International Journal of Heat and Mass Transfer 108 (2017) 822–829

Contents lists available at ScienceDirect

International Journal of Heat and Mass Transfer

journal homepage: www.elsevier.com/locate/ijhmt

Evaluation of clustering role versus Brownian motion effect on the heat conduction in nanofluids: A novel approach

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ARTICLE INFO

Article history:
Received 7 November 2016
Received in revised form 14 December 2016
Accepted 20 December 2016

Keywords:
Thermal conductivity
Brownian motion
Clustering
Image processing

ABSTRACT

In this study, the temperature and viscosity-dependent methods were used to identify the main heat conduction mechanism in nanofluids. Three sets of experiments were conducted to investigate the effects of Brownian motion and aggregation. Image processing approach was used to identify detailed configurations of different nanofluids microstructures. The thermal conductivity of the nanofluids was measured with respect to the dynamic viscosity in the temperature range between 0 and 55 °C. The results clearly indicated that the nanoparticle Brownian motion did not play a significant role in heat conduction of nanofluids, which was also supported by the observation that a more viscous sample rendered a higher thermal conductivity. Moreover, the microscopic pictures and the differences in the viscosity between theoretical and experimental values suggested the major role of particle aggregation and clustering.

1. Introduction

Nanofluids, i.e. well-dispersed nanoparticles in a liquid medium, have been found substantial improvement in many effective properties, which is promising for a range of applications ranging from medics to engineering. However it has been long debated on the level of property enhancement. For instance, there are many works reported an anomalous enhancement in the effective thermal conductivity (ETC) of nanofluids, and many theories have been proposed accordingly, including Brownian motion (BM), the existence of interfacial layer, clustering, etc.

Among the mentioned mechanisms, the Brownian motion (BM) and clustering have been intensively investigated. While some researchers have suggested that BM is the major heat transfer mechanism, many others proposed that its effect is negligible in comparison to other mechanisms. For instance, Keblinski et al. [1] suggested that the dependence of thermal conductivity enhancement on the particle Brownian motion maybe insignificant because the calculated Brownian diffusion of the nanoparticles was much slower than the thermal diffusion of the base fluid. Sarkar and Selvam [2] found that the nanoparticle movement was 28 times slower than that of the liquid phase. It was suggested that rather than the slow Brownian motion of nanoparticles, the highly enhanced and fast movement of the surrounding liquid (i.e. micro-convection) was proposed to be the main mechanism for the thermal conductivity enhancement. Similarly, a few researchers also suggested that the effect of Brownian motion on the thermal conduction enhancement was negligible [3–7]. On the contrary, some studies suggested that the Brownian motion effect was important, such as Shukla et al. [8]. Sun et al. [9,10] proposed that the micro convection effect due to the rotation of nanoparticles was the main reason for the effective thermal conductivity enhancement. Tsai et al. [11] also reported that for a low viscous fluid, the Brownian motion of the nanoparticles was much active, which rendered the enhancement in thermal conductivity.

It is accepted that lowering the base fluid viscosity would facilitate the Brownian motion of nanoparticles in a liquid [11–13]. Many methods have been used to alter the viscosity of the base fluid, which include temperature, elapsed-time and nanoparticles configuration [12,14], as well as different base fluids [11,15]. For instance, authors of Ref. [11] have altered the base fluid viscosity through implementing four combinations of two similar

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http://dx.doi.org/10.1016/j.ijheatmasstransfer.2016.12.071
0017-9310/

mediums. In another work, two completely different base fluids with different viscosities have been used to observe the role of the Brownian motion on the thermal conductivity change [15]. However, it is believed that detecting the ETC with viscosity change by adding diluent to the base fluid is not a reliable method to observe the Brownian motion effect. By adding diluent to the base fluid, both BM and TC fluctuations can alter the molecular-scale properties. As to the effect of temperature, some authors indicated that the temperature increase may magnify the effect of BM in the TC enhancement [4,16]. Based on the observation from Gao et al. [17] and Wang et al. [18], if the Brownian motion is important for the enhanced thermal conductivity, it is expected that a lower thermal conductivity would be observed for a frozen suspension. However, the effect of Brownian motion could be neglected if ETC was independent or in a reverse trend of temperature. This idea may support the negligible effect of the Brownian motion, but would not be sufficient to confirm it, particularly in the case of TC enhancement due to temperature increase.

It is expected that as the temperature increases, the vibration effect of the base fluid molecules may wrongly be attributed to the BM effect. The reason may refer to the point that the interaction between the base fluid molecules and the nanoparticle Brownian motion is not yet well-defined. Actually, such a high order of magnitude velocity difference between the fluid molecules and nanoparticles [2] may result in a complicated molecular regime, in which the behavior of low velocity nanoparticles cannot be easily predicted. The simulations by Prasher et al. [19] postulated that the random movement of nanoparticles might gather them together and forms clusters. However, Karthikeyan et al. [20] suggested that such a random movement can be referred as BM, which may result in a breakdown of nanoparticle clusters. One cannot discriminate whether the fast movement of fluid molecules exactly helps the BM enhancement or accumulation of nanoparticles in the sparse clusters. Therefore, it can be concluded that the employment of temperature change as an altering parameter to reveal the role of BM on thermal conductivity is not fully justified.

