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Lignin as dispersant for water-based carbon nanotubes nanofluids: Impact on viscosity and thermal conductivity $\stackrel{ ightarrow}{ ightarrow}$



P. Estellé ^{a,*}, S. Halelfadl ^b, T. Maré ^b

^a LGCGM EA3913, Equipe Matériaux et Thermo-Rhéologie, Université Rennes 1, IUT de Rennes, 3 rue du Clos Courtel, BP 90422, 35704 Rennes Cedex 7, France ^b LGCGM EA3913, Equipe Matériaux et Thermo-Rhéologie, Université Rennes 1, IUT de Saint-Malo, Rue de la Croix Désilles, CS51713, 35417 Saint-Malo Cedex, France

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ABSTRACT

The viscosity and thermal conductivity of water-based nanofluids containing carbon nanotubes, stabilized by lignin as surfactant, are measured. These experiments were performed at 20 °C and we study the effect of nanotube volume fraction which ranges from 0.0055% to 0.55%. In comparison with SDBS, we show that lignin reduces viscosity and shear-thinning behavior of nanofluids at high volume fraction, without penalizing thermal conductivity enhancement. Viscosity enhancement at high shear rate with respect to nanoparticle content is well modeled by the Maron–Pierce equation, suggesting that lignin is better than SDBS at dispersing carbon nanotubes within water at high volume fraction. The impact of surfactant on base fluid thermal conductivity is also reported and thermal conductivity enhancement of nanofluids with nanoparticle volume fraction is finally compared with previous published models.

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1. Introduction

Solving the development of well dispersed and stable nanofluids requires a combination of various techniques [1], such as physical and chemical methods, including the use of surfactants. This is particularly true when it comes to aqueous carbon nanotube (CNT) nanofluids due to their poor solubility in water. Many surfactants can be used to prepare dispersion of CNTs in water and solvents as reviewed in [2-4]. Typically, surfactant molecules adsorb on the nanotube surface and debundle nanotubes by steric or electrostatic repulsion. A number of studies related to the influence of surfactant in CNT dispersion and thermophysical properties of resulting nanofluids have been previously reported. Among all the surfactants, the most commonly used with CNT nanofluids are sodium dodecylbenzene sulfonate (SDBS) [5-8], sodium dodecylsulfate (SDS) [9-11], hemadecyltrimethylammonium bromide (CTAB) [12] and gum Arabic (GA) [13-15]. Octyl phenol ethoxylate was also used in [16] and the potential of bile salt (such as sodium deoxycholate, DOC) as a dispersing agent was investigated in [10,17]. Stable and homogeneous nanofluids of MWCNTs were obtained by [18] using a gemini cationic surfactant. The quantity of this surfactant was reported to possibly modify the viscosity of nanofluids following the surfactant concentration and temperature. Nasiri et al. [9] studied

E-mail address: patrice.estelle@univ-rennes1.fr (P. Estellé).

dispersion methods such as functionalization and use of SDS coupled with ultrasonication for preparing carbon nanotubes with different structures. They showed that thermal conductivity of nanofluids depends on nanoparticle content as well preparation methods. The effective thermal conductivity of DOC/SWCNT/EG dispersions was reported to increase with nanotube loading in [10]. A comparison with some theoretical models was also performed showing that these models fail to predict the experimental data.

The effects of SDS and SDBS on thermal performance of MWCNTs dispersed in water were investigated by Wusiman et al. [8]. They showed that the thermal conductivity of these surfactant water mixtures decreased with the concentration of both surfactants. The better thermal performance was obtained with SDBS in comparison with SDS dispersant. A study of PVP (polyvinyl pyrrolidone) CNT nanofluids and oxidized nanofluids was performed by Kim et al. [19]. The use of PVP additive is reported to increase the nanofluid viscosity as compared to oxidized nanofluids. In addition, both viscosity and thermal conductivity of PVP mixture nanofluids increase with the concentration of PVP up to 300% in weight of surfactant. The thermal conductivity of CNT nanofluids containing GA was investigated by Walvekar et al. [15] focusing of the effect of GA concentration and nanoparticle content. It was shown that GA does not contribute to thermal conductivity enhancement of nanofluids which was well predicted for low volume fraction by a theoretical model including diameter and dimensions of CNTs and temperature as well. The effect of chitosan, known as a natural polymer stemming from crustacean, as stabilizer in MWCNT nanofluids was studied by Phuoc et al. [20]. Based on experimental results, they concluded that the enhancement in thermal conductivity is independent

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^{*} Corresponding author at: IUT de Rennes, 3 rue du Clos Courtel, BP 90422, 35704 Rennes Cedex 7, France.

of nanofluid viscosity which increases with chitosan content. The viscosity enhancement with surfactant content increase was also reported in [21], considering water and SDBS as surfactants.

