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Experimental thermal conductivity and viscosity of nanodiamond-based propylene glycol and water mixtures



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ABSTRACT

Nanodiamond (ND) nanoparticles of primary particle size of 5–10 nm can be produced in large quantity by using commercially available ultra-dispersed diamond (UDD) powders. The UDD powders contains large quantity of carbon impurities, which can be removed using strong acid treatment, dry salt and dry sucrose to make UDD powders into single ND particles. The formation of carboxyl (COOH) groups on the surface of ND particles was analyzed by FTIR spectrum. The X-ray powder diffraction (XRD), transmission electron microscope (TEM) and Raman spectra was also performed on acid treated ND particles. The propylene glycol/water mixtures at different weight ratios were used as base fluids for the preparation of stable ND nanofluids. The thermal conductivity and viscosity of ND nanofluids were estimated experimentally at different particle concentrations and temperatures. Based on the results, the thermal conductivity enhancements are 18.8%, 16.8% and 14.1% at 1.0 vol.% of 20:80%, 40:60% and 60:40% PG/W based nanofluids. Similarly, the viscosity enhancements are 1.55-times, 1.50-times and 1.66-times for the same 1.0 vol.% of 20:80%, 40:60% and 60:40% PG/W based nanofluids at a temperature of 60 °C, respectively compared to their respective base fluids. New thermal conductivity and viscosity correlations have been developed based on the experimental data. Theoretical approach is used to identify the heat transfer benefits of the prepared propylene glycol/water based ND nanofluids in laminar and turbulent flow conditions using the thermal properties.

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1. Introduction

The carbon based materials such as multi walled carbon nanotubes (MWCNT), single walled carbon nanotubes (SWCNT), graphene and nanodiamond (ND) materials contain high thermal conductivity compared to other materials. Among all the carbon materials ND particles creates great interest because of their superior hardness, thermal conductivity, i.e. 1000 W/m K and Young's modulus, high electrical resistivity, attractive optical characteristics, and excellent chemical stability and biocompatibility. The single phase heat transfer fluids such as water (W), ethylene glycol (EG), propylene glycol (PG) and engine oil (EO) are primarily used in may industrial applications, among others, power, chemical and food industries. The heat transfer capability of these single phase heat transfer fluids is limited because of its poor thermal conductivity. The thermal conductivity of single phase heat transfer fluids may be enhanced by dispersing small quantity of solid particles. The Choi [1] and his team developed nanofluids (fluids contain nanometer size particles) and obtained higher thermal conductivity compared to base fluids.

The preparation of ND based nanofluids and their thermal properties are given below. The commercially available ultra-dispersed diamond

* Corresponding authors. E-mail addresses: sslingala@gmail.com (L.S. Sundar), mksingh@ua.pt (M.K. Singh). (UDD) powders of an average size of 0.1–2.0 µm have been used by Dolmatov [2] for the preparation of ND nanofluids, but he faced serious agglomeration problem with UDD powders in the base fluid. The reason for the agglomeration of UDD powders in the base fluid is due to the presence of carbon impurities. During the preparation of UDD powders, igniter is used in the detonation process which causes metal and carbon impurities and those are aggregates or attached to the outer surface of the ND particles. The purification of UDD powders is important for achieving good stable nanofluids. There are different methods to purify the UDD powders. The UDD powders can be treated with strong acids such as HNO₃, mixture of H₂SO₄/HNO₃, K₂Cr₂O₇ in H₂SO₄, KOH/KNO₃ to remove the carbon impurities [3,4]. The carbon impurities present in the UDD powders have been removed using mixture of H₂SO₄ and HNO₃ [5–9]. Another method for removing the carbon impurities from the UDD powders is with dry salt or dry sucrose [10]. Jee and Lee [11] used strong acids of H₂SO₄ and HNO₃ in the molar ratio of 3:1 for the purification of UDD powders and observed the removal of carbon impurities. They also observed the formation of carboxyl groups on the surface of ND particles based on the FTIR analysis. Xie et al. [12] also used acid mixtures of perchloric acid, nitric acid and hydrochloric acid (based on the procedure of Jang and Xu [13]) for the removal of carbon impurities from the UDD powders and prepared stable ND nanofluids in the base fluid of 55:45% ethylene glycol/water mixture and observed thermal conductivity enhancement of 18% at 2.0 vol.%. Sundar et al. [14] also

used strong acids of H₂SO₄ and HNO₃ in the molar ratio of 3:1 for the purification of UDD powders and prepared 20:80%, 40:60% and 60:40% ethylene glycol/water mixture based ND nanofluids and estimated both thermal conductivity and viscosity experimentally. Branson et al. [15] observed thermal conductivity enhancement of 12% at 0.9 vol.% of nanodiamond (ND)-poly (glycidol) polymer brush/EG nanofluids, similarly observed thermal conductivity enhancement of 11% at 1.0 vol.% of ND.oleic acid/mineral oil nanofluids. Ghazvini et al. [16] prepared 20% of water and 50% of engine oil based ND nanofluids and obtained thermal conductivity enhancement of 25% at 1.0 wt.% Taha-Tijerina et al. [17] prepared mineral oil based ND nanofluids and obtained thermal conductivity enhancement of 70% at 0.1 wt.%. Yeganeh et al. [18] observed thermal conductivity enhancements of 7.2% and 9.8% for water based ND nanofluids at 3.0 vol.% at temperatures of 30 °C and 50 °C, respectively. Tyler et al. [19] obtained viscosity enhancement of 80% for midel oil based ND nanofluids at 3.0 wt.%. Yu et al. [20] obtained thermal conductivity enhancement of 17.23% for ND/EG nanofluid at 1.0 vol.% at a temperature of 30 °C. The earlier works reveal the preparation, estimation of thermal conductivity and viscosity of water, ethylene glycol, mineral oil and midel oil based ND nanofluids.

