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Fabrication and thermo-physical characterization of silver nanofluids: An experimental investigation on the effect of base liquid

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ABSTRACT

Nanofluids (NFs) are solid-liquid composites prepared by stabilizing nanoparticles (NPs) in a base liquid, which is selected based on the technological area of application. For heat exchange applications the base liquid can be specified as water, ethylene glycol (EG), or their mixture. NFs have exhibited some potential to replace conventional heat transfer fluids due to enhancement of their thermal characteristics. The thermo-physical properties of NFs including thermal conductivity (TC) and viscosity may be affected by several factors including the base liquid, which is not well studied in the literature. Focus of the present work is to study the impact of base liquid by comparing the TC and viscosity of a commercial silver (Ag) NFs with lab-made water, EG and water/ethylene glycol (W/EG) mixture (50:50 by wt%) at different Ag NP loadings (1, 1.5 and 2 wt%). For this purpose, commercial water based Ag suspension (containing 1 wt% Ag NP) was acquired, which is used for the preparation of Ag NFs with different base liquids and NP loadings. Finally, the thermo-physical properties of NFs including TC and viscosity were measured and analyzed at 20 °C. The results revealed that W/EG based NFs containing 2 wt% Ag NP showed best performance with the highest TC enhancement of 12.4% and only 6.1% increase in viscosity, revealing that among different base liquids, W/EG based NFs are the most beneficial for heat transfer applications.

1. Introduction

Heat transfer fluids play important role in many industrial processes to remove excess heat. There are some challenges in conventional heat transfer fluids which must be solved to have effective cooling fluids. For instance, Water (W), ethylene glycol (EG) or their mixture (W/EG) as conventional heat transfer fluids have poor thermal conductivity (TC). One other challenge is about developing effective cooling methods for some high-tech applications for which traditional heat transfer fluids are not able to offer more effective solution. Transport, microelectronics, energy supply, microchannels and biomedical applications are some examples which have become priorities for the further development of effective heat transfer fluids [1–8]. Thus, there is a demand for efficient heat transfer fluids to solve these challenges. In the last decade nanofluids (NFs), which are nanotechnology based suspensions containing nano sized particles; nanoparticles (NPs) in conventional cooling liquids (i.e. base liquids), such as water EG and their mixture, have attracted attention due to their potential benefits in heat transfer applications [9]. Up to now several groups of materials and base liquids have been used to engineer NFs. Metal NPs such as Cu [10], Au [11] and Ag [12–14], ceramic compounds including oxide particles such as Al_2O_3 [15], TiO_2 [16], CeO_2 [17], mesoporous SiO_2 [18], or carbides such as SiC [19–21]. Two major techniques are commonly utilized to fabricate NFs; (i) one-step preparation method [22] which represents the direct formation of NPs inside the base liquids, and (ii) two-step method [23] wherein NPs are at first synthesized/acquired and then dispersed in the base liquids. NF system is a complex suspension which means that it's not simple dispersion of NP and base liquid and its characteristics such as thermo-physical properties including TC and viscosity maybe influenced by several factors, including composition and loading of NP and type of base liquid [24–25], morphology of NP (size and shape) [26], stability of suspension [27], NF fabrication method [28], and surface modifier [29]. Among all these factors, although the base liquids play a critical role by affecting TC and viscosity of NFs, its impact (base liquid effect) has not been studied in sufficient detail. A detailed literature survey showed that not only the number of studies reporting the impact of base liquid on thermo-physical properties of NFs (including TC and viscosity) is limited but also its effect on TC and viscosity of NFs is still under debate. Xie et al. [30–31] studied NFs with Al_2O_3 NPs in different base liquids and demonstrated that

