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

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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 conductedto 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 Resultsclearly indicated thatthe nanoparticle Brownian motion does 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 valuessuggested the major role of particle aggregation and clustering.

Keywords: Thermal conductivity; Brownian motion; clustering; Image processing

1. Introduction

Nanofluids, i.e. well-dispersed nanoparticles in a liquid medium, have been found substantial improvement inmany effective properties, which is promising for a range of applications ranging from medicsto 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 BMis 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 wasmuch 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 researchersalso suggested that the effect of Brownian motion on the thermal conduction enhancement was negligible [3-7]. On the contrary,some studies suggested thatthe 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 fora 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 nano-particles 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 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 besufficient to confirm it,particularly in the case of TC enhancementdue to temperature increase.

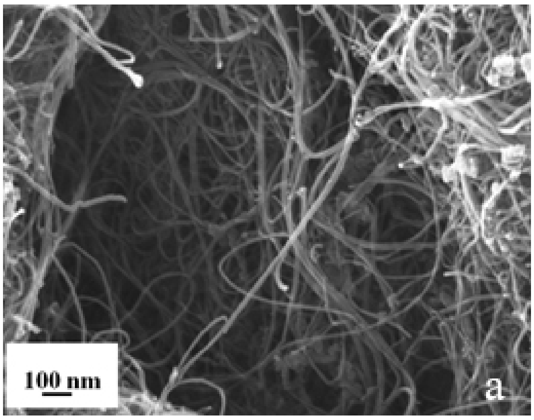
It is expected that as the temperature increases, the vibration effect of the base fluid molecules may wrongly be attributed to the BM effect. The reason may refer to the point that the interaction between the base fluid molecules and the nanoparticle Brownian motion is not yet well-defined. Actually, 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 byPrasher et al. [19] postulated that the random movement of nanoparticles might gather them together and formsclusters. 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 causethe 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].TheSEM analysis of the nanoparticles presented in Fig. 1a, 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.



100 nm

Fig. 1. SEM images of MWCNT a) before and b) after the ball-milling process

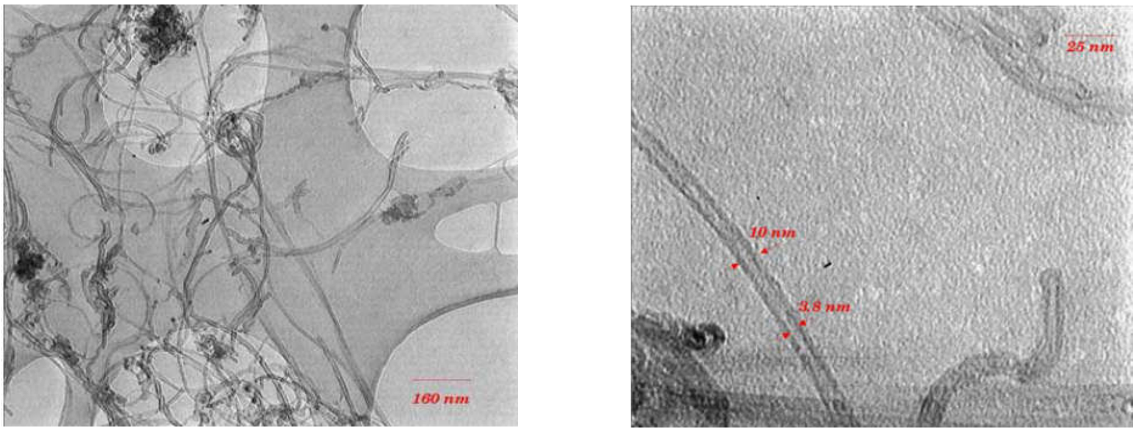


Fig.2. TEM images of MWCNT, showing its length and diameter

Table 1. Properties of base fluidat 25 ℃

Base fluid	Viscosity (cSt)	Thermal conductivity ($\frac{W}{mK}$)	Density ($\frac{Kg}{m^3}$)	Manufacturer
Silicone oil	150	0.144	980	KCC Co. Korea
Silicone oil	1000	0.144	938	KCC Co. Korea

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 hour. The comparison between the SEM images after and before ball milling, Figure 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 hoursat 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 onthe 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.3shows the procedure of the thermal conductivity measurement. Aconstant-temperature bath (Julabo, Inc., Germany) was used to fix the nanofluid temperature in the range of 0°C to 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.