Different weight or mass ratios of ethylene glycol/water (EG/W) or propylene glycol/water (PG/W) fluids are commonly used as engine coolants for automobile radiators and also used as coolant for electronic equipments, heating industrial and residential buildings in the cold countries of the world such as Alaska, Canada, Northern Europe and Russia, because of its low freezing point i.e. <-50 °C [21]. The low freezing point of water (below 0 °C) may be achieved by adding different ratios of ethylene glycol or propylene glycol, so that the EG/W or PG/W fluid can operate up to -50 °C without any difficult [22]. Namburu et al. [23] first time prepared 60:40% (weight ratio) of EG/W based CuO nanofluids and observed viscosity enhancement of 4.5-times and 3.1times at 6.12 vol.% at temperatures of -35 °C to 50 °C. Vajjha and Das [24] also prepared 60:40% (weight ratio) of EG/W based Al₂O₃, CuO and ZnO nanofluids and measured thermal conductivity up to 10.0 vol.%. Sundar et al. [25,26] also prepared 20:80%, 40:60% and 60:40% EG/W based Fe₃O₄ nanofluids and measured both thermal conductivity and viscosity experimentally. The earlier works reveals the preparation, estimation of thermal conductivity and viscosity of EG/W mixture based Al₂O₃, CuO, ZnO and Fe₃O₄ nanofluids.

On the other hand preparation of propylene glycol/water (PG/W) based nanofluids and estimation of thermal properties is also important before use in heat transfer equipments, because these PG/W fluids are also used as engine coolants for automobile radiators. Prasher et al. [27] prepared PG based Al₂O₃ nanofluids and estimated viscosity experimentally and proposed viscosity model. Naik and Sundar [28] prepared 30:70% PG/W based CuO nanofluid and estimated thermal conductivity and viscosity experimentally. Kole and Dey [29] prepared 50:50% PG/W based Al₂O₃ nanofluids and observed nanofluids obey Newtonian behaviour in the measured temperature range from 10 °C to 50 °C. Sundar et al. [30] also prepared 20:80%, 40:60% PG/W based Fe₃O₄ nanofluids and estimated thermal conductivity and viscosity experimentally. The earlier works reveals the preparation, estimation of thermal conductivity and viscosity of PG/W mixture based Al₂O₃, CuO, and Fe₃O₄ nanofluids. Therefore, there is no literature on the thermal properties of PG/W mixture based ND nanofluids.

The present work focused on the preparation of different weight ratio of PG/W (20:80%, 40:60% and 60:40%) mixture based ND nanofluids and the estimation of thermal conductivity and viscosity experimentally at different particle concentrations and temperatures. The carbon impurities present in the UDD powders is removed with strong acid treatment, dry salt and dry sucrose to make them into single ND crystals. The optimized method for purifying the UDD powders is established. The PG/W nanofluids are prepared with acid treated ND particles. The Prasher et al. [27] model and Mouromtseff number [31] is used to study the heat transfer benefits of PG/W based ND nanofluids in laminar and turbulent flow conditions.

2. Experimental methods

2.1. Purification of UDD powders

The ultra-dispersed diamond (UDD) powders were purchased from International Technology Centre, USA (http://www.itc-inc.org) with the following specifications: 98% purity, 5–10 nm particle core size, cubic shape, grey color, 300–400 m²/g specific surface area. The other chemicals such as sulfuric acid (H₂SO₄), nitric acid (HNO₃), sodium chloride (NaCl), sucrose ($C_{12}H_{22}O_{11}$) and propylene glycol ($C_{3}H_{8}O_{2}$) were purchased from Sigma-Aldrich Chemicals, USA.

The purchased UDD powders contain large quantity of carbon impurities and which can be surrounded at the outer surface of ND core. If the nanofluids prepared with UDD powders, there may be a problem of agglomeration of the particles in the base fluids. In order to avoid the agglomeration problem with UDD powders in base fluids, it is important to remove the impurities. There are various methods to remove the carbon impurities from UDD powders. In this paper three methods were used and the optimized method was suggested. The three methods are (i) UDD powders treated with strong acid solution (ii) UDD powders treated with dry salt powder (iii) UDD powders treated with dry sucrose powder.

In the strong acid treatment method, 5 g of UDD powders were dispersed in 300 ml of sulfuric acid (H₂SO₄) (18.4 M)/nitric acid (HNO₃) (16 M) mixture in the 3:1 M ratio and stirred continuously up to 72 h. After that the acid treated UDD powders were dried in an oven at 80 $^\circ$ C up to 12 h [14]. This method helps to remove the carbon impurities present in the UDD powders and the formation of carboxyl (COOH) groups on the surface of ND particles. In the dry salt powder method, 1 g of UDD powders were mixed with 7 g of dry salt powder and milled at 500 rpm for 4 h using 150 mm of steel grinding balls (Union Process, Inc., U.S.A.) in a 110 cm³ milling chamber. During the milling process, there may be a possibility of heat generation by friction and impact which causes the thermal damage of the UDD powders. This can be prevented by cooling the milling chamber. This method also helps for the formation of carboxyl (COOH) groups on the surface of ND particles. In the dry sucrose method, 1 g of UDD powders were mixed with 7 g of sucrose and milled at 500 rpm for 4 h using 150 mm of steel grinding balls in a 110 cm³ milling chamber. The schematic representation of milling procedure is shown in Fig. 1. The UDD powders milled with salt and sucrose as milling media, the final product is washed several times with distilled water to remove the carbon impurities and then dried in an oven at 80 °C for 12 h.



Fig. 1. Schematic representation of UDD powders milled with salt and sucrose media.

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