



Preparation and photo-thermal conversion performance of modified graphene/ionic liquid nanofluids with excellent dispersion stability



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ABSTRACT

Dispersion stability has been long considered as a critical issue for applying nanofluids in various fields, especially for the applications at elevated temperatures. Herein a novel route is explored to improve the dispersion stability of graphene (GE)/ionic liquid (IL) nanofluids for use as working fluids in medium- and high-temperature direct absorption solar collectors (DASCs), which involves modifying GE according to the molecular structure of the IL. Specifically, GE was modified using the reagents and process for synthesizing [HMIM]BF₄, followed by dispersing the modified GE (MGE) into [HMIM]BF₄. It is verified that the molecular chains similar to [HMIM]BF₄ have been grafted on the nanosheets of GE, and the MGE/[HMIM]BF₄ nanofluids exhibit much better dispersion stability than the one containing the unmodified GE, even at elevated temperatures. Moreover, the temperature profiles of the nanofluids containing MGE and GE were obtained both from the experimental measurement and the theoretical prediction using a one-dimensional transient heat transfer model. It is shown that the experimental data are in good agreement with the numerical ones for the MGE nanofluids, while a large deviation between them is found for the one containing the unmodified GE. And the MGE nanofluid shows enhanced receiver efficiency as compared to the GE one due to its much improved dispersion stability. Further, the transient model was used to predict the performance of the MGE nanofluid based DASCs under high solar concentrations. And by integrating the MGE concentration and the receiver height into a parameter, namely optical thickness, the optimization of the MGE nanofluid based DASC was carried out varying solar concentration, MGE concentration, nanofluid height and exposure time. It is revealed that the photo-thermal conversion performance of nanofluids greatly depends on its dispersion stability at elevated temperatures, and the MGE/[HMIM]BF₄ nanofluids possess excellent dispersion stability and show great potentials for use as the working fluids in DASCs. This work sheds light on effective routes for improving dispersion stability of nanofluids as well as numerical investigations on nanofluid based DASCs.

1. Introduction

Nanofluid refers to a new kind of heat transfer fluids (HTFs), prepared by dispersing a little amount of nanomaterials into conventional working media. Since this kind of HTFs was coined by Choi [1] in 1995, nanofluids have been demonstrated to exhibit enhanced thermal conductivity [2–5] and convective heat-transfer coefficient [6–8] as compared to the corresponding base fluids. And these good thermal characteristics make the nanofluids show potentials for use in various fields, such as vehicular and avionics cooling systems in the transportation industry [9,10], hydronic heating and cooling in buildings [11,12], and industrial process heating and cooling systems in petrochemical [13], textile [14], pulp [15], food [16] and other processing plants. However, due to the thermodynamic instability inherent in suspensions,

nanofluids suffer from the aggregation of the dispersed nanoparticles, which results in the decrease in their thermo-physical characteristics along with the clogging in practical heat transfer equipment [17–19]. Consequently, dispersion instability has long been a critical issue for nanofluids, greatly limiting their practical applications. Apparently, developing nanofluids with good dispersion stability, even at elevated temperatures, is a fundamental and challenging subject.

In order to improve the dispersion stability of nanofluids, several approaches have been explored. Addition of surfactant is a general route for increasing the dispersion stability of nanoparticles in aqueous suspensions. Popular surfactants that have been used in literature include sodium dodecylsulfate (SDS) [20–22], sodium dodecyl benzene sulfonate (SDBS) [23], and cetyltrimethylammoniumbromide (CTAB) [24]. With the aid of the surfactants, the hydrophobic surfaces of

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Nomenclature

P_l	density of fluid [kg m^{-3}]
C_{pl}	specific heat of liquid [$\text{J g}^{-1} \text{K}^{-1}$]
C_{pn}	specific heat of nanofluid [$\text{J g}^{-1} \text{K}^{-1}$]
C_{pb}	specific heat of basefluid [$\text{J g}^{-1} \text{K}^{-1}$]
k_l	thermal conductivity of liquid [$\text{W m}^{-1} \text{K}^{-1}$]
q	heat flux [W m^{-2}]
η	relative thermal storage capacity
T	temperature [K]
η_1	receiver efficiency [%]
t	time [s]
m	weight of nanofluid [kg]

G_s	irradiance of the solar simulator [W m^{-2}]
A	surface area of receiver [m^2]
$Q_{nanofluid}$	heat storage capacity of the nanofluid [J]
$Q_{basefluid}$	heat storage capacity of the basefluid [J]
H	nanofluid height [m]
$m_{p,n}$	weight of nanofluid [kg]
$m_{p,b}$	weight of basefluid [kg]
$K_{n,\lambda}$	extinction coefficient of nanofluid [cm^{-1}]
$K_{b,\lambda}$	extinction coefficient of basefluid [cm^{-1}]
M_f	slope
f	mass fraction of MGE
τ	optical thickness

