A bio-based, facile approach for the preparation of covalently functionalized carbon nanotubes aqueous suspensions and their potential as heat transfer fluids

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HIGHLIGHTS

• A novel, green method for covalent functionalization of MWCNTs was introduced.
• Highly stable clove-treated MWCNTs aqueous suspension was synthesized.
• The success of functionalization process was validated by characterization techniques.
• The solubility of the synthesized MWCNTs nanofluid was verified by zeta potential and UV–vis spectra.
• The synthesized clove-treated MWCNTs nanofluid showed remarkable thermo-physical properties.

ABSTRACT

In this study, we propose an innovative, bio-based, environmentally friendly approach for the covalent functionalization of multi-walled carbon nanotubes using clove buds. This approach is innovative because we do not use toxic and hazardous acids which are typically used in common carbon nanomaterial functionalization procedures. The MWCNTs are functionalized in one pot using a free radical grafting reaction. The clove-functionalized MWCNTs (CMWCNTs) are then dispersed in distilled water (DI water), producing a highly stable CMWCNT aqueous suspension. The CMWCNTs are characterized using Raman spectroscopy, X-ray photoelectron spectroscopy and transmission electron microscopy. The electrostatic interactions between the CMWCNT colloidal particles in DI water are verified via zeta potential measurements. UV–vis spectroscopy is also used to examine the stability of the CMWCNTs in the base fluid. The thermo-physical properties of the CMWCNT nano-fluids are examined experimentally and indeed, this nano-fluid shows remarkably improved thermo-physical properties, indicating its superb potential for various thermal applications.

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1. Introduction

The electrical, thermal and mechanical properties of carbon nanotubes (CNTs) combined with their small size, low density and high toughness [1–3], give these materials great potential for a wide range of applications including heat transfer and supercapacitors [4–8]. However, CNTs are known to have weak dispersibility in many solvents as a consequence of strong intermolecular \( \pi-\pi \) interactions. This hinders the processability of CNTs in industrial applications [9,10]. In order to tackle this issue, various techniques have been developed over the years to modify the surface of CNTs in order to improve their stability and solubility [7,11]. This enhances processing and manipulation of insoluble CNTs, rendering them useful for synthesizing innovative CNT nano-fluids with impressive properties that are tuneable for a wide range of applications.

Chemical routes such as covalent functionalization have been studied extensively, which involves the oxidation of CNTs via strong acids (e.g. sulfuric acid, nitric acid or a mixture of both) in order to set the carboxylic groups onto the surface of the CNTs as the final product or for further modification by esterification or amination [12–14]. Free radical grafting is a promising technique among covalent functionalization methods, in which alkyl or aryl peroxides [15], substituted anilines, and diazonium salts are used as the starting agents [12]. Free radical grafting of macromolecules (as the functional group) onto the surface of CNTs can improve the solubility of CNTs compared to common acid treatments which involve the attachment of small molecules such as hydroxyl onto the surface of CNTs. Indeed, the solubility of CNTs can be improved significantly by free radical grafting because the large functional molecules facilitate the dispersion of CNTs in a variety of solvents, even at a low degree of functionalization [16]. Even though these chemical routes are generally reliable to synthesize functionalized CNTs, these techniques are not environmentally friendly because the reagents used for synthesis are toxic and detrimental to the environment. More importantly, some of these chemical routes may cause defects in the lattice which will degrade the inherent characteristics of CNTs [12].

Hence, there is a critical need to develop a simple, efficient and environmentally friendly technique for the dispersion of CNTs, which will be greatly beneficial in the long term since it helps reduce pollution resulting from the use of toxic reagents during syntheses [17].

Cloves (i.e. buds of Syzygium aromaticum) are spices commonly used throughout the world and they are known for their valuable medicinal properties due to the presence of active components. The main bioactive compound of cloves is eugenol, which is typically present in concentrations of around 82.6 wt% [18]. Phenolic acid and gallic acid are among the active components of cloves, though at higher concentrations. Previous studies have also shown that ascorbic acid is present in clove flower buds at low concentrations up to 0.08 wt% [19]. The structure and unique properties of clove components make cloves an ideal candidate to enhance the functionalization of CNTs in aqueous media. In this study, we propose a facile, economical, environmentally friendly technique to synthesize highly dispersed multi-walled carbon nanotubes (MWCNTs) in aqueous media using the free radical grafting technique. We characterize the covalent-functionalized MWCNT (hereforth designated as CMWCNTs in the remainder of this article) by Raman spectroscopy, X-ray photoelectron spectroscopy (XPS), transmission electron microscopy (TEM), zeta potential and UV-vis spectra measurements in order to verify the success of the covalent functionalization. Following this, we prepare CMWCNTs nano-fluids by dispersing 0.08 and 0.05 vol% of CMWCNTs in distilled water, which we have selected as the base fluid. Lastly, we measure the thermo-physical properties of the CMWCNT nano-fluid as a function of the fluid temperature and particle concentration to verify the potential of this nano-fluid for thermal applications.

