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ON THE INCREASED HEAT CONDUCTION AND CHANGED FLOW BOUNDARY-LAYER OF NANOFLUIDS BY MOLECULAR DYNAMICS SIMULATION

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The present work is intended to reveal the microscopic physical essence of nanofluid convective heat transfer enhancement. Based on the idea that the improved convective heat transfer performance with nanofluids is caused by their increased heat conduction and changed flow boundary-layer, the present work performs molecular dynamics (MD) simulations with nanofluids from two aspects, including: heat conduction and boundary layer flow. MD simulation models for heat conduction and boundary layer flow are created separately. Thermal conductivity of water-based nanofluids is calculated by MD method and temperature effect for nanofluid thermal conductivity is discussed. Furthermore, influence of temperature for the absorption layer and micro-motions of nanoparticles have been examined. Flow behaviors of laminar sub-layer with nanofluids are studied. It is found that micro-motions of nanoparticles completely change the flow behavior of nanofluids, destroy the flow boundary layer, and therefore make heat conduction through laminar sub-layer easier. The results reported in this work provide direct evidence for explaining convective heat transfer enhancement in nanofluids.

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Keywords: Nanofluids; Thermal conductivity; Flow boundary layer; Molecular Dynamics; Mechanism

1. Introduction

The novel concept of nanofluids denotes the new class of heat transfer medium consisting of solid nanoparticles, with average sizes on the order of 1–100 nm, suspended in a conventional heat transfer liquid [1]. In recent years nanofluids have attracted great interest owing to their anomalously enhanced thermal properties. The most attractive attribute of nanofluids is their abnormal increased thermal conductivity. Researchers have found that by adding a small amount (less than 1% volume fraction) of nanoparticles to traditional fluids with poor thermal conductivity, the promotion of thermal conductivity can be more than 10% [2, 3]. The convection heat transfer performance of nanofluids is even better. For instance, Xuan et al. reported that under the condition of same Reynolds number the convection heat transfer coefficient is increased by 40% by adding 2.0% volume fraction of copper nanoparticles in water. While with the same volume fraction of copper nanoparticles addition, the thermal conductivity has been tested to have increased by merely 15% compared to that of water [4]. Nanofluids, with their improved thermal properties, could be applied in a variety of heat exchange equipment for the purpose of enhancing heat transfer [5]. However, researchers still cannot give thorough answer for their anomalously increased heat transfer properties, which limits the application process of nanofluids.

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Convective heat transfer is the most common heat transfer phenomenon. The convective heat transfer performance in nanofluids is experimentally found to be markedly improved compared to single-phase fluids. For instance, Wen et al. [6] measured the convection heat transfer performance of Al_2O_3-water nanofluids (1.6% volume fraction) in laminar flow, and their results showed that compared to that of water the Nusselt number of Al_2O_3-water nanofluids is increased by 30%. The obviously increased thermal conductivity is one reason for explaining convective heat transfer enhancement in nanofluids. For conventional single-phase fluids, the heat transfer performance is better when the heat conduction properties are better. The same rule holds for nanofluids. For instance, thermal conductivity of water with suspending Cu nanoparticles is higher than those of nanofluids with Al_2O_3 or CuO nanoparticles suspended. Then the Nusselt number of the former is also higher than the latter under the same conditions [7]. The other reason for explaining convective heat transfer enhancement in nanofluids is the changed flow behavior with destroyed flow boundary layer. This formulation is proposed with the essential understanding that: heat transfer is based on fluid flow; with changed flow field the heat transfer rate will definitely be changed. Researchers revealed through experiments that the longitudinal mixing of nanofluids is better, and vortex number is increased, which indicates the flow field of nanofluids is changed. Furthermore, the mass transfer properties of nanofluids are also found to be increased through adding nanoparticles. [8] The flow behaviors of nanofluids is obviously different from that of single-phase fluids.[9-11] As is known to all, the heat transfer capacity through convective heat transfer is equal to the quantity of heat conduction through laminar sub-layer (or called viscous sublayer). Nanoparticles, due to their small scale effect, may cause totally changed flow behavior in laminar sub-layer, which would be crucial for revealing convective heat transfer improvement in nanofluids. However, it is very hard with experiments to investigate this microcosmic mechanism.