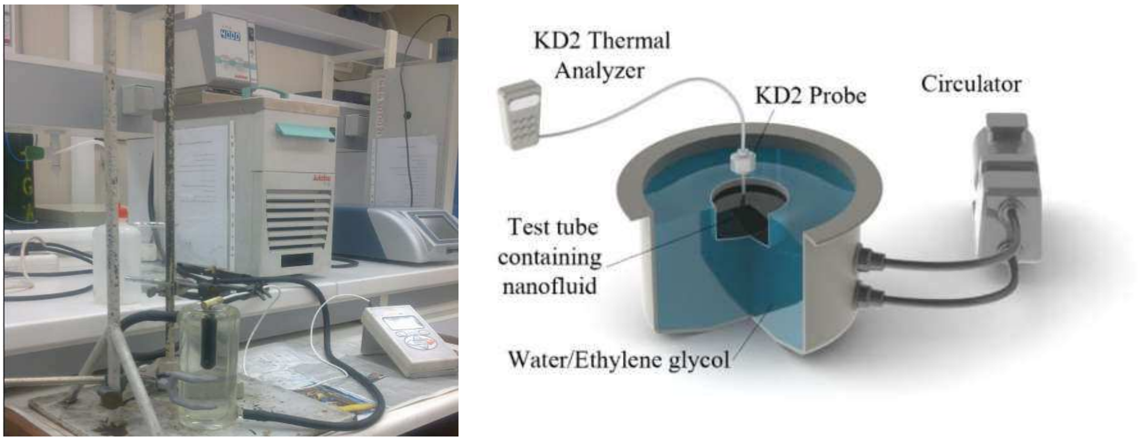


Fig. 3. Thermal conductivity measurement under the temperature range of 0°C to 55°C

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°C to 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 devicewasrecorded,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.

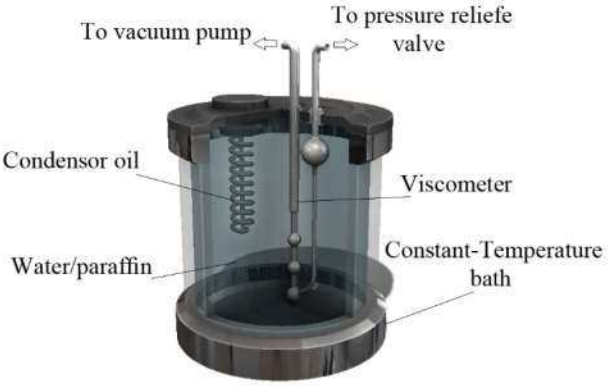


Fig. 4. Viscosity measurement under the temperature range of 0°C to 55°C

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 Matlabplatform.

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].

3.1. Primary process

The first step contains preliminary modifications. It equalized all original images by omitting unnecessaryelementsand prepared them for the next steps. The acquired images were in the RGB type, which was then converted to the gray-scale type. After that, the classical contrast enhancement procedures were applied, which consisted the background/foreground discrimination, noise elimination and segmentation [25,26].

