



Experimental investigation on thermo-physical properties of copper/diethylene glycol nanofluids fabricated via microwave-assisted route[☆]



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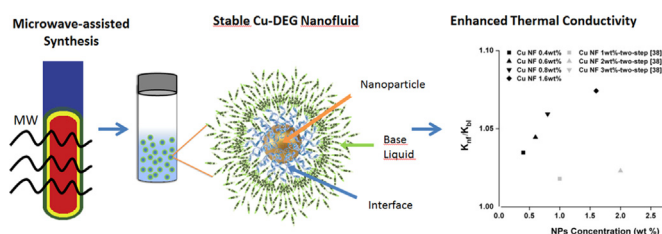
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HIGHLIGHTS

- Stable Cu nanofluids (NFs) have been fabricated by one-step microwave-assisted route.
- NFs with enhanced thermal conductivity and low viscosity increase are demonstrated.
- NFs thermo-physical properties are compared with proper models; deviations observed.
- Comparison between one-step and two-step preparation for NFs has been performed.
- NFs prepared by one-step route showed superior thermo-physical properties.

GRAPHICAL ABSTRACT



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ABSTRACT

This study investigates the fabrication, thermal conductivity and rheological characteristics evaluation of nanofluids consisting of copper nanoparticles in diethylene glycol base liquid. The fabricated Cu nanofluids displayed enhanced thermal conductivity over the base liquid. Copper nanoparticles were directly formed in diethylene glycol using microwave-assisted heating, which provides uniform heating of reagents and solvent, accelerating the nucleation of metal clusters, resulting in monodispersed nanostructures. Copper nanoparticles displayed an average primary particle size of 75 ± 25 nm from SEM micrographs, yet aggregated to form large spherical particles of about 300 nm. The physicochemical properties including thermal conductivity and viscosity of nanofluids were measured for the nanofluids with nanoparticle concentration between 0.4 wt% and 1.6 in the temperature range of 20–50 °C. Proper theoretical correlations/models were applied to compare the experimental results with the estimated values for thermal conductivity and viscosity of nanofluids. For all cases, thermal conductivity enhancement was higher than the increase in viscosity showing the potential of nanofluids to be utilized as coolant in heat transfer applications. A thermal conductivity enhancement of $\sim 7.2\%$ was obtained for nanofluids with 1.6 wt% nanoparticles while maximum increase in viscosity of $\sim 5.2\%$ was observed for the same nanofluid.

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Nomenclature

D	fractal index, –
K	thermal conductivity, W/mK
r_a	size of aggregates of the nanoparticles
r_p	primary size of nanoparticles
T	temperature, K

Greek letters

\emptyset	volume fraction, –
\emptyset_a	effective volume fraction, –
\emptyset_m	maximum particle packing, –
μ	viscosity, kg/ms

Subscripts

nf	nanofluid
p	nanoparticle
bl	base liquid

1. Introduction

Heat transfer is involved in many industrial processes by means of fluid flow to remove excess heat. Conventional heat transfer fluids such as water or ethylene glycol, like almost all substances in liquid phase at room temperature have poor thermal conductivity (TC) characteristics. For instance, compared to the metals liquid TCs are about two orders of magnitude lower. Moreover, developing highly efficient cooling methods for high-tech applications such as microelectronics, transportation, energy supply, manufacturing and biomedical applications has become one of the most important priorities for the further development of these technologies [1–6]. Consequently, there is a demand for effective heat transfer fluids to solve these challenges.

In the last decade nanofluids (NFs) [7], which is a new class of suspensions containing nano sized particles (NPs) in traditional cooling liquids, such as water or mixtures of ethylene glycol (EG) and water, have attracted increasing interest from many researchers because of their potential benefits in heat transfer applications. Up to now several groups of materials and base liquids have been used to engineer NFs. Metal NPs such as Cu, Ag and Au [8–10], oxide particles such as Al_2O_3 [11], SiO_2 [12], CuO [13], mesoporous SiO_2 [14], carbon nanotubes [15] and SiC [16]. Among these systems, suspensions containing copper nanoparticles (Cu NPs) have attracted attention because of their expected high TC.