Molecular Dynamics (MD) method has been proved to be effective for revealing microscopic mechanisms of heat conduction enhancement with nanofluids by many researchers [12-17]. The advantage of MD simulation is the deterministic simulation results, which is able to provide some key microcosmic mechanisms. However, regarding to the comprehensive mechanisms for explaining convective heat transfer improvement with nanofluids, including: the increased thermal conductivity and changed flow behaviors, more MD simulation work still needs to be done.

In order to comprehensively reveal microscopic mechanisms for convective heat transfer enhancement with nanofluids, the present work conducts MD simulations from two aspects, including: heat conduction and boundary layer flow. Simulation models are created respectively. Thermal conductivity of water-based nanofluids is calculated by MD method. Temperature effect for nanofluid thermal conductivity is discussed. Influence of temperature for the absorption layer and micro-motions of nanoparticles have been examined. Flow behavior of laminar sub-layer with nanofluids is studied. It is found that micro-motions of nanoparticles completely change the flow behavior of nanofluids, destroy the flow boundary layer, and therefore make heat conduction through laminar sub-layer easier.

2. Simulation Method and model

2.1 MD method

MD method assumes that the motions of particles (atoms or molecules) could be described by classical dynamics theory. [18] For a system consisted of *N* particles, the motion of each particle satisfies the differential equation of Newton's second law, which is written as:

$$\frac{d}{dt}\left(m_i \frac{dr_i}{dt}\right) = -\frac{\partial \Phi}{\partial r_i} \tag{1}$$

where m_i represents the quality of the *ith* particle, r_i is the position vector of the *ith* particle, t represents time and Φ is potential energy.
Through numerically solving motion equation for each particle in the simulation system, the coordinate positions and momentum of each particle could be obtained, which is the trajectory of particles in phase space. Furthermore, the macroscopic transport property could be obtained by statistical calculation on the basis of MD simulation.

MD method relates the thermal conductivity of fluid to equilibrium heat flow autocorrelation function through Green-Kubo equation [12], which is written as:

$$k = (1/V3k_BT^2)\int_0^\infty \langle J(t)\cdot J(0)\rangle dt \tag{2}$$

where k is thermal conductivity of fluid, k_B is Boltzmann's constant, T is thermodynamics temperature, V is volume, J is instantaneous microscopic heat flux vector, $<J(t)\cdot J(0)>$ is the heat flow autocorrelation function, and angular brackets denote taking overall average. Heat flow vector can be calculated by:

$$J = (1/V)\left[\sum_j (1/2)e_j v_j + (1/2)\sum_{i\neq j}(r_{ij} : F_{ij})\cdot v_j\right] \tag{3}$$

where F_{ij} represents the interaction between atom i and atom j, which is ruled by interaction potential function. And e_j represents surplus energy of the atom j, which is calculated by:

$$e_j = \sum_j (1/2)m_j v_j^{\,2} + (1/2)\sum_{i\neq j}\Phi_{ij} \tag{4}$$

2.2 Potential function

The accuracy of MD simulation relies on potential function [12]. Generally, MD simulation employs empirical or semi-empirical potentials to describe interactions between atoms. Lennard-Jones (L-J) is a commonly used potential for describing interactions between liquid atoms, which is written as:

$$u_{ij} = 4\varepsilon\left[\left(\frac{\sigma}{r_{ij}}\right)^{12} - \left(\frac{\sigma}{r_{ij}}\right)^{6}\right] \tag{5}$$

where r_{ij} represents the distance between atom i and atom j (r_{ij}=r_j-r_i), ε is characteristics potential energy of interactions between molecules, σ is characteristics diameter of molecule. The first item represents the repulsive force caused by overlap of the inner electrons or ions in an atom, and the second item represents the electrostatic attraction between dipoles.

