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Study of environmentally friendly and facile functionalization of graphene nanoplatelet and its application in convective heat transfer

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ABSTRACT

The chemical functionalization of carbon-based nanomaterial typically involves toxic and corrosive inorganic acids that are harmful to environment and human health. In this study, an environmentally friendly, facile and cost effective procedure for synthesizing a novel and highly dispersed functionalized graphene nanoplatelets (GNPs) nanofluids for use as a heat transfer fluids was developed. In the new approach, GNPs were functionalized covalently with Gallic Acid (GA) in a one-pot free radical grafting method. The Gallic acid-treated graphene nanoplatelets (GAGNPs) were dispersed in distilled water (DI water) with different weight concentrations to prepare GAGNPs-water nanofluids (nano-coolants). The effectiveness of the functionalization process was verified by the Fourier transform infrared spectroscopy (FTIR), thermogravimetric analysis (TGA) and transmission electron microscopy (TEM). The UV–visible absorption spectroscopy was used to show a sustained stability of nanoparticles. Thermo-physical and rheological properties of GAGNPs aqueous suspensions with different weight concentrations were experimentally investigated. This was followed by measurement of the convective heat transfer coefficient, Nusselt number and friction factor for fully developed turbulent flow of GAGNPs nanofluids at a constant heat flux. The experimental results were compared with those of the base fluid. The GAGNPs nanofluids showed a significant increase in the convective heat transfer coefficient and Nusselt number, while the increases in the friction factor and pumping power were small. Furthermore, the comparison showed that the overall performance index was higher than 1. The novel and eco-friendly GAGNPs nanofluids have the potential to be used as highly effective working fluids for various heat transfer applications.

1. Introduction

Energy optimization has been identified as the most important issue in sustainable development including the infrastructure, transportation, defense and many industries. To realize energy optimization, different kinds of cooling systems with various designs have been employed over the past century. Convective heat transfer plays an important role in most of thermal equipment including cooling devices, solar collectors and heat exchangers which usually use water, propylene glycol, ethylene glycol and oil as working fluids. These fluids, however, show low thermal conductivity that leads to low heat transfer efficiency [1]. In the last decade the potential usage of suspension of nanoparticles with high effective thermal conductivity has gained considerable attention [2–10]. Such suspension, referred to as nanofluids have attracted researcher's interest since the materials in the nanometer size has unique physical and chemical properties. In particular, nanofluids exhibit high thermal conductivity and good heat transfer coefficient which makes them suitable candidate as high performance heat transfer fluids [3,11,12]. Various nanoparticles, such as carbon nanotubes (CNT), Graphene nanoplatelets (GNPs), Graphene oxide (GO), fullerene, copper oxide (CuO), aluminum oxide (Al_2O_3), and silicon dioxide (SiO_2) have been used to produce nanofluids with enhanced thermal conductivity. More recently, remarkable enhancement in thermo-physical, rheological and heat transfer properties of carbon based nanofluids was reported in the literature [13–19].

Lee et al. [20] experimentally studied the thermal conductivity of alumina (Al_2O_3)–water, alumina–ethylene glycol (Al_2O_3-EG) and CuO–EG. They reported about 23% enhancement in the thermal

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conductivity of EG in presence of CuO. Murshed et al. [21] investigated the thermal conductivity of TiO_2-water nanofluids. Their results suggested that there was a nonlinear trend between thermal conductivity and volume concentration of nanoparticles. Pak and Cho [22] investigated the turbulent convective heat transfer characteristics of water–Al_2O_3 and water–TiO_2 nanofluids flowing through a horizontal tube. The results showed that the Nusselt number of nanofluids increased with increasing Reynolds number and the volume concentration. Xuan and Li [23] explored experimentally the flow and convective heat transfer of copper based-water nanofluids, and reported that the friction factor of the nanofluids for dilute concentration was approximately the same as that of water. The thermal conductivity and heat transfer of multiwalled carbon nanotubes (MWCNTs)-based water nanofluids were investigated in previous investigations [24,25] where a noticeable enhancement of heat transfer was reported that was attributed to the thinning of the thermal boundary layer by MWCNTs and the associated reduction of the thermal resistance.

Among various carbon-based nanostructures, the graphene-family nano-materials have received more attention due to their attractive thermal, electrical and mechanical properties [26–29]. Graphene, an atomically thin, two dimensional lattice of Sp^2-hybridized carbon has exhibited exceptional thermo-physical properties [30]. These unique properties make it a promising additive for many applications such as inkjet printing, conductive thin films, solar cells, polymer composites, aerogels and heat exchangers [26,31]. In addition to high specific surface area, graphene presents tendency to agglomerate by strong π-π stacking interaction, while the dispersibility of graphene in aqueous media is considered as one of the most important factor in heat transfer equipment, films and composites [32]. Hence several methods were employed to improve stability of graphene in aqueous and organic media by chemical and physical methods containing covalent and non-covalent functionalization of graphene.

