



Highly dispersed reduced graphene oxide and its hybrid complexes as effective additives for improving thermophysical property of heat transfer fluid



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ABSTRACT

The present research reported on the benign and facile preparation of highly stable reduced graphene oxide (RGO) and its role on enhancing the thermophysical properties of heat transfer liquid. Graphene oxide (GO) was prepared via chemical exfoliation route and subsequently reduced using tannic acid (TA) which served as natural based environmentally benign reducing agent. Further, a meticulous amount of different high purity carbon sources (i.e. multiwall carbon nanotube (MWCNT), carbon nanofiber (CNF) and graphene nanoplatelets (GnP)) was introduced to the RGO sheets aiming to extend the limits of its physicochemical properties for diverse applications. Results obtained led to the fact that GO was successfully reduced based on the material characterization evidences. Moreover, the addition of highly conjugated carbon structures on RGO has proven to be pivotal in promoting highly efficient thermal transport with minimal penalty on viscosity increment. It was believed that, similar to the previously documented electrical conductivity enhancement, the rise in thermal property was attributed to the modified structural network of RGO originating from the addition of different carbon allotropes which overlaid series of conducting paths for efficient phonon transport both in lateral and out of plane modes. As high as 25% increase in thermal conductivity was recorded with RGO–CNT complexes in comparison to base fluid (i.e. DI water) while a modest 4% rise in viscosity was proven to be insignificant. It was suggested from the results that morphological structure of the carbon based additives may play significant roles in different degree of enhancement of the hybrid materials.

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Abbreviations: CNT, carbon nanotube; CNF, carbon nanofiber; DI, deionized water; d, interlayer spacing between graphitic layers; D, defect within molecular structure; GO, graphene oxide; G, graphitic domain within molecular structure; GnP, graphene nanoplatelets; I, peak intensity of Raman spectra; k, thermal conductivity (W/m K); L, length (m); MWCNT, multi wall carbon nanotube; NIST, national institute of standards and technology; OD, outer diameter; RGO, reduced graphene oxide; TA, tannic acid; T, temperature, K; TEM, transmission electron microscope; UV, ultraviolet; wt, weight percentage; XRD, X-ray diffraction.

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1. Introduction

Graphene, ideally envisaged as a single layer atomic structure consisting of pristine carbon atoms interconnected in two dimensionally arranged honeycomb lattice via sp^2 molecular bond has been at the center of intense research both in academia and industry [1,2]. This newly discovered structure within the family of graphite carbon allotrope has amazed the forefront physicists due to its ability to maintain its 2D planar structural integrity by which it was once considered to be impossible to exist [3]. Fundamental research efforts on graphene have unraveled series of astonishing discoveries particularly in its response towards optical, electrical, thermal and mechanical excitations [4–6]. Due to its remarkable properties, reaching as near to the material theoretical limit, graphene has rapidly drawn vast attention from research

Nomenclature

Greek

μ	viscosity, Pa s
ρ	density, kg/m ³

Subscripts

w	water
n	nanocolloids

community. Hitherto, graphene has shown its promising potential in diverse areas such as electronics and photonics [7] medicine [8] and sensors [9].

It is evident that several routes have been pursued on graphene synthesis namely micromechanical exfoliation technique, epitaxy of graphene layer on ruthenium, chemical vapor deposition (CVD), Arc discharge method, microwaves assisted exfoliation of intercalated graphite, unzipping of CNTs, electrochemical approach and chemical reduction of exfoliated graphene oxide (GO) [10–15]. Among these, chemical reduction method is by far the most popular route taken among researchers owing to its facile nature in chemical synthesis, and require simple apparatus and easily accessible reagents within its production protocols [16].

It is evident that GO, as precursor material for graphene, suffers from extremely low electrical, thermophysical and optical properties due to its loss of systematically arranged conjugated structure during oxidation process [17]. Thus, extensive research effort has been dedicated on restoring GO to attain the original pristine graphite based molecular structure in order to harness its potentials in diverse technological spectrum. The typical route for repairing (GO) is via the removal of its oxygen rich functional groups with the use of reducing agent which catalyzes electron transfer. Other methods that have been pursued include electrochemical and thermal reduction [18,19]. This process leads to the restoration of the sp² double bond between carbon atoms dictating a repaired conjugated structure. Although the resultant product still contains holes and defects [20], the transformation enables its unique quantum effects to be explored and vindicated. It was also known that the restoration of graphitic properties in graphene serves as the essential key for ballistic phonon and electron transport to give exceptionally high thermal and electrical properties [17].