During the MD simulation, resultant force on each atom meets the Newton's law[18]:

$$m_i \frac{d^2 r_i}{dt^2} = \sum_{j\neq i} F_{ij} \tag{6}$$

where m_i and r_i represent the quality and position of atom i, and the interaction between atom i and atom j can be written as:

$$F_{ij} = -\frac{\partial u_{ij}}{\partial r_{ij}} \tag{7}$$

Substitute equation (5) to equation (7), the interactions between molecules can be written as:

$$F_{ij} = \frac{48\varepsilon}{\sigma^2}\left[\left(\frac{\sigma}{r_{ij}}\right)^{14} - \frac{1}{2}\left(\frac{\sigma}{r_{ij}}\right)^{8}\right] r_{ij} \tag{8}$$

In the present work, the nanofluid model for thermal conductivity calculation is composed of water molecules and copper nanoparticle. While the nanofluid model for boundary-layer flow consists of argon atoms and copper nanoparticles. The potential functions that decide interactions between water molecules or copper atoms are SPCE and EAM potentials, respectively. And the parameters for these potentials could be easily found in literatures [14,19,20]. L-J potential is used for deciding interactions between Ar-Ar, Ar-Cu, and H_2O-Cu. The L-J potential parameters that are suitable for H_2O-Cu and Ar-Cu can be calculated according to Lorentz-Berthelot mixing rule, which is written as [12]:

$$\sigma_{sl} = \frac{\sigma_{ss} + \sigma_{ll}}{2} \tag{9}$$

$$\varepsilon_{sl} = \sqrt{\varepsilon_{ss} \varepsilon_{ll}} \tag{10}$$

The L-J potential parameters used in the present work are listed in Table 1.

Table 1 L-J potential parameters for nanofluids

Atom 1	Atom 2	ε (g Å^2/*fs*^{2})	σ (Å)
O	O	1.0568e-028	3.1506
Cu	Cu	6.5582e-027	2.338
O	Cu	8.3251e-028	2.7443
Ar	Ar	1.6540e-028	3.405
Ar	Cu	1.0415e-027	2.872

2.3 Simulation model

The MD simulation model for thermal conductivity is consisting of H_2O molecules and Cu nanoparticles, as shown in Fig.1. The nanoparticle is spherical in shape with the diameter of 2nm. The simulation box is cubic with the side length of 5.9386 nm. Therefore the volume concentration of nanofluids is 2%. The total amount of particles in the simulation box is 4682, which satisfies the requirement of particle number independence as Sarkar et al. reported [12]. The particles (H_2O molecules or Cu atoms) in the simulation box are initially placed according to FCC (Face Centered Cubic) lattice. With a time period of relaxation, the particles in the simulation box would adapt to a state of relative balance. The relaxation process lasts for 250ps, with time step of 2fs. Fig.2 shows the internal energy of simulation system evolution during relaxation. The figure indicates that the simulation system is in equilibrium state after relaxation and ready for thermal conductivity calculation.