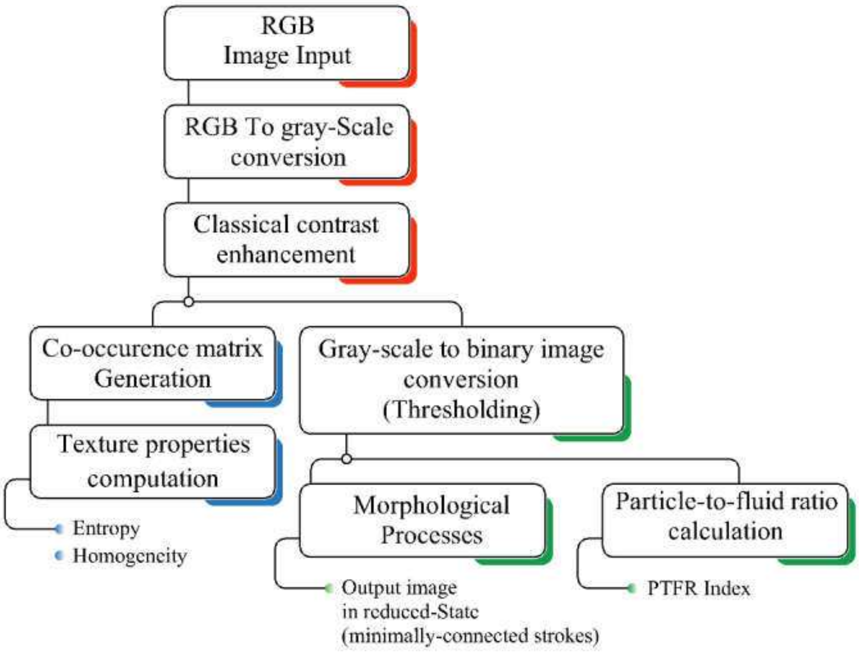


Fig. 5. Image-processing algorithm

3.2.Texture analysis

Describing an image texture would need to take into account not only the gray-level of single pixels but also their relative positions in association with the neighboring pixels [23, 27]. Generating the co-occurrence matrix would take care of this action. This matrix was then used for the calculation of some moments of intensity called 'Entropy' and 'Homogeneity'. The image entropy is a unique index describing the level of randomness in the distribution of foreground objects (nanoparticles micro-clusters). In fact, a higher entropy stands for an image with better percolated micro-clusters all over the entire base fluid. The Matlabcalculatedthe image entropy using the following equation [28]:

Entropy: $H = -\sum_{i=1}^{n} p_i \log{(p_i)}$ (1)

where p_i is the probability of occurrence of the event i and:

$$\sum_{i=1}^{n} p_i = 1, \qquad 0 \leq p_i \leq 1. \qquad (2)$$

The algorithm is also used to calculate the image homogeneity which is an indicator for the texture uniformity [29]:

$$\text{Homogeneity:} \quad U = \sum_{i=0}^{L-1} p^2(z_i) \qquad (3)$$

where L is the number of possible intensity levels, p(z) is the histogram of the intensity levels in a region and z is a random variable indicating intensity. A texture with higher number of homogeneity contains larger 'cluster-free-zone' and lower 'degree of randomness' in the objects (clusters) distribution.

3.3.Morphological processes

In this step, by using the 'bwmorph' function, binary images went through an 'object-thinning' process. The micro-clusters in the output image were in the form of minimally-connected strokes. By this step, the configuration of micro-clusters was better estimated visually. Thin rods revealed that the clusters were connected and permeated through the base fluid. Also, further analysis calculated the ratio of the surface area (number of pixels) of the objects (black pixels) to the background (base fluid). This index is reported here as “Particle to Fluid Ratio (PTFR)”. This quantity is an indicator of the amount of nanoparticles clusters that are grown up to the micro scale. Therefore, a lower object-to-background ratio shows less formation of micro-clusters.

4. Result and Discussion

4.1. Temperature effect

This part discusses the Brownian motion in nanofluids through investigating the temperature-dependent thermal conductivity and shape evolution of clusters. For this purpose, a sample containing 0.8% CNT silicone oil-based was prepared, and the TC characterization was performed following the method described in the previous section. Fig. 6 shows the behavior of TC changes for both base fluid and nanofluid when the temperature was varied from 0°C to 55°C. It can be observed that the nanofluid has a smaller decreasing slope of TC in comparison with the base fluid. It means that the ETC, as a result of adding nanoparticle, is gradually increased with the temperature rising.