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enhancement in TC of NFs take places if base liquid with lower TC is employed. Tsai et al. [32] reported the alteration of the base liquid viscosity (from 4.2 to 5500 cP, by mixing two base liquids with approximately the same TC) which resulted in a decrease in the TC of the Fe_2O_3 NFs as the viscosity of the base liquid increased. Timofeeva et al. [33–34] studied SiC NFs in water (W) and W/EG mixture with controlled concentration, particle sizes and pH. Their investigations revealed that the TC enhancement is higher in W/EG based NFs compared to the same NFs with water base liquid. We recently reported on water and W/EG based NFs containing α-SiC NPs to demonstrate the effect of base liquid on TC and viscosity of NFs [19–20]. Based on findings at 20 °C, NFs with W/EG base liquid exhibited more favorable performance than the similar water based NFs. In other research work, we studied the effect of base liquid on thermo-physical properties of ethylene and diethylene glycol (EG and DEG) based copper NFs containing various Cu NP loading (1, 2 and 3 wt%). Based on our results, EG based NFs exhibited more favorable characteristics than that of DEG based suspensions at the same Cu NP loading and temperature [35]. There is still a serious need to perform systematic investigation to study the effect of base liquid on thermo-physical properties of NFs. Ag nanoparticles (Ag NPs) have attracted great interest due to their expected high TC as well as excellent chemical and physical stability [14]. Although, there are reports on Ag NFs [12–14], there is no study on the (experimental and theoretical) impact of base liquid on thermo-physical properties of Ag NFs including TC and viscosity. For this purpose, we designed systematic experiments to study effect of base liquid on TC and viscosity of Ag NFs. In this work, our aim is to fabricate Ag NFs with different base liquids and various Ag NP loadings, and investigate the effect of base liquid and NPs loading on the thermo-physical properties of NFs including TC and viscosity. For this purpose, stable Ag NFs with different base liquids (water, W/EG and EG) and various NPs loading (1, 1.5 and 2 wt%) were fabricated. The thermo-physical properties of NFs including TC and viscosity were measured at 20 °C and our findings are presented in detail.

2. Experimental

2.1. Materials and methods

Commercial water based suspension containing 1 wt% Ag NP was purchased and NFs with 1.5 and 2 wt% were prepared using original suspension. Ethylene glycol (EG) was acquired from Sigma Aldrich (Germany). All the suspensions/liquids were used as received, without further purification.

2.2. Fabrication of Ag NFs

In order to fabricate NFs with different base liquids and various Ag NPs loading, water based suspension containing 1 wt% Ag NP (as original sample) was used. For fabrication of water based NFs at higher Ag NP loading (1.5 and 2 wt%), the centrifuge and sonication were used. To keep sample history as well as the particle size the same, NFs with higher concentrations were obtained by ultracentrifuging and concentrating these samples. It should be noted that the ultracentrifuge technique is a well-known method which can be used for separating and precipitating suspended NPs in suspensions. For example, to prepare 25 mL of Ag NF with 2 wt% Ag NP loading, 50 mL of NF at NP loading of 1 wt% was centrifuged at 10000 rpm for 10 min at 20 °C. It was observed that the sample was separated in two phase including the precipitated Ag NPs and liquid free from NPs. Then 25 mL of the solvent decanted to concentrate Ag NFs. New NF with higher concentration of Ag NPs was sonicated, for dispersing the NPs, and evaluated for TC and viscosity properties. The same procedure was followed to fabricate NFs with other NP loading. The EG based NFs containing 1, 1.5 and 2 wt% Ag NP loading also were fabricated using original suspension using the same method. Removal of water from the Ag NPs is necessary as water has higher TC than EG and the presence of water may result in incorrect TC data. Therefore, the NPs obtained were finally dispersed in ethylene glycol under an argon environment in a glovebox. Finally, W/EG based Ag NFs with different Ag NP loading of 1, 1.5 and 2 wt% were prepared by mixing 50% (by wt%) of each water and EG based Ag NF systems. All fabricated NFs (Table 1) were stable for six months without any visual precipitation.

Table 1
Ag NFs with different base liquids evaluated for TC and viscosity properties.