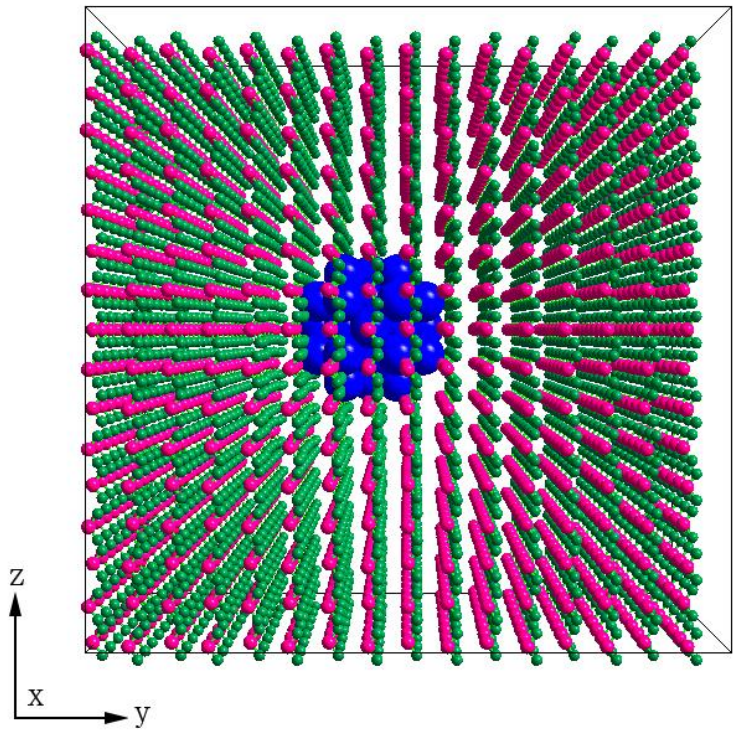


Fig.1 Simulation model for thermal conductivity of nanofluids

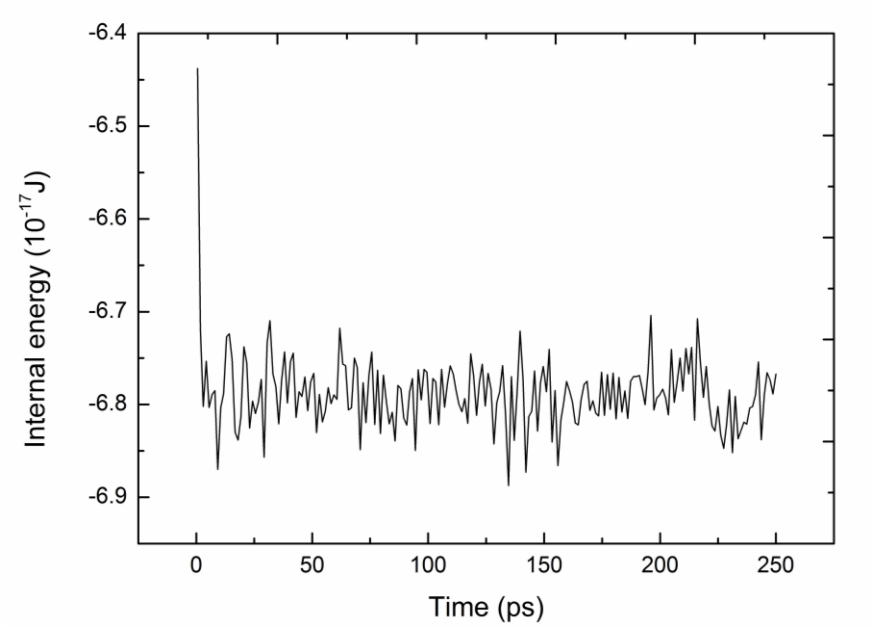


Fig.2 Internal energy of water-based nanofluids during relaxation

The MD model for nanofluid boundary-layer flow is consisting of Ar atoms as the base fluid and 35 Cu nanoparticles, as shown in Fig.3. The model chooses Cu plate as the fixed wall. In order to calculate a computational domain as large as possible, Ar is chosen as the base fluid rather than water because of the computing workload. The size of simulated fluid region is 8.853nm×61.971nm×45.9675nm. The Cu nanoparticle is spherical with the diameter of 2nm. In the model, 7 Cu nanoparticles are installed along the y axis and 5 nanoparticles are installed along the z axis. The volume concentration of nanofluids in the flow model is 1%. All the particles in the models are initially arranged in FCC lattice. The total amount of particles in the simulation box is

more than 350 thousand. Along the x and y axis periodic boundary is adopted, and along the z axis the boundary condition is defined as needed. The relaxation process lasts for 1600ps, with time step of 2fs. Fig.4 shows the internal energy of simulation system evolution during relaxation. The figure indicates that the simulation system is in equilibrium state after relaxation and ready for boundary-layer flow calculation. In order to imitate the boundary-layer flow, a five layer of Ar atoms at the top of the fluid region is set as the "moving plate" with a constant velocity along y axis to impose flow driving force. In this case, the velocity is set to be 5m/s. The simulation system is created by "LAMMPS" code [21], which is a famous freely available molecular dynamics code developed by Sandia National Labs.