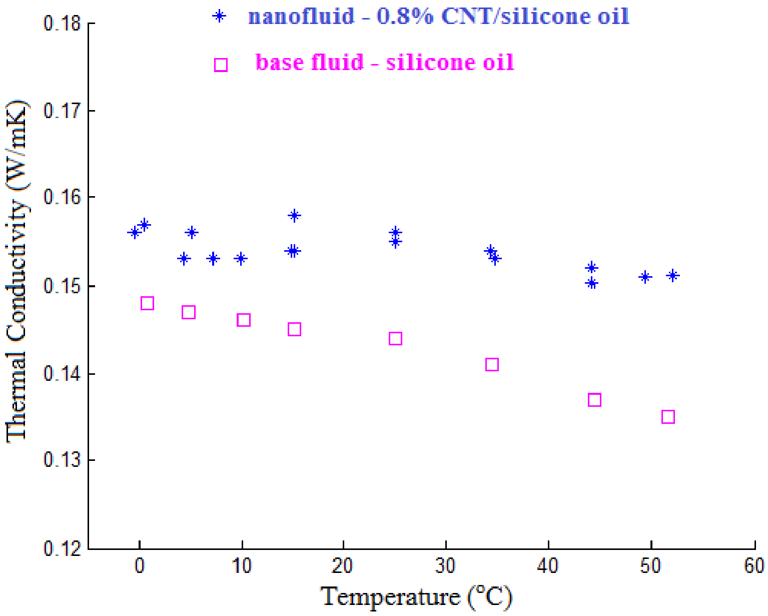


Fig. 6. Variation of thermal conductivity of base fluid and nanofluid in the range of 0°C to 55°C

The microscopic images (Fig.7) were taken from the sample at three different temperatures to show the clustering configuration change.The left image was at a temperature around -70 °C, the middle one at room temperature and the right one at 100 °C. According to Fig. 7, a well-diffused percolated structure can be identifiedas temperature increased. At low temperature, the clusters tend to accumulate and make isolated regions. As a result, cluster networks become disconnected, forming “particle free-zones” and “particle rich-zones” regions [19]. The increasing trend of ETC can be explained by the clusters configuration. At the higher temperature, the well-diffused thermal pathways would facilitate the heat transport and lead higher ETC; while the isolated clusters at a lower temperature would act in an inverse manner, which causes lower ETC. The mentioned well-diffused structure and the isolated regions can clearly be seen in the in Fig.8.

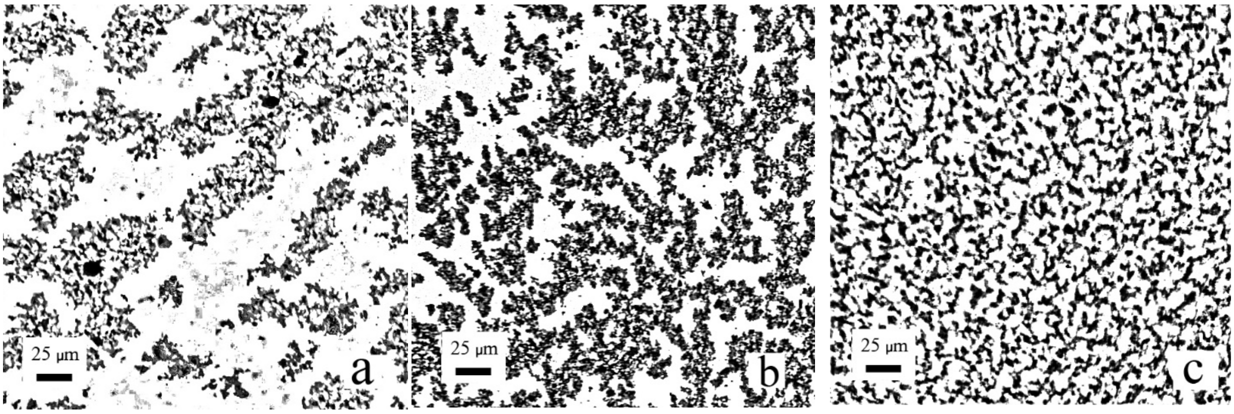


Fig. 7. Microscopic images containing CNT: a) frozen sample, b) at room temperature, c) at 100 ℃