In the current context of sustainable development it could be promising to use natural products like GA as nanofluid stabilizers. So, the purpose of this paper is to demonstrate the potential of lignin as a dispersing agent in water-based carbon nanotube nanofluids. This is investigated measuring both the rheological and thermal properties of water-based nanofluids containing carbon nanotubes formulated with lignin as surfactant. These experiments were performed at 20 °C and we study the effect of nanotube volume fraction which ranges from 0.0055% to 0.55%. For comparison purposes, we use previous published data of nanofluids with the same nanotubes stabilized with SDBS, and identical volume fractions [21,22].

2. Materials and experiments

2.1. Nanofluids

As in our previous works [21–23], MWCNTs (density 2.1 g/cm³; conductivity 3000 W/mK) of 1.5 µm in average length and 9.2 nm in average diameter were used here to produce nanofluids with a mixture of distilled water and lignin as base fluid. Lignin is a by-product from wood industry with highly branched molecular structure of OH groups, which was recently shown as a suitable dispersing agent for multiwalled carbon nanotubes [24]. A starting suspension containing 1% in weight fraction of nanotubes (0.55% in volume fraction) was prepared and provided by Nanocyl. In this suspension, the weight fraction of lignin was fixed at 2% as previously used with SDBS [21]. It should be noted that the quantity of surfactants, which was used to stabilize the nanoparticles within the base fluid in the starting suspension, was initially selected by Nanocyl. Nanofluids with lower volume fraction up to 0.0055% were obtained from dilution, as reported in [21–23]. The dilution was performed to maintain a constant surfactant/carbon nanotube weight ratio of 2 followed by mechanical stirring to disperse the nanoparticles in the base fluid. A mixture containing distilled water and lignin (2 wt.%) was also prepared by Nanocyl, and then used to obtain the base fluids corresponding to the nanofluids previously prepared.

The morphology and the nanotube dispersion state of the nanofluid with 0.55% in volume fraction were characterized from scanning electron microscopy (SEM-JEOL-JSM-6301F). In this experiment, the nanofluid was preliminary dried; an accelerating voltage of 10 kV and a working distance of 8 mm were used. SEM characterization of the same suspension with SDBS as surfactant was previously performed, showing that at this concentration the nanotubes are mainly entangled and can form aggregates [1,21]. Fig. 1 shows the SEM picture of the nanofluid with 0.55% in volume fraction and lignin as surfactant. Fig. 1 indicates that the nanotubes are entangled and randomly oriented and form a connected network without significant presence of agglomerates.

2.2. Rheological and thermal conductivity measurements

A stress controlled rheometer (Malvern Kinexus Pro) with a cone and plate fixture was used for the viscosity measurements. The diameter and the angle of the conical geometry were 60 mm and 1° respectively. A Peltier system located below the lower plate allows the sample temperature to be controlled with an accuracy of ± 0.01 °C. Experimental configuration, set-up of the samples and viscosity measurement under steady-state conditions were similar to the ones used in our previous works [21,22]. The experiments were performed for both the nanofluids and the base fluids at 20 °C within the shear rate range 0–1000 s⁻¹. The uncertainty in viscosity measurement was previously reported to be less than 4% within this shear rate range.



Fig. 1. SEM picture taken from dried nanofluid with lignin as surfactant with 0.55% in volume fraction of nanotubes.

The thermal conductivity of nanofluids and base fluids was measured from the transient hot wire method using a KD2 Pro thermal property analyzer (Decagon Devices Inc.). The experimental set-up for thermal conductivity measurement was previously used and described elsewhere [22]. Calibration with distilled water for temperatures ranging from 20 to 50 °C was also previously performed, leading to a standard relative deviation of less than 3.5% [22,23]. Presently reported thermal conductivity values at 20 °C consist of an average of ten measurements.

3. Results and discussion

3.1. Viscosity

Fig. 2 first shows the viscosity of base fluid for the starting nanofluid at 0.55% in volume fraction of nanotubes. A Newtonian behavior is observed within the shear rate range investigated, similar to the base fluid containing SDBS with a close shear viscosity within the experimental uncertainty. The shear viscosity value is 1.074 mPa \cdot s. This enhancement, in comparison with the viscosity of distilled water at 20 °C, is attributed to the presence of lignin. In Fig. 2, the viscosity of the nanofluid at 0.55% in volume fraction of nanotubes is also reported in function of shear rate. It is observed that this nanofluid is shear-thinning as the viscosity decreases when the shear rate is increased. The shear-thinning region is extended up to 200 s⁻¹, then followed by



Fig. 2. Viscosity of nanofluids and base fluids versus shear rate – Influence of surfactant.

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