Applied Thermal Engineering 114 (2017) 346–359

Contents lists available at ScienceDirect

Applied Thermal Engineering

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

Research Paper

# Percolating micro-structures as a key-role of heat conduction mechanism in nanofluids

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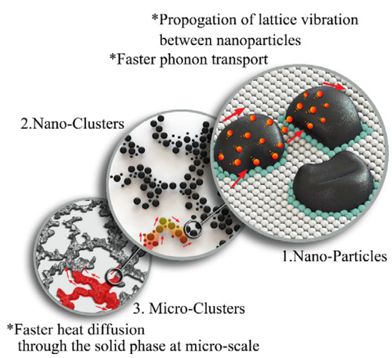
## HIGHLIGHTS

- New application of image processing is used to study micro-clusters in nanofluids.
- Effect of working factors on nanofluid are experimentally investigated.
- Micro-clusters morphology of five sets of nanofluids is studied.
- Nanoparticles clusters may grow up to the micro-scale.
- Well-percolated micro-clusters enhance heat conduction in nanofluids.

## GRAPHICAL ABSTRACT




## ARTICLE INFO

*Article history:*
Received 27 August 2016
Revised 29 October 2016
Accepted 28 November 2016
Available online 29 November 2016

*Keywords:*
Percolating micro-structures
Clustering
Image processing
Thermal conductivity enhancement
Carbon-based nanofluids

## ABSTRACT

The primary working mechanism behind nanofluids thermal conductivity can be distinguished as one of the most controversial issues of nanofluids. Between several theories proposed in literature, nanoparticles' clustering is the sole mechanism that can be observed even at the micro-scale level. Several parameters including nanofluid preparation method, particles concentration, particles morphology, temperature, and elapsed time are experimentally altered to investigate the role of nanoparticles micro-clusters in nanofluids thermal conductivity. It is observed that a minor variation in the altering parameters, would lead a remarkable change in the micro-clusters configuration and subsequently, in the thermal conductivity of nanofluids. In this paper, it is focused on this phenomenon by introducing a new application of image processing as a tool to study the morphological characteristics of micron-sized clusters. Through this approach, it is demonstrated that higher thermal conductivity is obtained when particles micro-clusters are well percolated all over the base fluid. As a result, well-diffused percolating micro-structures are known to have a significant role in heat conduction of nanofluids.


## 1. Introduction

Anomalous enhancement in effective thermal conductivity of nanofluids observed by some researchers is a controversial topic and many authors have discussed it earlier. A vast majority of experiments report increase in thermal conductivity beyond Maxwell limit, while some other researchers claim that enhancement in thermal conductivity beyond Maxwell limit seems to be impossible [1]. Apart from the debate, several mechanisms have been proposed to explain the physics behind this phenomenon such as

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http://dx.doi.org/10.1016/j.applthermaleng.2016.11.197
1359-4311/

clustering, phonon transport regime, Brownian motion (BM), and existence of interfacial layer.

Nanoparticles' clustering is one of the most challenging topics in nanofluid thermal conductivity studies. Although there are so many papers proving that clustering affects positively in thermal conduction enhancement, others claim that it has the adverse effect. Karthikeyan et al. [2] have found that when the mesh-like structure is formed, the thermal conductivity began to drop. Their experiment was based on the time-dependent thermal conductivity investigation of water based CuO nanofluid, in which the thermal conductivity decreased with elapsed time due to microscopically clustering of nanoparticles. Several other studies similarly have concluded that conduction clustering causes a dramatic decrease of thermal conductivity [2–5]. On the other hand, Prasher et al. [6] theoretically have demonstrated that the thermal conductivity of nanofluids can be significantly enhanced by the aggregation of nanoparticles into clusters. Feng et al. [7] proposed a new model for the effective thermal conductivities of nanofluids based on the nanolayer and nanoparticles' aggregation. Later, Kole and Dey [5] confirmed that the thermal conductivity enhancement agrees well with Feng's model, which explains the thermal conductivity of nanofluids dependent on both nanolayer and nanoparticles cluster. Other theoretical works [8–10] have confirmed the impressive effect of aggregation on thermal conductivity enhancement in comparison to the other mechanisms. The experimental results [11–14] also suggested that nanofluid with clusters show significant enhancement of thermal conductivity. In an experimental research works by Gao et al. [15] and Wang et al. [16] the thermal conductivity mechanism was investigated in both liquid and solid states. This paper's results strongly suggest that clustering holds the key to the thermal conductivity enhancement of nanofluids. Also, their results indicate that the Brownian motion of nanoparticles is not the main cause of the thermal conductivity enhancement.