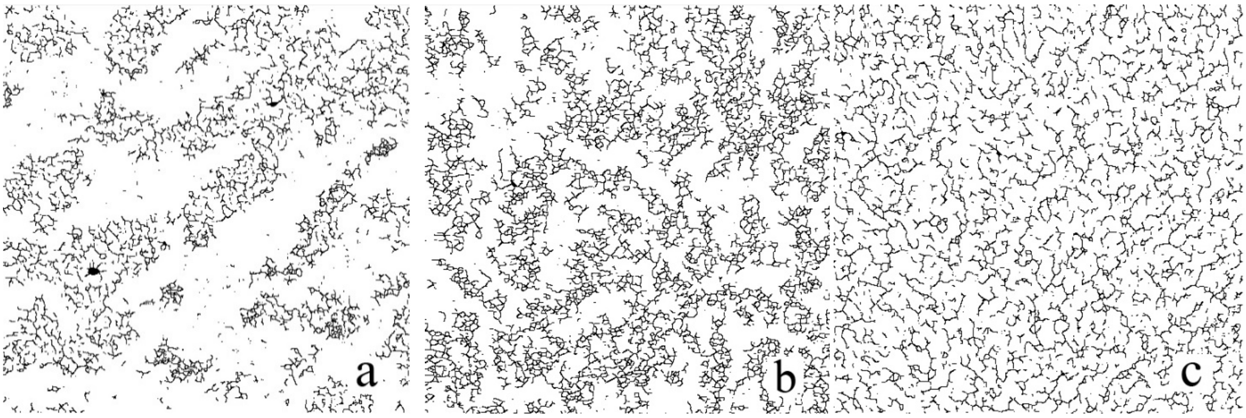


Fig. 8. Eroded images of: a) frozen sample, b) at room temperature, c) at 100 ℃

The positive role of clustering can also be confirmed by the entropy-based image processing results for the above microscopic images,which are presented in Table 2.The higher entropy stands for the higher thermal conductivity in the sample (c). It can also be seen visually that thermal pass-ways in the sample (c) are percolated all over the base fluid. While, in other samples,particles are concentrated in large clusters and accumulated in sparse regions.The homogeneity also indicates the relative state of particle-free zones in samples (a) to (c). Samples (a), owns higher homogeneity number due to the larger particle-free zone. This is because of the accumulation of particles in the thick micron-size aggregations. The existence of such aggregations would limit the probability of heat transferthrough the solid structure all over the base fluid.

Table 2. Image processing analysis of set 1

	Homogeneity	**Entropy**	**PTFR**
Sample (a)	0.5721	8.219	0.362
Sample (b)	0.4944	9.798	0.522
Sample (c)	0.3989	11.088	0.5358

Current microscopic images and the image processing results suggest that the trend of ETC change can be justified by clustering. However, altering the temperature (as an external stimulus) may not be sufficient to evaluate the effect of BM. There is no enough evidence to neglect the BM effect, and also it is not reasonable to assign this trend of ETC to the BM because the improved kinetic energy and vibration of the base fluids molecules may reinforce either the BM effect or the clustering. The temperature changes may affect other nanofluid properties such as viscosity. Therefore, it would be essential to scrutiny the correlation between temperature, viscosity and thermal conductivity in order to clarify theprominent thermal conductivity mechanism.

4.2. Viscosity effect

In this experiment,two base fluids with all the same properties, but different viscosity, were used. It is of note that they have equal thermal conductivity, too. Silicone oil with two different values of viscosity was used as the base fluid where MWCNTs were dispersed in the base liquid. Both samples were prepared using the same method and characterized at the same conditions.

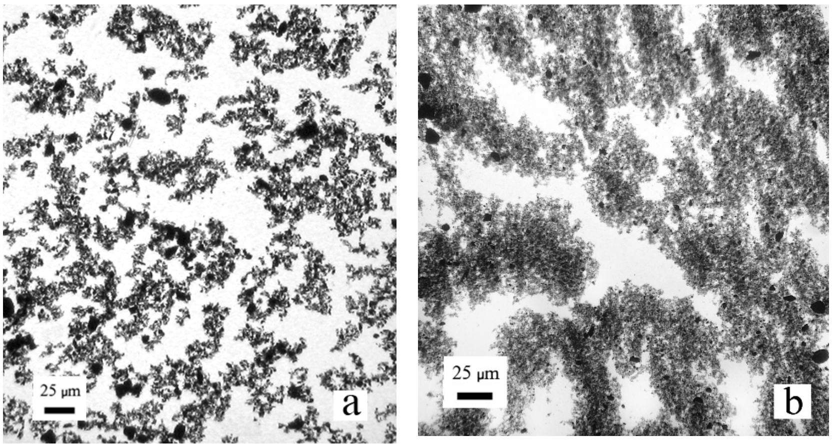


Fig. 9. Sample with silicone oilas base fluid with a viscosity equal to a) 150 cst and b) 1000 cst

Fig. 9 shows the micro-scale clusterconfiguration of two nanofluids used in our experiment. It can be seen, in both images thatthe clusters are of high level of percolation. This may justify the high percentage of TC enhancement in both nanofluids, which is reported in Table 3.