Recently, chemical functionalization with the hydrophilic organic group like carboxyl, esters, alkalis and amine group on the surface of GNP has promised the stability of the graphene sheet in aqueous media. Oxidizing GNP by the usage of strong acids, the mixture of sulphuric acid and nitric acid, is reported as an effective method of synthesizing water soluble GNP [32,33], although there are many covalent reactions on carbon based nanomaterials such as, radical addition, alkali metal reduction, Bingel cyclopropanation, Nitrene cycloaddition, 1,3-dipolar cycloaddition, electrophilic addition, nucleophilic addition, carbene addition and Diels-Alder cycloaddition [34,35]. The free radical coupling is a promising method of covalent functionalization of carbon nanostructure. In this method, proxides and aryl diazonium salts have substituted anilines and benzophenone were utilized as starting materials [36]. In comparison with other fictionalizations methods, the covalent functionalization technique may produce defects on the GNP sheets and also generates hazardous and toxic wastes.

The non-covalent functionalization of GNPs based on the π-π staking interaction and polymer wrapping of surfactants such as sodium dodecyl benzene sulphonate (SDBS), sodium dodecyl sulfate (SDS), gum arabic (GA) and cetyltrimethylammonium bromide (CTAB) improves GNPs solubility in polar solvents. However, there are some undesirable effects of the above mentioned surfactants on the thermophysial properties of carbon based nanofluids such as increasing viscosity and foam formation in the colloidal suspensions which have limited surfactant applications [37]. In addition, both strong acids and some organic solvents often cause environmental pollution, equipment corrosion and health hazard. Therefore, it is imperative to develop an environmentally friendly method to functionalize carbon base nano-structures [38].

Recently, green chemistry and green chemical processes has attracted considerable attentions due to their benign effects on the environment and their ability to function well or even better than other more toxic traditional options [39,40]. Green chemistry was proposed by chemists which contains some important principles such as 1 – the elimination of chemical waste, 2 – design of safer chemicals 3 – reduction of hazardous chemical synthesis, 4 – real time analysis for pollution prevention, 5 – improved energy efficiency, 6 – use of renewable source, 7 – inherently safer chemistry for accident prevention [40,41].

Among the green materials, Gallic acid (GA) is a natural polyphenol antioxidant extracts from green tea, berries, grapes, wine and also found in some hard wood plant species [42]. It can act as a non-toxic corrosion inhibitor by adsorbing on the metal surfaces. [43]. Moreover, Gallic Acid has been shown to be an effective stabilizing agent for the protection of biodiesel oxidation [44].

Due to the Gallic acid structure (phenol) and green properties, it can be a suitable candidate for improving the functionalization of GNPs in aqueous media. The main objective of the present study is developing an environmentally friendly, cost-effective, and industrially scalable method for synthesizing covalent Gallic acid-treated graphene nanoplatelets (GAGNPs). The other objective is to develop a stable nanofluid of suspension of GAGNPs in aqueous media, and demonstrate its effectiveness in improving the convective heat transfer in a closed conduit. To validate the successful implementation of the functionalization, the treated nanoparticles were characterized by Fourier transform infrared spectroscopy (FTIR), thermogravimetric analysis (TGA), transmission electron microscopy (TEM), and UV–visible absorption spectroscopy. In addition, the thermo-physical and rheological properties of the synthesized nanofluids were measured at different weight concentrations and temperatures. Furthermore, the convective heat transfer and friction factor were experimentally evaluated for the nanofluids at different concentration and the data were validated against the empirical correlations. Afterwards, the heat transfer coefficient, Nusselt number, friction factor, pumping power and performance index were evaluated for the synthesized water-based GAGNPs nanofluids under turbulent flow conditions in a horizontal stainless tube.

2. Materials and methods

Pristine Graphene nanoplatelets (GNPs) of maximum particle diameter of 2 μm, specific surface area 750 m^2/g and purity 99.5% were obtained from, XG Sciences, Lansing, MI, USA. Rest of the chemical materials were of analytical grade such as Gallic acid (3,4,5-trihydroxybenzoic acid) and hydrogen peroxide (H_2O_2, 30%) were procured from Sigma–Aldrich.

2.1. Preparation of the GA-treated GNPs aqueous suspensions

Free radical grafting of Gallic acid onto GNPs was achieved using hydrogen peroxide (H_2O_2) and heat, respectively, as redox and thermal initiator. As a first step hydrogen peroxide as a free-radical oxidizer that generates non-toxic by products and leaves no chemical residue at high temperatures becomes unstable and decomposes spontaneously into hydroxyl radicals. These producing hydroxyl radicals will then attack Gallic acid to produce free radicals on the Gallic acid structure, which leads to linkage of the activated molecules onto the surface and edges of GNPs. In addition, the hydroxyl radicals can attack the GNPs directly, leading to formation of hydroxyl groups on the surface of GNPs. For synthesizing GA-treated GNP, 5 g pristine GNPs and 15 g Gallic acid was poured into a vessel filled with 1000 ml of distilled water (DI water) and then stirred for 15 min at 80 °C to reach a uniform black suspension. Concentrated hydrogen peroxide (35 ml) was slowly poured into the vessel during the sonication time. The