2. Material and methods

2.1. Chemicals and sample preparation

Here, we outline our innovative procedure to prepare the environmentally friendly CMWCNTs. The procedure consists of two primary steps: (1) the preparation of the clove extract solution, and (2) the covalent functionalization of the MWCNTs. In the first step, we purchased the cloves from a local market in Iran and we prepared the clove extract solution (which is a source of eugenol and ascorbic acid [19]) as follows. Firstly, 4 g of ground cloves were added into a vessel filled with 200 ml of distilled water. The process was carried out under heating at 80 °C and the solution was homogenized at 1000 rpm for 30 min. Lastly, the clove extract solution was then filtered through 45 μm PTFE membrane under ambient conditions. Scheme 1 shows a pictorial representation of the clove extract solution preparation procedure.

In the second step, 1 g of pristine MWCNTs (Nanostructured & Amorphous Materials Inc.) was poured into a vessel filled with 200 ml of clove extract solution and then stirred for 15 min. Following this, 7 ml of hydrogen peroxide (30%) (Sigma-Aldrich Co., Selangor, Malaysia) was added drop by drop into the mixture throughout the sonication time. The resultant mixture was ultrasonicated for 10 min, followed by heating up to 80 °C under reflux for 14 h. The suspension was centrifuged at 14,000 rpm and washed with distilled water several times to remove unreacted materials until the solution attained a pH of 7. The functionalized sample was subsequently dried overnight in a vacuum oven at 60 °C. Following this, CMWCNTs-water nano-fluid is prepared by dispersing (10 min ultra-sonication) 0.05 and 0.08 vol.% of covalently functionalized MWCNTs nanoparticles in distilled water. We observed that the CMWCNTs were well-dispersed in the aqueous media.

Scheme 2(a) shows the initiation reaction for the free radical grafting. In the initiation step, the ascorbic acid (i.e. vitamin C) reacts with hydrogen peroxide (a free-radical oxidizer that generates non-toxic by-products and leaves no chemical residue), producing hydroxyl radicals [20]. At high temperatures, the hydrogen peroxide becomes unstable and decomposes spontaneously into hydroxyl radicals. These reactions continue to take place at 80 °C in harmless water [21]. Scheme 2(b) shows that most of the generated hydroxyl radicals will attack eugenol to produce free radicals on the eugenol structure, which leads to the linkage of the activated eugenol onto the surface of MWCNTs. In addition, the hydroxyl radicals can attack the MWCNTs directly, leading to formation of hydroxyl groups on the MWCNT surface.

2.2. Instrumentation

Ultrasonication was conducted via ultrasonic liquid processor (Misonix Inc., Farmingdale, New York, NY, USA) having an output of 600 W. We used Axis Ultra-DLD system, Kratos Analytical X-ray photoemission spectrometer (XPS) with Al Kα X-ray source (hv = 1486.8 eV) to recognize the functional groups in Clove-treated MWCNTs nanoparticles. We used the CASA XPS programme with Gaussian-Lorentzian mix function and Shirley background subtraction for deconvolution of the XPS spectra. Hitachi HT7700 transmission electron microscope (high-resolution digital TEM) was used to examine the morphological characteristics of

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the samples. For the TEM analysis, the samples were prepared by ultrasonically dispersing the nanoparticles in ethanol prior to collection on Lacey carbon grids. The Raman spectra were recorded using a Renishaw inVia Raman with laser excitation at 514 nm. A Zetasizer Nano ZS two-angle particle and molecular size analyser (Malvern Instruments Ltd., Malvern, UK) was used to measure the zeta potential of the nano-fluid samples. The UV Spectrophotometer (UV-1800, Shimadzu Corporation, Kyoto, Japan) was used to evaluate the dispersibility of the nano-fluids. To permit for detectable wavelengths for the UV–vis spectrometer to pass through the samples, a dilution ratio of 1:20 with distilled water was used. Using special quartz cuvettes suitable for the UV region, light absorbance of all samples was measured at different time intervals for 63 days. The thermal conductivity of the samples was measured using a KD-2 PRO portable field and laboratory thermal property analyser (Decagon Devices, USA). The KS-1 probe has a length and diameter of 60 and 1.3 mm, respectively. The accuracy of the thermal conductivity measurements is around 5%. In order to ensure equilibrium of the nano-fluids, an average of 20 measurements were recorded over a 5-h period for each sample and
temperature. Calibration of the instrument was also conducted with DI water prior to the nano-fluid measurement.