In the literature reviewed above, all have focused on nanoscale agglomerations. Despite of adding nanoscale solid particles to a conventional fluid, it is believed that thermal behavior of the suspension is to a great extent related to the configuration of micron-sized clusters. Observations by some other researchers [2,3,11,15–19] clearly prove the formation of micron-sized clusters engendered from the growth of the nanoscale clusters. For instance, Wang et al. [16] attributed the abrupt thermal conductivity enhancement (from 12% to 36%) to the formation of micro-structure as high heat conducting pathway.

Although formation of micron-size clusters is mentioned in several studies, it is not deeply investigated as a critical issue. In this paper, our main approach is to highlight the role of clustering as the key mechanism from the micro-scaled point of view. For this purpose, it is focused on basic principles of thermal transport behavior in nanofluids. Then, by using image processing tool, some evaluating parameters are defined to quantitatively study the configuration of micron-sized clusters. Microscopic images of five different sets with altering parameters of preparation method, particles concentration, particles morphology, temperature, and elapsed time are presented and analyzed through image processing algorithm. Our experiment shows that by changing the considered variables, meaningful variations in size and shape of micron-sized clusters can be observed. Then, an analytical interpretation out of the obtained results, based on the proposed theories in literature is presented. It is believed that making any changes to nanofluids altering parameters, would not affect the thermal conductivity unless they alter the clusters morphology, this case is described in this paper as a novel work. The focus is to discuss how grown-up clusters can explain heat conduction enhancement in nanofluids.

## 2. Heat conduction mechanisms of nanofluids

### 2.1. Clustering

Observations by researchers have evidenced formation of particle aggregations due to some interacting forces [12,20,21]. In literature, these aggregations are mostly called "clusters" or "agglomerations". Regardless of what they are called, this phenomenon is proved to be responsible for effective thermal conductivity fluctuations. Clustering stands against particles dispersal referring to the state in which they tend to stick together and form a solid state rich zone in a colloidal suspension [5]. To facilitate the study of clusters, Morphological examinations reveal some identical numbers such as radii of gyration or fractal number [6,22]. These indexes show clusters shape and size and the way they spread out through the base fluid.

Basically, in solid state material especially those with crystalline lattice, thermal energy can flow faster than other states. That is why adding solid Nano particles to conventional fluids leads to effective thermal conductivity enhancement. This fact is depicted in Fig. 1 and as animation in online format. Accordingly, aggregation of particles which provides smaller inter particles distance and lower thermal resistance pathway [6], would be a strong proof for those who advocate clustering role in increasing thermal conductivity coefficient. In this paper, it is supposed to establish proper reasoning to support this postulation.

### 2.2. Brownian motion

Brownian motion (BM) defined as random movement of nanoparticles in response to their frequent collision with base fluid molecules [23], is one of the working mechanisms regarding the large body of theoretical and experimental works [2,5,22,24,25]. Brownian motion is a general topic by which couple of mechanisms can be recognized.

First, some authors believe that nanoparticles act as heat carriers so that they can transport thermal energy due to their translational movement [26]. On the contrary, others challenge the possibility of such event by providing enough molecular dynamic simulations in which they claim that thermal diffusion in media is faster than translational movement of particles [27–29]. The second possible function defines by micro convection effect. In practice, this effect can be expected in two ways: First, convectional heat transfer due to translational displacement of particles. Second, micro-mixing effect because of fast rotation of Nano particles [24,30–33]. This theory seems to be more realistic but the problem is the amount of enhancement which according to molecular dynamic simulations cannot be responsible for such an anomalous increase [29,34]. Considering BM as dynamic interaction of nanoparticles and fluid molecules, some other authors have shown that it is not the Brownian motion of Nano particles that enhances thermal conductivity, but the fast movement of liquid atoms due to adding solid nanoparticles [35,36]. Indeed, adding solid Nano scale particles will result in an increased movement of fluid molecules which consequently, can enhance thermal transport through the suspension media. The mentioned definitions of the Brownian motion is depicted in Fig. 2, in which the translational movement of particles as heat carriers, micro-convection around particles, and vibration of medium molecules are shown, and the related animation is available in online format.

### 2.3. Nanoscale thermal transport

Optimizing thermal properties of nanofluids is of a great importance as they are employed in heat transfer applications. Without