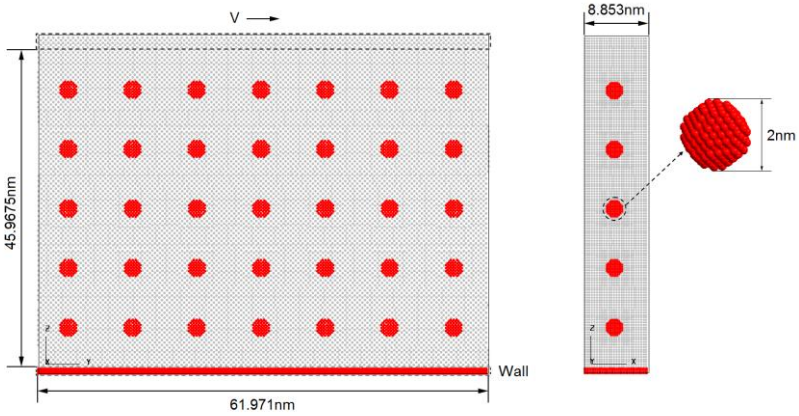


Fig.3 Simulation model for boundary-layer flow of nanofluids

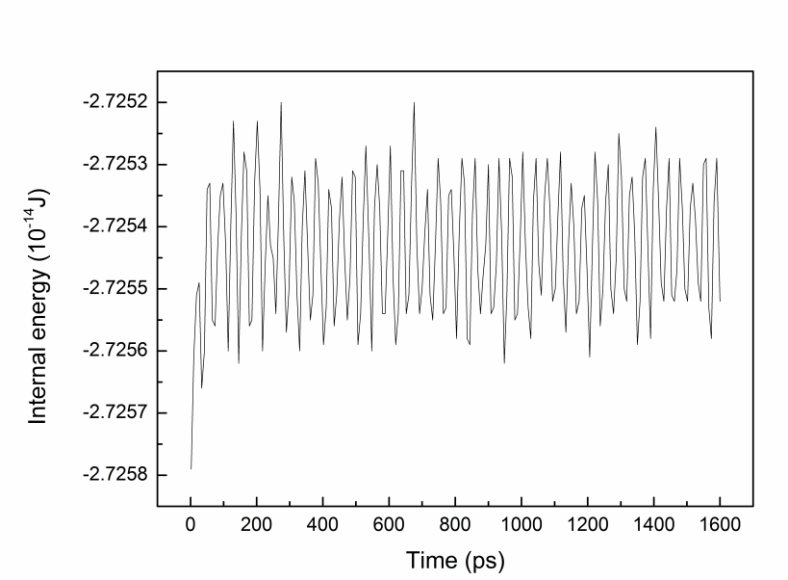


Fig.4 Internal energy of Ar-based nanofluids during relaxation

3. Results and discussion

3.1 Enhanced thermal conductivity of nanofluids

Thermal conductivity of single-phase water at different temperature is firstly calculated by MD simulation. The results are shown in Fig.5. By comparing the calculated results with experimental data, it is found that under the condition of this work at 313K the calculated thermal conductivity of water (0.63518W/m•K) is the closest to experimental data (0.635W/m•K). When calculating thermal conductivity of water by MD simulation the maximum error is 11.4% at 343K.

