku Ultima III, employing a scanning rate of 4° per min. with CuK$\alpha$ radiation ($\lambda = 0.154$ nm). Raman spectrum is obtained over the range of 500–3200 $cm^{-1}$on the rGO flakes using Renishaw Invia Raman Microscope. An FT-IR spectrum is recorded on a Perkin Elmer over the range of 4000–500 $cm^{-1}$ to identify the functional groups. The surface morphology and crystalline nature are examined by SEM (Tescan Vega3) whereas the size and thickness of rGO flakes are obtained by AFM (Agilent, US).

The XRD pattern of graphite, GO, and rGO is shown in Fig. 1(a). In this figure, graphite shows a sharp diffraction peak at $2\theta = 26.4°$, which is indexed to the (0 0 2) peak of graphite structure of carbon. Due to the insertion of oxygenated functionalities and water