In addition to the image, we considered two other indexes to explain the cluster configurations. Table.3 shows thattheir PTFR numbers are not the same although they have the same volume fractions. Surprisingly, despite the higher PTFR of sample (a), it does not show higher TC. It clearly indicates the importanceof clusters percolation quality and the way the clusters are dispersed. This quality is quantitatively reported in Table. 3. It can be observed that the values of ETC for the considered samples verify the image processing interpretation since its valuesare 16.67% and 20.83% for the sample (a) and (b), respectively.

Table 3. Image processing analysis of viscosity effect set

	ETC (%)	Homogeneity	Entropy	PTFR
Sample (a)	16.67	0.4579	10.5328	0.872
Sample (b)	20.83	0.3777	13.2686	0.392

The results of this experiment indicate that the BM cannot be responsible for the observed enhancement of TC.In fact, if the Brownian motion is the key mechanism, it is expected to see a higher ETC for the nanofluid with a lower viscosity. However, the TC measurement contradicted such a statement. In addition, the microscopic images and image processing results of the samples confirm that the trend of the TC changes with viscosity can be well explained by the clustering.

4.3. Nanoparticle's concentration effect

To strengthen ourargument, it should take care of some other important parameters affecting the nanofluids viscosity. According to the Einstein equation [30], the solid weight fraction plays avital role on the nanofluids viscosity.

$$\left(\frac{\mu_{nf}}{\mu_{bf}}\right) = 1 + 2.5\phi (4)$$

Where the subscripts "nf" and "bf" refer to nanofluid and base fluid, respectively, and the coefficient ϕ stands for the nanoparticles concentration.

The temperature has been indicated to have a decreasing effect on the viscosity of nanofluids [31, 32]. As a result, the effects of both solid mass fraction and temperature were experimentally investigated.

The dynamic viscosity of the synthesized nanofluids was measured under the influence of solid weight fraction of 0.2% and 0.6%,in four temperature sets of 10 °C, 25 °C, 40 °C and 55 °C. In addition, the experimental results were compared to Einsteinmodel.Both viscosity and thermal conductivity were found to increase with the increase of the solid particles concentration [33-35]. Fig. 10((a) to (d)) shows our experimental data of nanofluids viscosity behavior with respect to the solid weight fraction. In all four states, it can be seen that the proposed model deviate from the experimental data as the weight fraction increases. In fact, the viscosity increase would be more significant at higher solid weight fractions [19].