nanoparticles could be modified to become hydrophilic or vice versa for non-aqueous liquids. However, the bonding between surfactants and nanoparticles would be damaged at the temperatures above than 60 °C [25]. Consequently, the nanofluids lose their stability, leading to the sedimentation of nanoparticles at the elevated temperatures. Besides, controlling pH is another method to improve the dispersion stability of nanofluids, since the stability of nanofluids directly links to their electrokinetic properties. Xie et al. [26] reported that a carbon nanotube suspension gained a good dispersion stability in water through a simple acid treatment on the carbon nanotubes and found that the good stability was attributed to a hydrophobic-to-hydrophilic conversion on the surfaces of the carbon nanotubes. Lee et al. [27] worked on the nanofluids containing Al_2O_3 at different pH values. Their experimental results indicated that, when the nanofluids had a pH of 1.7, the agglomerated particle size was reduced by 18%; and when the nanofluids had a pH of 7.66, the agglomeration size was increased by 51%. However, although the addition of surfactants and the adjustment of pH play an effective role in improving the dispersion stability of nanofluids, the damage of the bonding between surfactants and nanoparticles at the temperatures above than 60 °C and the decrease in zeta potential with an increase in temperature make these two routes inapplicable to the nanofluids for medium- and high-temperature applications [28]. And it has been reported that high temperatures intensified the agglomeration of nanoparticles, resulting in the remarkable decrease in thermal conductivity of the nanofluids [29]. Therefore, new approaches need to be explored for preparing nanofluids with good dispersion stability at elevated temperatures.

Ionic liquids (ILs) are a group of molten salts with a wide liquid temperature range, high density and heat capacity, good thermal and chemical stability and low vapor pressure [30,31]. These favorable thermophysical properties make ILs show great promise to be used as HTFs for medium- and high temperature applications. And it has been reported that the ILs based nanofluids, prepared by dispersing nanoparticles into ILs, show enhanced thermal conductivity, making them more promising for use as HTFs [32,33]. In our previous work, graphene (GE), a 2D carbon nanomaterial with extraordinarily high thermal conductivity [34], was dispersed into ILs to prepare the GE/IL nanofluids, and their thermophysical properties were measured in wide temperature range [35]; Further, the optical absorption property and photo-thermal performance of the GE/IL nanofluids as well as the performance of the GE/IL nanofluid based direct absorption solar collectors (DASCs) were experimentally and numerically investigated [36,37]; The obtained results show that graphene is an excellent nanoadditive for preparing nanofluids, and the GE/IL nanofluids are a kind of promising working fluid for medium- and high temperature DASCs; However, it has been also found that the agglomeration of GE increased with temperature during the GE/IL nanofluids were irradiated, leading to the decrease in the photo-thermal performance of the GE/IL nanofluid. Therefore, in view of the strong influence of dispersion stability of the IL-based nanofluids on their thermophysical

properties and photo-thermal performance, it is urgent to prepare the GE/IL nanofluids with good dispersion stability at elevated temperatures for their practical applications.

In this work, to make GE disperse stably into 1-hexyl-3-methylimidazolium tetrafluoroborate ([HMIM]BF₄), a novel method for preparing the nanofluids was explored, which is quite different from the previously-reported approaches. First, GE was modified according to the molecular structure of [HMIM]BF₄ by grafting the molecular chains similar to [HMIM]BF₄ on its surface using the reagents and the process for synthesizing [HMIM]BF₄. The morphology and structure of MGE were characterized. Second, the [HMIM]BF₄ based nanofluids were prepared by dispersing the modified GE (MGE) sample into the IL at different loadings. The dispersion stability of the [HMIM]BF₄ based nanofluids was investigated in the temperature range from room temperature to 180 °C, and their thermal conductivity, specific heat and transmission spectra were measured. Then, the performance of the DASCs based on the MGE/[HMIM]BF₄ nanofluids were theoretically and experimentally investigated. Specifically, a transient one-dimensional numerical model was used to predict the temperature distribution of the MGE nanofluids under an irradiation of 2300 W m^{-2} based on the extinction coefficients of the nanofluids. And a cylindrical simulative receiver was set up to experimentally measure the temperature profiles of the DASCs under the irradiation with the same incident light intensity. The obtained experimental results were compared with the aforementioned numerical ones to validate the model. Further, the model was employed to predict the temperature profiles of the MGE/[HMIM]BF₄ based DASCs with various geometrical parameters under different operating conditions. And thus the effects of the parameters including the MGE concentration, the receiver height and the solar concentration on the thermal storage capacity of the DASCs were systematically investigated. What's more, the MGE concentration of the nanofluid and the receiver height of the DASC were integrated into one parameter, namely optical thickness, and the optical thickness was optimized under the fixed solar concentration. Finally, the exposure time was optimized based on the optical thickness, and the systematical investigation along with optimization on the MGE/[HMIM]BF₄ based DASCs were conducted under the concentrated solar incident radiation. It is found that the MGE/[HMIM]BF₄ nanofluids possess good dispersion stability at elevated temperatures due to the good compatibility of MGE with [HMIM]BF₄, and the optimized nanofluid based DASC exhibits large thermal storage capacity, making it show great potentials for practical applications. This work sheds light on the approaches for increasing dispersion stability of nanofluids and makes an improvement on numerical investigations on nanofluids based DASCs.

2. Experimental section

2.1. Materials and reagent

Carboxyl graphene (GE-COOH) was purchased from Nanjing

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