There are two main methods for the fabrication of NFs: One-step method in which particles are formed directly in the base liquid [17], while in the two-step method particles are synthesized/acquired and then dispersed in the base liquids [18]. Most of the literature reports are on the preparation of two-step NFs by using commercially available NPs. However, there are challenges in fabrication of NFs by the two-step method. In this route because of drying, transportation and storage process of NPs, agglomeration of the particles can take place, which result in clogging of the microchannels causing decrease of the TC, or of the heat transfer coefficient of NFs [19]. A comparison between two-step and one-step method has shown that although the two-step method may be applicable for fabrication of NFs with oxide particles, it is not a suitable choice for making NFs containing metal particles [2]. In order to overcome the above-mentioned challenges connected to the two-step method, such as clogging of microchannels because of NPs agglomeration, development of a new fabrication method is essential.

There are several one-step methods for making suspensions containing metal NPs such as conventional heating of metallic salts in a base liquid or submerged arc NPs synthesis system. Among the metal NPs, synthesis of Cu NPs by conventional one step methods for cooling applications has received considerable attention [17,19,20]. Because of several limitations in conventional procedures of NFs fabrication such as cost and dispersion quality, developing a new route to avoid these challenges is desirable. The microwave-assisted method is a promising route of fabrication, which provides a rapid and uniform heating of reagents and solvent, accelerating the reduction of metal precursor and the nucleation of metal cluster very rapidly, resulting in monodispersed nanostructures, which may in turn improve thermal characteristics of NFs. There are few reports in the literature about the synthesis of Cu NFs/Cu NPs suspensions by the microwave-assisted heating method using EG as the base liquid [21,22]. Based on our former experience and the literature data, NFs fabricated by different groups using low viscosity base liquids exhibit different thermal characteristics, making their comparison and the influence of NPs inconclusive. Several studies report on the use of base liquids with lower TC and higher viscosity and conclude that this type of fluids will benefit most from the addition of solid particles [23,24]. In this work diethylene glycol (DEG) was selected as the base liquid for the demonstration of concept because of its higher viscosity and lower TC values as compared to other base liquids such as water, EG or their mixtures. There are very few reports on the synthesis of NPs in DEG [25], with no mention of their use/evaluation for heat transfer applications. In this work we present the fabrication of Cu NFs in DEG by the microwave-assisted method and for the first time the TC and viscosity of this type of NFs are evaluated experimentally and theoretically. Following our recent attempt in fabrication of Cu NFs using DEG as base liquid via the two-step method [38], the authors focused on the use of the one-step (microwave-assisted) method to overcome some challenges such as stability of NFs and finally obtain NFs with improved TC properties.

2. Methods

2.1. Materials

Polyvinylpyrrolidone (PVP, MW: 10,000 g/mol), diethylene glycol (DEG, $C_4H_{10}O_3$, $\geq 99\%$) copper acetate monohydrate ($Cu(ac)_2 \cdot H_2O$, MW: 199.65 g/mol, $\geq 99\%$), and L-ascorbic acid ($C_6H_8O_6$, MW: 176.12 g/mol, $\geq 99\%$) were purchased from Sigma Aldrich. All the reagents were used as received, without further purification.

The microwave reactor system utilized in this study was Biotage Initiator™ 60 microwave synthesizer operating at a frequency of 2.45 GHz with power output of 0–400 W. The pressure can be varied between 0 and 20 bar and the temperature between 40 and 250 °C. The cavity volume has a range from 0.2 to 20 mL.

2.2. One step fabrication of Cu NFs

The one-step synthesis procedure of Cu NF was adapted from Blosi et al. [25] in which molecular weight of PVP was changed from 55,000 Da to 10,000 Da-which results in a bigger solvodynamic particle size. In order to synthesize the Cu NFs, PVP was dissolved in 10.5 mL of DEG and this solution was heated to 170 °C for 10 min in a microwave oven under magnetic stirring. After reaching the synthesis temperature, two DEG solutions were added into the hot PVP solution: 2 mL of $Cu(ac)_2 \cdot H_2O$ –DEG solution followed by the addition of 3.5 mL of the ascorbic acid–DEG solution. After adding metal precursor, a green solution was obtained, which turned to dark red when the ascorbic acid solution was added, indicating the nucleation of metallic Cu NPs. The one-step Cu NFs synthesized

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