The measurements were made in triplicate for each sample and temperature. Specific heat capacity of the samples were evaluated with a differential scanning calorimeter (DSC 8000, Perkin Elmer, USA) with an accuracy of ±1.0%. The viscosity of the nano-fluids was measured using a Physica MCR rheometer (Anton Paar, Austria). The rotational rheometer consists of a moving cylindrical plate and a stationary cylindrical surface placed in parallel with a small gap between them. The density of the samples was measured using a DE-40 density meter (Mettler Toledo, Switzerland), with an accuracy of 10⁻⁴ g/cm³.

3. Result and discussion

3.1. Functionalization analysis

Raman spectroscopy provides fundamental information for us to examine the structure of sp² and sp³ hybridized carbon atoms in carbon-based materials and therefore, we chose Raman spectroscopy at the first characterization technique. In addition, Raman spectroscopy elicits essential information on the covalent functionalization of MWCNTs by following the variations in disorder and defects [22].

Fig. 1(a) shows the Raman spectra of the MWCNT and CMWCNTs. The spectra of pristine MWCNTs and C-MWCNT illustrate D bands, which is related to the amorphous/disordered carbon (sp³) and G band which is due to the graphitic carbon (sp²-hybridized carbon). The peak centre and intensity for the MWCNTs and C-MWCNTs are presented in Table 1. The enhancement in the Raman intensity ratio (I_D/I_G) indicates a change in hybridized carbon from sp³ to sp², which is a direct evidence of covalent functionalization [23]. As can be seen in Table 1, The pristine MWCNTs have an I_D/I_G ratio of 0.36 whereas the CMWCNTs have an I_D/I_G ratio of 0.8. The larger I_D/I_G ratio obtained for the CMWCNTs indicates the presence of a higher number of sp³ carbons and the occurrence of an electrophilic addition reaction, which confirms the successful covalent functionalization of the MWCNTs.

We used X-ray photoelectron spectroscopy (XPS) to examine the nature and elemental compositions of the MWCNTs before and after covalent functionalization with clove leaves and the results are shown in Fig. 1(b). We fitted each peak to the binding energy of standard carbon, i.e. 284.6 eV. It is apparent from the XPS spectra for both the MWCNTs and CMWCNTs that the highest peak intensity occurs at around 284.6 eV (C1s) and 532.8 eV (O1s). Based on these results, we can deduce that the pristine MWCNTs have a very small amount of oxygen (Table 2). However, upon functionalization, the intensity of the O1s peak increases considerably, as indicated by the oxygen to carbon (O/C) ratio, which increases from 0.024 (MWCNTs) to 0.241 (CMWCNTs). This confirms the successful free radical grafting of different oxygen functional groups onto the CMWCNTs.

Fig. 2 shows the high resolution of C1s and O1s XPS results and the deconvoluted peaks (bonding components) for both the pristine MWCNTs and CMWCNTs. The C1s XPS spectrum for the CMWCNTs (Fig. 2(a)) shows that the CMWCNTs have three oxygen functional groups at 285.8, 287.02 and 288.78 eV, which are assigned to the C=O (hydroxyl and epoxy), C=O (carbonyl) and O−C=O (carboxylic acid) bonds, respectively. Table 2 represents an overall and elemental composition and atom percentage of MWCNTs and CMWCNTs. The C1s XPS spectrum for the MWCNTs (Fig. 2(b)) also shows these peaks but their intensities are lower than those for CMWCNTs, indicating that large fractions of oxygen atoms are present as either C=O and O−C=O for the CMWCNTs (Table 2). In addition, the peaks at 284.6 and 290.3 eV (for both the pristine MWCNTs and CMWCNTs) are related to the C=O (aromatic) and π−π interaction bonds, respectively [24–27].

Fig. 2(c, d) shows three oxygen peaks arising from C=O (~530.3 eV), C−O (carbonyl ~352.4 eV) and O−C=O (~533.8 eV) groups for MWCNTs and two oxygen peaks arising from C=O (~530.3 eV) and O−C=O (~532.9 eV) groups for CMWCNTs. After functionalization, the three peaks related to C=O/C=O/O−C=O increases from 1.44%, 1.2% and 0% for MWCNTs to 4.34%, 11.89% and 3.01%, respectively for CMWCNTs. The pristine MWCNTs show general functionalities such as C=O−C and O−C=O, as well as C−OH and C=O in C1s and O1s, respectively (Fig. 2(d)). It is likely that these groups were generated unintentionally when the MWCNTs were exposed to the environment containing natural oxidizing agents such as ozone and hydroxyl radicals [23]. Full XPS analysis data is given in Table 2.

