



Effect of particle size and viscosity on thermal conductivity enhancement of graphene oxide nanofluid[☆]



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ABSTRACT

In this study, first, graphene oxide nanosheets were synthesized based on the modified Hummers methods. The physicochemical properties of fabricated graphene oxide were characterized using X-ray diffraction analysis (XRD), a scanning electron microscope (SEM), and UV–Vis spectrophotometry. Second, graphene oxide nanofluids were prepared at different concentrations (0.01, 0.05, 0.1 and 0.5 wt.%) in water (base fluid). Particle-size distribution and stability of the colloidal solution of the graphene oxide nanofluids were investigated using dynamic light scattering and zeta potential techniques. Also, rheological behavior of graphene oxide nanofluids was studied at different temperatures (25 °C, 40 °C, and 60 °C) and different shear rates (10–100 1/s). Results show that both particle size and viscosity of graphene oxide nanofluids increased linearly by increasing the graphene oxide concentration from 0.01 to 0.1 wt.%, but there was a very sharp increment on average particle size and viscosity by increasing the concentration to 0.5 wt.%. The thermal conductivity of graphene oxide nanofluids was measured at different temperatures. Thermal conductivity of graphene oxide nanofluids depends on both particle-size distribution of graphene oxide and viscosity of graphene oxide nanofluids. All graphene oxide nanofluids showed enhanced thermal conductivity compared to the base fluid, water. Increasing the graphene oxide concentration from 0.01 wt.% to 0.1 wt.% resulted in 8.7% and 18.9% thermal conductivity enhancement at 25 °C, respectively. However, further concentration increment to 0.5 wt.% increased thermal conductivity to 19.9%. This behavior shows that there is an optimal concentration of graphene oxide at which particle size and viscosity of graphene oxide nanofluids exhibit significant thermal conductivity enhancement, and further concentration increments have no significant effect on thermal conductivity enhancement.

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1. Introduction

Nanotechnologies have been proven to enhance efficiency in research areas including pharmaceuticals [1], environmental processes [2,3], petroleum [4], automobiles [5] and power generation [6]. The critical demand for highly efficient thermal transport solution has become a major challenge for industries, especially in terms of energy and power supply [7]. Heat transfer plays a major role in many types of industries, such as transportation, air conditioning, power generation, process engineering and electronics [8]. Therefore, there is a need to improve heat transfer capabilities. The advantages of nanoparticle properties have resulted in the introduction of a nanofluid (NF). In general, an NF is a suspension of ultra-fine particles, rods or tubes with an extremely high thermal conductivity compared to the conventional base fluid [9]. NFs could enhance thermal conductivity and heat transfer efficiency and also diminish process costs. There is a universal motivation for

discovering the optimal conditions of fabrication, characterization and implications of NF to achieve their maximum advantages.

Nanoparticle physicochemical properties such as size, shape and surface charge and also interaction between nanoparticles and base fluid are controlling factors for NF thermal conductivity. Hong and Yang [10] compared the effect of Fe and Cu nanoparticles less than 10 nm in particle size on thermal conductivity and reported that suspension of highly thermal conductive materials is not always effective to improve thermal transport property of NF. Li et al. [11] investigated the effect of particle size on thermal conductivity of NF. They compared 36 and 47 nm particle size Al₂O₃ nanoparticles in water base nanofluids and reported that each of the nanoparticles showed different thermal conductivity increment trend due to their particle size. They concluded that thermal conductivity of Al₂O₃/water has a nonlinear relationship with temperature, volume fraction and nanoparticle size, and there is an optimal combination condition for each of these factors at a certain bulk temperature. However, there is an inconsistency in reported data regarding the effect of particle size on thermal conductivity. For example, Chopkar et al. [12] showed that Al₂Cu nanoparticles with smaller particle size (~30 nm) significantly increased NF thermal conductivity

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compared to Al₂Cu nanoparticles with larger particle size (~ 150 nm) in water and ethylene glycol based NFs. In a similar study, Moghadasi et al. [13] reported that as the particle size of CuO decreased, the NF thermal conductivity improved. The findings of Beck et al. [14] showed that thermal conductivity of Al₂O₃-water NFs increased by increasing the particle size from 8 nm to 282 nm. Also, Every et al. [15] observed that the thermal conductivity of zinc-sulfide increased by adding large particles of highly conducting diamond, but lowered by addition of sub-micron size particles of diamond.

There is a relationship between rheological, aggregate size and NF thermal conductivity. Available literatures show that NFs can exhibit both Newtonian and non-Newtonian rheological behaviors, depending on particle size, shape and concentration [16]. Kwak et al. [17] studied the relationship between viscosity and thermal conductivity of copper oxide NFs dispersed in ethylene glycol and reported that based on the non-Newtonian rheological property of NFs, substantial enhancement in thermal conductivity was attainable only when particle concentration is below the 0.002 volume fraction. Namburu et al. [18] tested the rheological properties of copper oxide nanoparticles in a mixture of ethylene glycol and water and showed that for the particle volume concentration of 0% to 6%, NFs exhibited the Newtonian behavior in temperature ranging from – 35 to 50 °C.