Sample	NP	Base liquid	NP loading (wt%)	pH
Ag NF-EG-1 wt%	Ag	EG	1	–
Ag NF-EG-1.5 wt%	Ag	EG	01.5	–
Ag NF-EG-2 wt%	Ag	EG	2	–
Ag NF-W-EG-1 wt%	Ag	W/EG[a]	1	6
Ag NF-W-EG-1.5 wt%	Ag	W/EG[a]	1.5	6
Ag NF-W-EG-2 wt%	Ag	W/EG[a]	2	6
Ag NF-W-1 wt%	Ag	W	1	6
Ag NF-W-1.5 wt%	Ag	W	1.5	6
Ag NF-W-2 wt%	Ag	W	2	6

[a] Water/ethylene glycol mixture (50:50) by wt%.

2.3. Characterization techniques

Microstructure and morphology of Ag NPs were evaluated by using Scanning Electron Microscopy (SEM; FEG-HR Zeiss-Ultra 55). Transmission Electron Microscopy (TEM) analysis of the particles was carried out using JEOL 2100 at 200 kV acceleration voltage. Zeta potential analysis of Ag NPs was performed for evaluating NFs stability region; average hydrodynamic/solvodynamic particle size distribution of Ag NPs was evaluated by Beckmann-Coulter Delsa Nano C system. The TC measurements of NFs at various NP loading was measured using KD2 Pro thermal property analyzer, based on the principle of transient hot wire (THW) method [36]. The KS-1 sensor was oriented vertically in the sample and the measurements were recorded at low power mode with read time of 1 min. All TC measurements of NFs were carried out at 20 °C and repeated for three times for each NF systems. The validity of the THW instrument was checked by comparing with a standard source for thermodynamic properties of water (IAPWS reference) [37]. Compared to the reference the accuracy of measurement for water was within 2.5%. Finally, the viscosity of NFs was assessed using DV-II + Pro-Brookfield viscometer and the validity of this equipment was also verified using distilled water.

3. Results and discussion

3.1. Morphology analysis

The structure and morphology of NPs was investigated by SEM analysis. Agglomerated spherical NPs with average diameter of 40 ± 5 nm were observed. Microstructure and electron diffraction pattern of Ag NPs were analyzed by TEM which revealed Ag NPs have near spherical morphology with an estimated average particle size of 25 ± 4 nm from the TEM micrographs. Selected area electron diffraction (SAED) pattern displayed in inset images in Fig. 1(b), was indexed for Ag (ICDD no: 05-0664), reveals crystalline nature of the Ag NPs.

3.2. Zeta potential/dynamic light scattering (DLS) analyses

Characterizing NPs dispersions and understanding the role of various parameters, which may affect colloidal properties, are important for any NF system. The pH of a suspension has important role not only on the rheological property of suspension but also in terms of fabrication of stable suspension, which is related to the electrostatic charge on

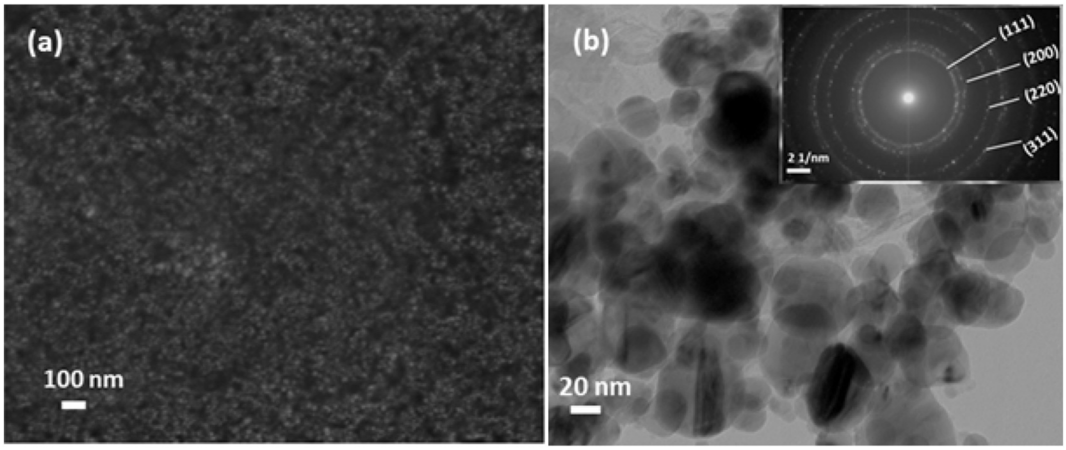

Fig. 1. (a) SEM micrograph of Ag NPs and (b) TEM micrographs of Ag NPs (inset: SAED pattern).