Even though transmission electron microscopy (TEM) does not provide any information on the functional groups, this technique enables us to examine the surface alteration of MWCNTs, which serves as supplementary evidence of successful functionalization. Fig. 3(a–d) shows the TEM images of the pristine MWCNTs and CMWCNTs. In addition, it is evident from Fig. 3(b, c and d) that most of the tips and ends of the CMWCNTs are well-separated, which differs significantly from Fig. 3(a) which shows unscathed MWCNT tips and ends. We believe that this may be due to the conversion of sp² hybridized carbons to sp³ hybridized carbons, which can potentially defect graphitic CNTs.

It is worth noting here that the stability of CMWCNT nano-fluid is closely linked with its electro-kinetic properties. The high
The surface charge density of MWCNTs will generate strong repulsive forces, which helps in the attainment of well-dispersed, stable MWCNT suspensions [28]. Hence, we conducted zeta potential measurements to investigate the electrophoretic behavior and further understand the dispersion behavior of MWCNTs in water.

Fig. 4(a) shows the measured zeta potential values as a function of pH for the CMWCNT aqueous suspension. It can be observed that the CMWCNT nano-fluid has high negative values (−20.7 mV to −42.9 mV) within the pH varies from 2.55 to 10.95. More importantly, the zeta potential values for the CMWCNT nano-fluid are far from the isoelectric point (i.e. point of zero charge), which indicates that this pH range (3.5–10.95) results in strong electric repulsion forces between the CMWCNTs. This prevents aggregation of the CMWCNTs by non-covalent interactions such as π–π interactions or H-bonding. We attribute this phenomenon to the oxygen functionalities at the surface of the CMWCNTs. Hence, the CMWCNT aqueous suspension is stable when the pH is more than 3.5 (where the corresponding zeta potential is around −30.6 mV). We intend to highlight that in this study, the CMWCNT aqueous suspension is stable even in slightly acidic conditions. In more acidic solutions (pH < 3.5), the material tends to agglomerate and undergo intermolecular dehydration catalyzed by H⁺, leading to the coupling of CMWCNTs via ether linkages. We noted that the CMWCNT nano-fluid becomes more stable by adding alkali to the aqueous suspension, which leads to the generation of additional negative charge in the nanoparticles.

### Table 1
Raman spectra analysis results for pristine and clove-treated MWCNTs.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Peaks</th>
<th>Centre (cm⁻¹)</th>
<th>Intensity (a.u.)</th>
<th>I_D/IG</th>
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<tr>
<td>MWCNTs</td>
<td>D band</td>
<td>1339.63</td>
<td>2253.20</td>
<td>0.36</td>
</tr>
<tr>
<td></td>
<td>G band</td>
<td>1595.67</td>
<td>6118.35</td>
<td></td>
</tr>
<tr>
<td>CMWCNTs</td>
<td>D band</td>
<td>1353.56</td>
<td>5454.51</td>
<td>0.80</td>
</tr>
<tr>
<td></td>
<td>G band</td>
<td>1586.29</td>
<td>6825.58</td>
<td></td>
</tr>
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</table>

### Table 2
XPS analysis results for pristine and clove-treated MWCNTs.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Overall Percentage (atom%)</th>
<th>Elemental composition (atom%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>C1s</td>
<td>O1s</td>
</tr>
<tr>
<td>MWCNTs</td>
<td>97.09</td>
<td>2.91</td>
</tr>
<tr>
<td>CMWCNTs</td>
<td>80.56</td>
<td>19.44</td>
</tr>
</tbody>
</table>

Fig. 2. C1s XPS spectra for (a) pristine MWCNTs and (b) CMWCNTs, and O1s XPS spectra for (c) pristine MWCNTs and (d) CMWCNTs.
Fig. 3. (a) TEM images of pristine MWCNTs and (b–d) CMWCNTs.

Fig. 4. (a) Zeta potential values of the C-MWCNT nano-fluid as a function of pH, (b) colloidal stability of Clove-treated MWCNTs dispersed in DI water, and (c) photograph of non-functionalized and Clove-functionalized MWCNTs dispersed in DI water after three months.
Colloidal stability of CMWCNTs nano-fluids were recorded via UV–Vis spectra and the data are plotted in Fig. 4(b). It is clear that the relative concentration of the nano-fluids reduces with the number of the days. However, the nano-fluids have a fairly constant concentration after Day 38. The maximum magnitude of sedimentation is ~11%, which confirms the colloidal stability of the nano-fluids containing CMWCNTs. Digital images of both pristine and clove-functionalized MWCNTs (CMWCNTs) dispersed in DI water after 90 days, are represented in Fig. 4(c).