It is worthy to highlight that while the reduction process enables the restoration of graphene chemical properties, another challenge arises on the subject of improving its solubility in solvents which act as vehicle for further material processing. Controlling the dispersion of graphene has become a separate field of research due to its immense importance in expending the technological barriers. Both covalent and non-covalent functionalization approaches have been explored for graphene sheet isolation in organic liquids [21,22]. While covalent functionalization is favored for producing exceptionally high stability graphene colloids [22], it suffers from lost in electronic conjugation due to the opening of sp² carbon structure, leading to a formation of defect sites in order to pave ways for covalent reaction with other organic molecules [23]. The perturbed aromatic structures prevent ballistic phonon and electron transport leading to the deterioration of its electronic and thermal properties reflective to GO [17,24].

A much facile approach for attaining stable graphene dispersion is through non-covalent functionalization technique which strongly depends on the short range molecular interaction between dispersant and numerous active sites on graphene. It was reported in the literature that some polymers, surfactant and polyaromatic based molecules were capable of isolating graphene sheets [25]. Researchers have classified the interaction into three main categories namely the π - π , cation- π and van der Waals bonding [26]. Several researchers have recently reported successful functionalization of graphene using aqueous based polyaromatic

organic compound [27]. Polymer and surfactant mediated graphenes have also been proposed by several researchers which rely on van der Waals attraction as the root of stabilization mechanism [28,29].

On the subject of GO reduction via wet chemical approach, specific endeavors have been explored to shift from the ever-reliance use of conventional reducing agent such as hydrazine, dimethylhydrazine, and Sodium Borohydride (NaBH₄) towards environmentally friendly and less toxic chemicals [30,31]. It was known that hydrazine in particular, although highly efficient in oxygen removal of GO, suffers from inherent toxicity owing to its highly corrosive and flammable nature while its carcinogenic disposition poses a major deterrence to prolonged exposure in human [32]. Further, the use of hydrazine may not be compatible and manufacturing friendly in areas such as photovoltaic cell technology where traces of reducing agent may induce detrimental effect to the overall solar absorption capacity [33]. Meanwhile NaBH₄, although being much efficient reductant in comparison to hydrazine based on the C/O ratio [34], produces heteroatoms in the form of alcohol as residual byproduct that are covalently attached to GO during the reduction process. Another persisting challenge on the use of the above chemicals lies on the irreversible aggregation of graphene sheet upon reduction which pronouncedly affect its packing efficiency and thus require further stabilization strategy [14].

Recently, specific attention has been directed on exploring much safer route for producing graphene by employing environmentally benign naturally inspired organic substances [30,35,36]. In this context, based on literature evidences, much emphasis has been given on the use of plant based extracts to enable the restoration of graphene conjugated structure via redox reaction, promoting electron transfer between the active sites of the interacting species [30,37]. Interestingly, the fundamental molecular building block of these plant extracts enables reduction and functionalization to be accomplished simultaneously. This single-pot RGO synthesis and stabilization inspires simplicity and scalability in post processing as well as promoting product compatibility in variety of solvents [31].

Motivated by the above favorable attributes of the so-called 'green reductant' on reducing and stabilizing RGO, the present research would embark on extending the application boundary of water soluble RGO by conducting series of observations and measurements to evaluate the thermophysical properties of the phytochemical reduced GO in order to extend its application boundary within thermal science. On this note, tannic acid (TA) has been employed as the reducing agent to perform oxygen scavenging task in GO. Tannic acid is known to contain abundance of catechol and pyrogallol as its main structural units [30]. In the context of RGO synthesis, the hydroxyl component of these building blocks would actively participate in electron transfer process, leading to a formation of quinones at the end of the reduction process along with H₂O and restored conjugated structure of RGO. Further, the aromatic structure of both RGO and quinone would come into contact via π - π staking interaction to stabilize the RGO.

Also, the present study would indulge in exploring the potential of RGO in offering extended stability to different carbon allotropes by forming RGO based hybrid complexes. This concept of hybridization was previously introduced to enhance the

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