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Optical properties of carboxyl functionalized carbon nanotube aqueous nanofluids as direct solar thermal energy absorbers

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Abstract

Nanoparticle suspensions in liquids have shown remarkable changes in thermo-physical and optical properties of their base media. In this study, carbon nanotubes are chemically functionalized in order to overcome their inherent hydrophobic nature and make them dispersible in a polar liquid such as water. Dispersions of pristine and functionalized carbon nanotube nanofluids in deionized water were prepared and their optical properties at room and elevated temperatures were measured. Quantitative analysis using absorption spectroscopy demonstrates higher radiation absorption of carbon nanotube dispersions with respect to pure water which is decreased by increasing temperature. Optical characterization shows that functionalized carbon nanotube nanofluids show remarkable stability after undergoing thermal cycling and after extended periods of time (up to three months) in comparison to pristine carbon nanotube nanofluids.
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1. Introduction

In recent years nanofluids have gained significant interest because of their ability to enhance thermo-physical and radiative properties of commonly used heat transfer fluids in various heat exchange systems. Of particular interest here is the ability of nanoparticles to significantly enhance the radiative properties of pure base fluids. This phenomenon motivated the concept that nanofluids can be used as direct volumetric absorbers for solar radiation, leading potentially to more efficient solar energy conversion compared to typical surface absorbers. The potential use of nanofluids in direct absorption solar thermal collectors has been explored extensively (Tyagi et al., 2009; Otanicar et al., 2010; Lenert et al., 2010; Taylor et al., 2011; Lenert and Wang, 2012; Lee et al., 2012). Optical properties of nanofluids have been the subject of much recent study with more focusing on room temperature measurements (Mu et al., 2009; Zhu et al., 2013; Sani et al., 2010; Taylor et al., 2011; Kameya and Hanamura, 2011; Han et al., 2011; Karami et al., 2014). Since liquid-based solar collectors can operate anywhere from 50 °C to 500 °C- based on their working fluid boiling point, evaluation of these properties at elevated temperatures is of great importance. A limited number of studies focused on temperature dependent optical properties of nanoparticles (Kreibig, 1974; Link and El-Sayed, 1999).

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The effect of temperature variation on optical behavior of nanoparticle suspensions within a base fluid was recently investigated by Otanicar et al. (2012) (in a range of 25–70 °C) and (Otanicar et al., 2013) (up to 45 °C). Their studies demonstrated how increased temperature results in decreased absorption. Among nanoparticles, carbon nanotubes (CNTs) appear very promising in enhancing radiative properties when suspended in heat transfer fluids. CNT nanofluids have high spectral absorptivity over the majority of the solar spectrum and consequently, these nanofluids can absorb almost 100% of the irradiation with relatively low CNT concentrations (Hordy et al., 2014). Spectral absorbance of carbon nanohorn-based nanofluids (CNHs) Sani et al., 2010, 2011, multi-walled carbon nanotube nanofluids (MWCNTs) Hordy et al., 2014 and alkaline functionalized carbon nanotubes (Karami et al., 2014) were studied.

In most of the applications proposed in the literature, nanoparticle suspensions will experience some amount of heating and thermal cycling. At high temperatures, nanoparticles have an increased chance to collide and agglomerate (Otanicar et al., 2013). The unique properties of the suspension, attributed to small size and homogeneous dispersion of the particles, are lost if the nanoparticles agglomerate and settle out over time and under thermal cycling (Hordy et al., 2014).

Typically, nanoparticles in a base fluid are stabilized through the use of a surfactant (Yu and Xie, 2012; Taylor et al., 2013). However, one well-known issue is irreversible deterioration of the surfactant at rather modest temperatures as low as 60 °C (Ghadimi et al., 2011), whereas an appropriate solar nanofluid must remain stable at elevated temperatures. Dispersion stability of nanofluids at high temperature with respect to their optical applications has seen limited academic study. Mercatelli et al. (2011) studied particle aggregation over five cycles of heating and cooling for single-wall carbon nanohorns dispersed in water and glycol. For water suspensions good stability was observed for temperatures up to 120 °C whereas for glycol dispersions a higher stability, up to 150 °C was detected. Otanicar et al. (2013) performed thermal cycling experiments (up to 80 °C) for aqueous silica and gold nanofluids. They observed that theses nanofluids exhibit good stability and only soft agglomerates were present after over 200 thermal cycles. One disadvantage of carbon nanotubes is their insolubility in any solvent. Only suspensions of CNTs can be produced which tend to agglomerate at elevated temperatures and under thermal cycling. Attachment of functional groups to the nanotubes can cause the particles to favor interactions with the host liquid and dramatically increase the solubility of nanotube material (Dettlaff-Weglikowska et al., 2002). In order to obtain a fine dispersion of CNTs in the selected solutions or matrices, several methods have been developed including covalent or non-covalent modifications which were extensively reviewed by Meng et al. (2009). Among these studies, stable aqueous MWCNT nanofluids were synthesized by surface functionalization via radio-frequency (RF) glow discharge plasma treatment (Vandsburger et al., 2009; Hordy et al., 2013). Plasma treatment has been shown to covalently graft oxygenated functionalities, such as carboxylic groups onto the surface of the MWCNTs, thus rendering them polar and highly dispersible in water (Hordy et al., 2013). High-temperature stability of these plasma-functionalized MWCNTs nanofluids were quantitatively examined by Hordy et al. (2014). They found that no agglomeration occurred after heating to 85 °C and 170 °C for the aqueous and glycol based nanofluids, respectively.

Possible chemical functionalization of carbon nanotubes make them interesting for increasing the overall efficiency of direct absorption solar thermal collectors. In the course of the work reported here, to overcome their inherent hydrophobic nature, carbon nanotubes were chemically functionalized by acid treatment. This process covalently bonds polar functionalities such as carboxylic groups onto the single-walled carbon nanotubes (SWCNT) surface, making them dispersible in water. The presence of carboxyl groups was proved by scanning electron microscopy (SEM) and Fourier transform infrared (FTIR) measurements. Dispersions of pristine and functionalized carbon nanotube nanofluids in deionized water in various concentrations were prepared and their optical properties at room temperature were evaluated and compared. Samples with maximum absorption values were selected for temperature dependent optical measurements, thermal cycling and long term stability testing. In addition to measuring the optical properties, the particle size distributions of the as-prepared and thermal treated colloidal suspensions were measured using a dynamic light scattering (DLS) system. Based on the measurements, in comparison with pristine SWCNT nanofluids, acid functionalized SWCNT dispersions show better stability at elevated temperatures, after thermal cycling and after long term settling. Making them very promising for use in direct absorption solar thermal energy collectors.

2. Experimental

2.1. Materials

Singe wall carbon nanotubes with well defined structure i.e. 1.2–1.5 nm average diameter and 2–5 μm average length were ordered with purity above 90% on the gram scale from Sigma–Aldrich. Polyethylene glycol (PEG) 4000, nitric acid (65%, AR grade), hydrochloric acid (37% AR grade) and sulfuric acid (98%, AR grade) were purchased from Merck Millipore.

2.2. Functionalization of SWCNT

In order to functionalize the pristine carbon nanotubes (P); two acid treatment procedures were performed. In the first method (F1), 50 mg of SWCNTs powder was dispersed in aqua regia; a solution of concentrated nitric and

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