3.2. Thermo-physical properties

We determined the thermo-physical properties of the CMWCNTs nano-fluids as a function of the fluid temperature and nanoparticle concentration from experiments and the results are shown in Fig. 5(a–d). The measured thermal conductivity of the DI water shows excellent agreement with the NIST data [29], whereby the error is less than 1%. Fig. 5(a) clearly shows that the thermal conductivity of the CMWCNT nano-fluids is significantly higher than that for DI water and this thermal conductivity increases with an increase in both fluid temperature and nanoparticle concentration. We believe that the main mechanism underlying the thermal conductivity enhancement of the CMWCNT nano-fluid is the Brownian motion of the suspended nanoparticles [30], which increases at higher temperatures. The nanoparticle agglomerates will break down more easily by the loading temperature and thus, the CNTs will disperse more uniformly in the water. This in turn, improves heat conduction in the nano-fluid. Furthermore, the random movement of the nanoparticles increases with an increase in temperature and consequently, the heat energy is transmitted more faster in the fluid [31]. Interestingly, we obtained a maximum thermal conductivity enhancement of ~20% at 50 °C.

Fig. 5(b) shows that the effective viscosity of the CMWCNT nano-fluids is higher than that of DI water, though the difference in the values is not marked. Moreover, it is obvious that the viscosity of the CMWCNT nano-fluid decreases with an increase in fluid temperature, which is close to that for DI water. We attribute this trend to the weakening of intermolecular forces with increasing temperature [30]. The slight increase in the effective viscosity upon the addition of CMWCNTs at a low particle loading is an important advantage of the CMWCNT nano-fluid since a higher viscosity will undermine the overall positive impact of enhanced thermal conductivity in heat transfer systems due to the pumping fluid penalty [32]. We also measured the density of the CMWCNT nano-fluid and DI water as a function of temperature and the results are shown in Fig. 5(c). We observed that the density of the CMWCNT nano-fluid and the base fluid decreases slightly with increasing temperature, which is due to the thermal expansion of the liquid. The density of the nano-fluid decreases by approximately 0.6% when the fluid temperature increases from 20 °C to 40 °C for a particle loading of 0.08 vol%. There is a slight enhancement in the density for the CMWCNTs nano-fluids with an increase in nanoparticle concentration. The increase in density can be attributed to the density of the CMWCNTs, which is higher than that of the base fluid. Hence, increasing the particle loading will increase the density of the nano-fluid. In addition, it can be seen from Fig. 5(d) that the specific heat capacity of the CMWCNT nano-fluid and base fluid increases in response to the increase in fluid temperature. The slope is similar for the specific heat capacity curves, which conform to the findings of Pak and Cho [33,34]. Additionally, the experimentally measured dynamic viscosity data is depicted in Fig. 6(a–c) as

![Fig. 5](image_url)
the functions of shear rate and temperature for CMWCNTs aqueous suspensions and DI water. Fig. 6(a–c) clearly indicates that all nano-fluids similar to water exhibit Newtonian behavior with almost constant viscosity with respect to increasing shear rate. Therefore, from the thermal transport perspective, this material is deemed suitable for enhancing heat transfer performance at negligible pumping penalty.

4. Conclusions

In this study, we have successfully developed a facile, bio-based, environmentally friendly technique to synthesize well-dispersed clove-functionalized multi-walled carbon nanotubes (CMWCNTs). Our results have proven that the technique improves the thermo-physical properties and stability of MWCNTs in aqueous media. We have verified the success of the covalent functionalization procedure by Raman spectroscopy, X-ray photoelectron spectroscopy and transmission electron microscopy. We have also provided further evidence of the hydrophilic functionalized groups for the CMWCNTs by zeta potential measurements. Based on the UV–vis absorption spectra, the CMWCNTs nano-fluids have remarkable stability over the course of 63 days after preparation. The measured thermal conductivity of the DI water shows excellent agreement with the NIST data [29], whereby the error is less than 1%. Our results are very encouraging since we obtained a low viscosity of loading (0.08 vol%) of CMWCNTs in DI water. More importantly, we found that the viscosity and density of the CMWCNT nano-fluid show close resemblance to those for water. Based on the results, we conclude that this environmentally friendly nano-fluid has superb potential for use in heat transfer systems which require heat transfer fluids with high thermal conductivity and low viscosity.

Acknowledgments

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