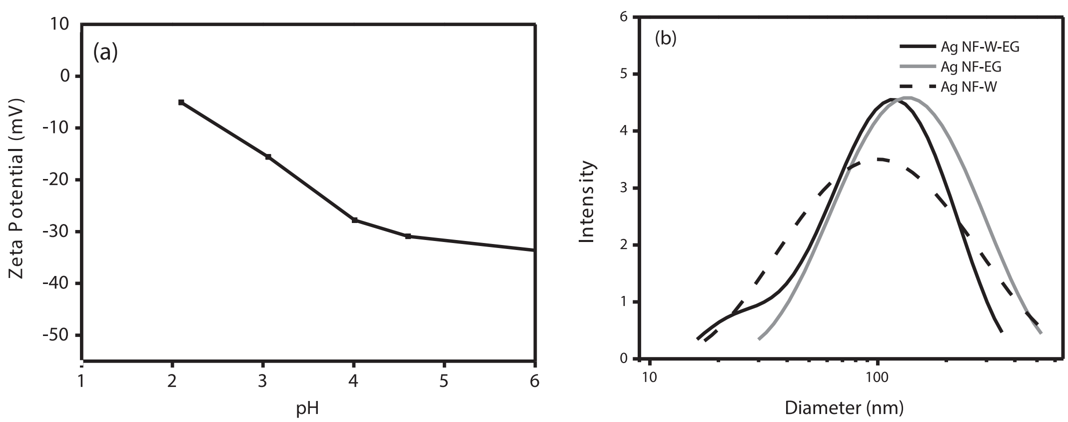

Fig. 2. (a) Zeta potential as a function of pH for Ag NPs, and (b) Particle size distribution of Ag NPs in water and W/EG and EG base liquids measured by DLS.

particles. Zeta potential analysis was performed in the pH region from 2 to 6 to study the stability region of NFs and the results are shown in Fig. 2(a). Based on that, a stable suspension with a strong negative charge is achievable in the pH range between 4 and 6. In order to understand the influence of effective size of dispersed silver NPs in the liquid media, DLS analysis was carried out to estimate the dispersed size of Ag NPs in liquid medium. The results for Ag NPs in W, W/EG and EG media are displayed in Fig. 2(b). Solvodynamic size for Ag NPs in EG, W/EG as well as water media was estimated to be in the range of 30–565, 16–350 and 15–650 nm with an average hydrodynamic/solvodynamic size of 175, 110 and 165 nm, respectively. The difference between primary size obtained from SEM/TEM and the size estimated by DLS is ascribed to surface modifiers in the commercial suspensions, which may have caused particle clustering/agglomeration. The results also reveal that the size of agglomerates is smaller in case of W/EG base liquid.