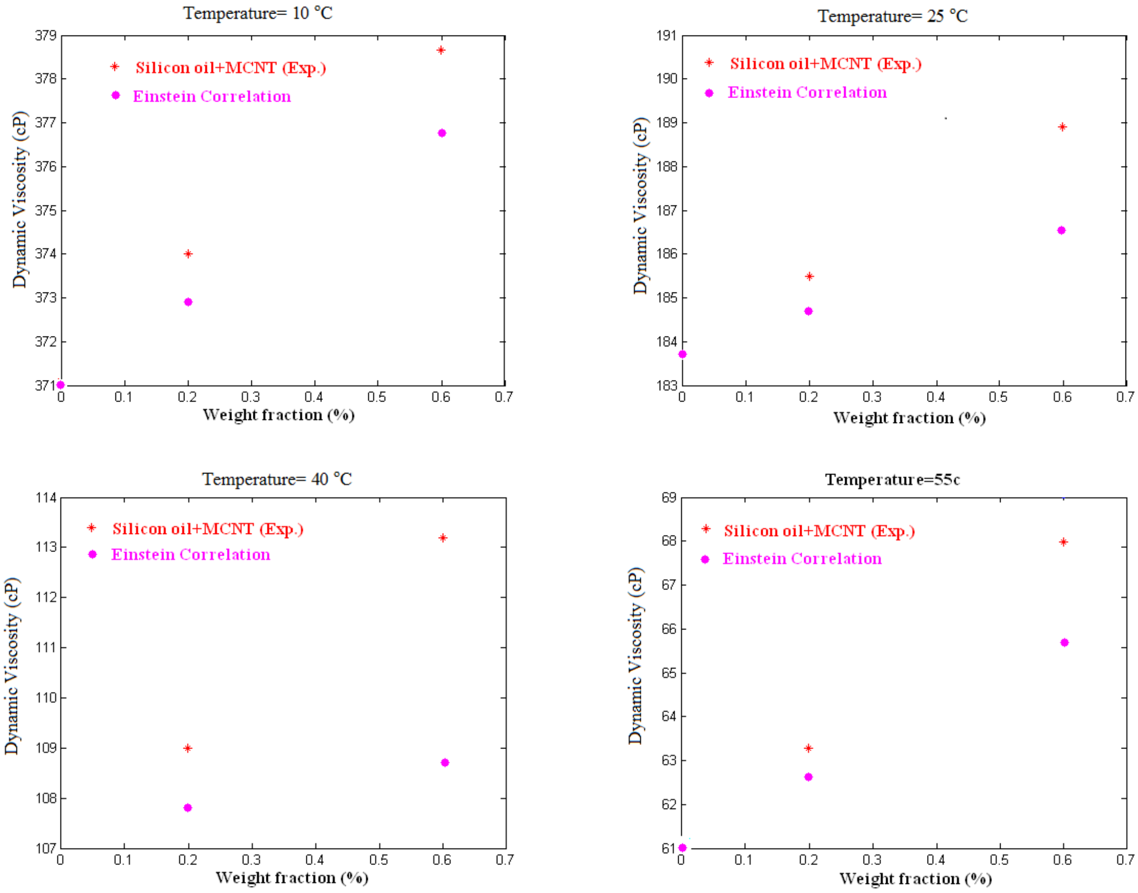


Fig. 10. Nanofluids viscosity behavior, measured at two weight fractions of 0.2% and 0.6% for CNT, and at four temperatures of 10 °C, 25 °C, 40 °C and 55 °C

As the solid weight fraction increases, the average inter-particles distance decreases, which promote the formation of nanoparticle aggregations. Asimilar result was also obtained byHalelfadl et al. [36]. Recent studies on nanofluid showed that the presence of particles aggregation would increase the viscosity of nanofluids [37]. In fact, the concentration of nanoparticles in large and scattered sites may behave as internal solid structures, increasing the internal resistance to the fluid flow [4].Such a postulation has been both experimentally and theoretically proved by other researchers [38, 39]. It can be safely drawn that the clustering phenomenon is one of the most important issues of nanofluids influencing their viscosity, extending from micro to nanoscale [13, 20, 40-42].

This debate comes to the conclusion thatthe BM may not be distinguished as the dominant heat transfer mechanism in nanofluids.The higher TC observed in the more viscous sample showsa negligible influence of Brownian motionon heat conduction. On the other hand, it seems that the formation of particles aggregation suitably justifies the enhancement in thermal conductivity.

5. Conclusion

We investigated the rheological and thermophysical properties of carbon silicon oil-based nanofluids to illuminate the dominant heat conduction mechanism in nanofluids.Considering the controversies evidenced in the reported experiments over the role of the Brownian motion, three sets of supplementary experiments were performed. First, taking temperature as an altering parameter to investigate the Brownian behavior of nanoparticles may not necessarily indicate the dominancy of this mechanism. It is believed that increasing temperature would definitely increase the level of complexity of solid particles-base fluid interactions. In such a regime, one cannot anticipate the exact behavior of solid particles in response to the high-frequency collisions of base fluid molecules, which may result in the formation of clusters or breakdown of particle aggregates. Secondly, the configuration of micro-scale aggregations revealed by image processing also supports such a statement. Thirdly, separated experiments were conducted to assess the effect of viscosity, which indicatedan insignificant role of nanoparticles Brownian motion innanofluids heat conduction mechanism.Convincingly our work supports the dominance of particle aggregation effect in nanofluids heat conduction.

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