Our strategy to distinguish the role of Brownian motion versus clustering in heat conduction mechanism of nanofluids is to utilize both viscosity and temperature alteration in a complementary way. We have used silicone oil with different Polydimethylsiloxane (PDMS) chain lengths. Different PDMS chain length is the key parameter that relates heat conduction mechanism to the Brownian motion of nanoparticles. This would introduce an improved implementation of viscosity as an altering parameter for detecting heat conduction mechanism. If the Brownian motion plays the key role, it is expected that a nanofluid with a higher viscosity (i.e., with larger polymer chain length) would cause the blockage of the nanoparticles motion, consequently decrease the ETC. Also, the behavior of nanofluids viscosity versus mass fraction is studied under different temperatures. This would suitably expand our approach to heat conduction mechanism in nanofluids through the viscosity fluctuations. In addition, the structure of micro-scale clusters of prepared nanofluids is comparatively studied with the help of image processing.

2. Material and method

2.1. Material fabrication

The Multi-Walled Carbon Nanotube (MWCNT) was used in this study, which was synthesized at the Nanotechnology Research Center of Research Institute of Petroleum Industry (RIPI) with 90–95% purity. The CNTs were synthesized by the catalytic decomposition of 20% methane in hydrogen over Co–Mo/MgO catalysts at 800–1000 °C [21]. The SEM analysis of the nanoparticles presented in Fig. 1a and b, shows the tangled structure of nanotubes before and after ball-milling. The TEM images in Fig. 2 also show the inner/outer diameter and approximate length of the nanotubes. The dimensions of nanotubes are determined as 3.8 nm and 10 nm respectively for the inner and outer diameter of the tubes. It can be seen that the length of the particles varies from 5 to 10 μm. Silicone oil was considered as the base fluid, whose thermo-physical properties at 25 °C are provided in Table 1.

First, the ball milling process was used to separate the entangled and agglomerated particles, using a planetary ball mill at a speed of 200 rpm for 1 h. The comparison between the SEM images after and before ball milling, Fig. 1, ensures that the particles were not disturbed during this process. The liquid and solid phases were weighted on a scale with a resolution of 0.0001 g. The solid and liquid phases were mixed by a magnetic stirrer for 4 h at a speed of 900 rpm in order to facilitate the sonication dispersion. The sonication procedure was done using both an ultrasonic bath and a probe. The non-continuously sonication [22] with Qsonica ultrasonic probe was implemented on the nanofluid to prevent probable disturbance of nanoparticles due to the sharp increase of temperature.

2.2. Nanofluid characterization

The thermal conductivity of the prepared nanofluids was measured in the Research Institute of Petroleum Industry (RIPI), by using a KD2 Pro thermal property analyzer (Decagon devices, Inc., USA). This device is based on the transient hot-wire method, which is widely used by different researchers. Due to the ±5% measurement tolerance of the device, each sample was measured five times, and the reported values are the overall average values. Fig. 3 shows the procedure of the thermal conductivity measurement. A constant-temperature bath (Julabo, Inc., Germany) was used to fix the nanofluid temperature in the range of 0–55 °C. The water was used to calibrate the KD2 Pro and to be sure about the device accuracy by validating the reference data with the measured one.

The dynamic viscosity of the prepared nanofluids was measured by a glass viscometer (Petrotest Co.) in a constant-temperature bath in the Research Institute of Petroleum Industry (RIPI).The constant-temperature bath provides viscosity measurement at the steady-state conditions in the temperature range of 0–55 °C. Fig. 4 shows a schematic procedure of the viscosity measurement. By applying vacuum conditions at one end of the viscometer, the time of passing nanofluid from the gradation of the device was recorded, and then the viscosity value was determined. In the case of viscosity measurement in the temperature of over 100 °C, paraffin can be used instead of water.

3. Image processing

In order to establish a reasonable comparison between different nanofluids' microstructure, we need to quantify the texture content of the taken microscopic images [23]. Therefore, an algorithmic image-processing procedure, based on the statistical information extracted from micro-clusters, was performed. The prepared nanofluids were taken several microscopic images to study the configuration of nanoparticles micro-clusters. Fig. 5 shows the schematic flow chart of the process, which was developed in the Matlab platform.

The process included three major steps, in eight subsets with four output parameters, as described below. These three steps were successfully approved as a tool in morphological study of micro-structures by Tahmooressi et al. [24].