3.3. TC and viscosity measurements of Ag NFs

TC and viscosity are the two main thermo-physical characteristics representing the performance of heat transfer fluids. It should be mentioned that to have an efficient NF for heat transfer applications, fabrication of NF with higher TC enhancement and minimal increase in viscosity (over the base liquid) is highly desired [38]. Thermo-physical properties of Ag NFs including TC and viscosity were tested at 20 °C. Each data point represents the average of three measurements at a particular NP loading at 20 °C. Fig. 3(a) displays the TC enhancement values vs Ag NP loading in the related base liquid defined by $[(K_{nf} / K_{bl}) - 1 * 100]$, at 20 °C. K_{nf} and K_{bl} stand for TC of NFs and TC of base liquid, respectively. All NFs displayed higher TC values (K_{nf}) than base liquids indicating enhancement of TC due to the presence of Ag NP, which scales with Ag NP loadings for all base liquids. Among all NFs systems with different base liquids, NFs with W/EG base liquid displayed the most efficient thermal characteristics compared to the similar NFs systems with water and EG base liquids indicating the base liquid effect. W/EG based NF containing 2 wt% Ag NP loading (Ag NF-W-EG-2 wt%) exhibited the maximum TC enhancement of 12.4%. Viscosity of NFs was also measured and the results are presented in Fig. 3(b), where its increase is defined by $[(\mu_{nf} / \mu_{bl}) - 1 * 100]$ (μ_{nf} and μ_{bl} are the viscosity of NF and of base liquid, respectively) at the same Ag NP loadings and temperature. All Ag NFs containing showed Newtonian behavior at the tested temperature. Moreover, all NFs exhibited higher viscosity values than the relevant base liquid. The minimum and maximum increase of ~3.5% and 6.5% increase in viscosity was observed for W/EG and water based NF containing 1 wt% and 2 wt% Ag NPs loading (at 20 °C), respectively. As a result, among all NF systems, W/EG based NF containing 2 wt% Ag NP (Ag NF-W/EG-2 wt%) displayed the most promising performances with 12.4% TC enhancement with only 5.6% increase in viscosity. This not only indicates the impact of using W/EG mixture (as base liquid effect), but also displays the capability of using this kind of NF as efficient NF for heat transfer applications. Sharma et al. [12] synthesized EG based silver NFs using silver nitrate (precursor), EG (reducing agent), and polyacrylamide-*co*-acrylic acid as dispersion stabilizer and measured the TC of NFs using THW method with no report on viscosity. At 1 wt% NP loading with NP size of 30–40 nm and room temperature, they reported 18% TC enhancement; which is higher than the values we obtained at the same NP loading and almost the same NP size. The difference may be related to

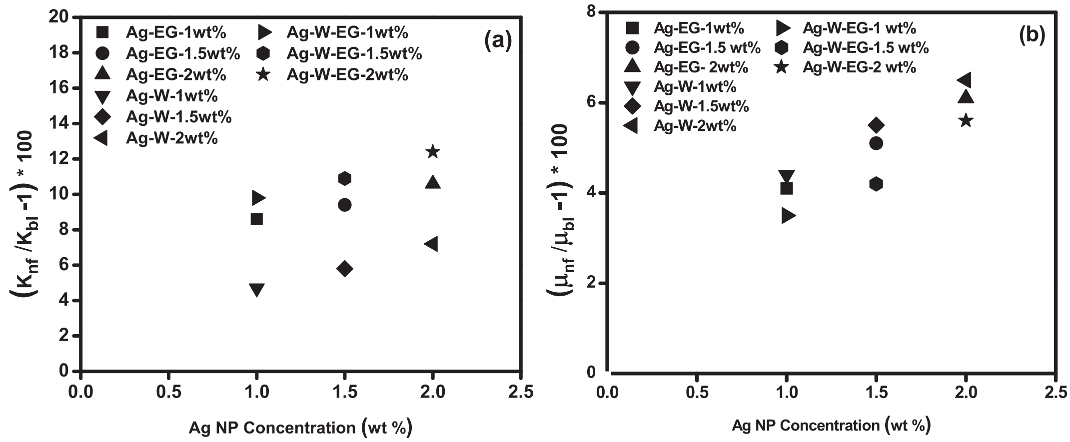

Fig. 3. (a) TC enhancement and (b) increase in viscosity of Ag NFs as a function of Ag NPs concentration with various base liquids all measured at 20 °C.