Recently, graphene-based particles received notable attention due to their large intrinsic thermal conductivity and low density in comparison to metals or metal oxides nanoparticles [19]. For example, Ghozatlo et al. [20] reported that adding 0.075 wt.% of graphene to the base fluid resulted in an improvement of thermal conductivity up to 31.83%. Yu et al. [21] developed the ethylene-glycol based nanofluids containing graphene nanosheets that showed up to an 86% thermal conductivity increment. In contrast to the theoretical study of the thermal conductivity of graphene, the experimental work about the heat transfer property of graphene-based materials is rare due to their unique physicochemical properties and their different compatibility with other compounds [21]. Also, very few research studies have been performed on the effects of viscosity and particle size on thermal conductivity of graphene oxide nanofluids [22]. It can be seen from the literature that there is a need for a more in-depth understanding of the relationship among particle size, rheological behavior and thermal conductivity improvement using graphene oxide.

In this study, graphene oxide was fabricated and then characterized using scanning electron microscope (SEM), X-ray diffraction, and Ultraviolet–Visible (UV–Vis) spectroscopic methods. The graphene oxide NFs were prepared at 0.01, 0.05, 0.1 and 0.5 wt.% concentrations, and thermal conductivity of each sample was measured at three temperatures (25, 40 and 60 °C). The rheological behavior and aggregation behavior of each sample were measured, and their effects on thermal conductivity were studied.

2. Materials and methods

2.1. Materials

Graphite powder (99.99%, 45 m) was purchased from Bay Carbon, Inc. (Bay City, Michigan, USA). Sulfuric acid (H₂SO₄), hydrochloric acid (HCl), sodium nitrate (NaNO₃), potassium permanganate (KMnO₄) and hydrogen peroxide 30% (H₂O₂) were purchased from Fisher Scientific (Pittsburgh, PA, USA). All the chemicals were of analytical reagent grades and used as received, without further purifications. Deionized (DI) water was produced by the Millipore DI system Direct-Q 3 UV (18.2 MΩ cm) (Massachusetts, USA).

2.2. Fabrication of graphene oxide and preparation of nanofluids

Graphene oxide was prepared from graphite powder based on the modified Hummers methods [23]. Briefly, 2.5 g of graphite powder was treated with 57 ml of concentrated sulfuric acid in an ice bath to

prevent the overheating. Sodium nitrate (1.75 g) was added slowly to the mentioned solution and mixed for 1 h. Then, 7.5 g of potassium permanganate was added to the solution and stirred for 12 h. The resulting solution was diluted by adding 1250 ml of DI water under vigorous stirring. Hydrogen peroxide (12.5 ml, 30%) was added to the solution and stirred for 2 h to make sure the potassium permanganate reaction was complete. The final mixture was washed with 57 ml of concentrated hydrochloric acid and 5 l of DI water, respectively. Finally, the mixture was filtered and the graphene oxide cake was collected and dried for 12 h at 40 °C. In general, the mixture of sulfuric acid, hydrochloric acid, and potassium permanganate oxidize the natural graphite powders in the water-based solution. During this process, bonding of hydroxyl, carbonyl and epoxy groups to the graphite structure and insertion of H₂O and ions into the graphene layer result in an interlayer spacing increment [24–26].

Graphene oxide NFs were prepared by dispersing different masses of graphene oxide in DI water. Graphene oxide in amounts of 0.01, 0.05, 0.1 and 0.5 g was dispersed in an appropriate mass of DI water, Table 1, and stirred for 5 h. Then, graphene oxide NF colloidal solutions were sonicated at 130 W and 42 kHz using an ultrasonication instrument (Branson Ultrasonics, Danbury, CT, USA) for 1 h. Table 1 shows graphene oxide NF sample specifications.

2.3. Dynamic light scattering and zeta potential

The dynamic light scattering method was used for investigating particle size and particle size distribution of graphene oxide particles in DI water (base fluid). The DLS measurements were performed using the Malvern Zetasizer ZS instrument (Malvern Instruments Ltd., Malvern, United Kingdom) equipped with an He–Ne laser operated at $\lambda = 633$ nm [27]. A 173° angle was set for the detector position to take advantage of the back scattering mode. Automatic position of the attenuator was selected to achieve the optimum laser intensity based on the sample concentration. The intensity-based particle size distribution was obtained from fitting the recorded data to a single or multi-exponential correlation function, measuring the mean size (z-average diameter), and estimating the width of the distribution (polydispersity index) according to the International Standard ISO13321, Methods for Determination of Particle Size Distribution.

The same Malvern Zetasizer ZS instrument was used for zeta potential measurement of graphene oxide NFs. The scattered light was detected at an angle of 13° by the detector. The zeta potential (ζ) was calculated from electrophoretic mobility (EM) of particles using the Henry equation by considering the Smoluchowski approximation [27,28]. Three replicates of each sample were tested and an average value reported.

2.4. Characterization techniques

The crystal structure of graphene oxide was investigated by an X-ray diffractometer (XRD), Rigaku Ultima IV X-Ray Diffractometer, Rigaku, Texas USA. Data was collected from 5° to 80° at a scan rate of 3 degree/min. Morphological characterization of fabricated graphene oxide was examined by an environmental scanning electron microscope (ESEM), FEI Quanta 200, 149 FEI Company, Hillsboro, OR, USA.

Table 1
Specification of graphene oxide nanofluid samples.

Sample name	Graphene oxide (wt.%)	Deionized water (wt.%)
GO-NF-0.01	0.01	99.99
GO-NF-0.05	0.05	99.95
GO-NF-0.1	0.1	99.90
GO-NF-0.5	0.5	99.50

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