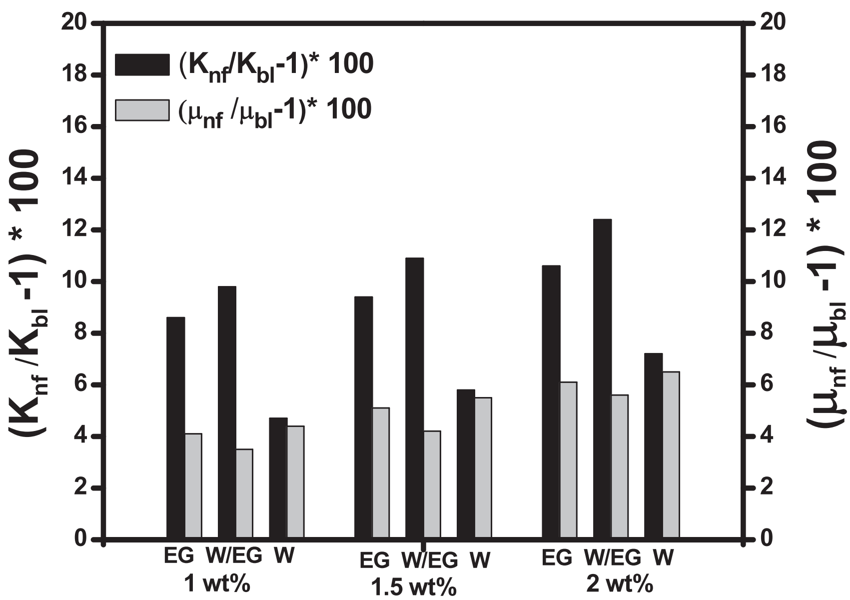

Fig. 4. Comparison of TC enhancement with increase in viscosity at 20 °C for NFs, with different base liquids of water, EG and W/EG containing 1, 1.5 and 2 wt% of Ag NPs; all at 20 °C.

different fabrication method and dispersion stabilizer. Parametthanuwat et al. [14] fabricated water based silver NFs (0.5 wt%) via two-step method using oleic acid (OA) and potassium oleate surfactant (OAK +) as surface modifiers (with different concentration of 0.5, 1 and 1.5 wt%) and investigated the thermal properties of suspensions. The hydrodynamic size for Ag NF containing OA and OAK + were reported as 100 and 95 nm, respectively. They reported 11% and 16% TC enhancement for the water based NFs with 1 wt% OA and OAK + at 20 °C, respectively. Both showed higher values than our fabricated water NF systems even at lower NP loading. Use of different surface modifiers, dissimilar NP size, different sonication time may be responsible for this difference. To have an easier conclusion, the values of TC enhancement and increase in viscosity of Ag NFs with different base liquids, as W and W/EG and EG at different Ag NP loading are summarized in Fig. 4. It should be noticed that base liquid effect obtained in different NF systems is most likely related to better wettability (lower value of the interfacial thermal resistance) of Ag NPs in W/EG medium than in EG or W based NFs [29]. Based on Fig. 4, W/EG based NFs show ~1.2%–1.8% and 4.9%–5.2% higher TC enhancement compared to NFs in EG and water base liquid at the same NPs loading, respectively. At the same NPs loading, W/EG based NFs display 0.5%–1% and ~1%–1.4% lower increase in viscosity compared to NFs in EG and water base liquid, respectively. This suggests that if W/EG is selected as the base liquid, compared to the EG and W base liquid; a lower pumping power is required for heat transfer applications while reverse behavior was observed for water based NFs. Therefore, as heat transfer fluid; W/EG based NFs containing Ag NPs displayed highest efficiencies compared to the similar EG and water based Ag NFs, due to the effect of the base liquid.

4. Conclusions

We presented on the fabrication and evaluation of highly stable Ag NFs in different base liquids as water, W/EG mixture and EG, containing various concentrations of Ag NPs, intended for heat transfer applications. Thermo-physical properties of NFs including TC and viscosity in different base liquids and with various Ag NPs loading were measured at 20 °C. TC enhancement for all NFs systems with different base liquids were observed, due to the presence of Ag NPs. In all NF systems the values of TC enhancements were higher than the number of increase in viscosity indicting the potential of using these kinds of NFs for some heat transfer applications. Among all suspensions, W/EG based NFs with 2 wt% Ag NPs (Ag NF-W-EG-2 wt%) showed the most promising performance; highest TC enhancement of 12.4% with only 6.1% increase in viscosity. As base liquid effect, the W/EG based Ag NFs displayed more favorable performance as heat transfer fluids than the similar water and EG based NFs.

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