The subsequently calculated thermal conductivity of nanofluids is compared with the MD simulated thermal conductivity of water. The calculated thermal conductivities of nanofluids at different temperature are shown in Fig.6. In the figure the MD simulated thermal conductivity for single-phase water are also shown for comparison. It could be easily found that the thermal conductivity of nanofluids is higher than that of single-phase water under the same temperature, which is attributed to the adding of nanoparticle. By MD simulation the temperature effect for thermal conductivity of nanofluids is successfully simulated. The increase ratio of nanofluid thermal conductivity is over 30% compared to that of base fluid. The markedly increased thermal conductivity is one key reason for explaining convective heat transfer enhancement in nanofluids.

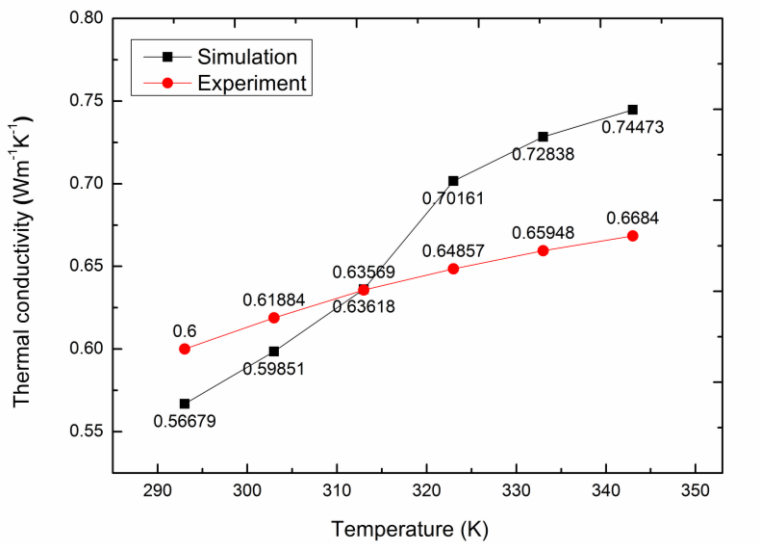


Fig.5 Thermal conductivity of water at different temperature

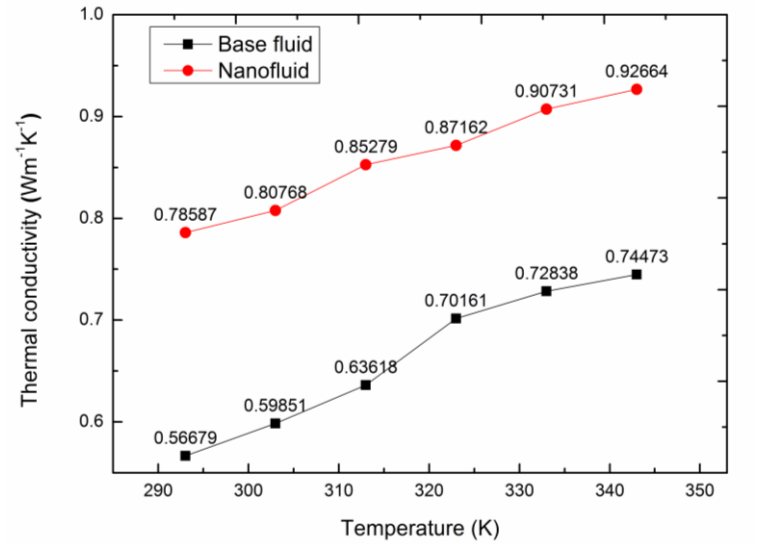


Fig.6 Thermal conductivity of water-based nanofluids at different temperature

The absorption layer around nanoparticles is believed to be an important mechanism for heat conduction augmentation in nanofluids [13,22,23]. Fig.7 shows the calculated number density of absorption layer on the surface of nanoparticles at different temperature. It could be found that the influence of temperature on absorption layer is mainly reflected in the first peak value of number density curve. In the figure, the first peak value of number density at 353K is the largest,

while the first peak value of number density at 293K is the smallest. This phenomenon indicates that the absorbed liquid molecules are more compact at high temperature. The main reason is that the random motion of molecules at high temperature is more intense. The liquid molecules have more opportunities to approach nanoparticle which is conducive to absorbing.

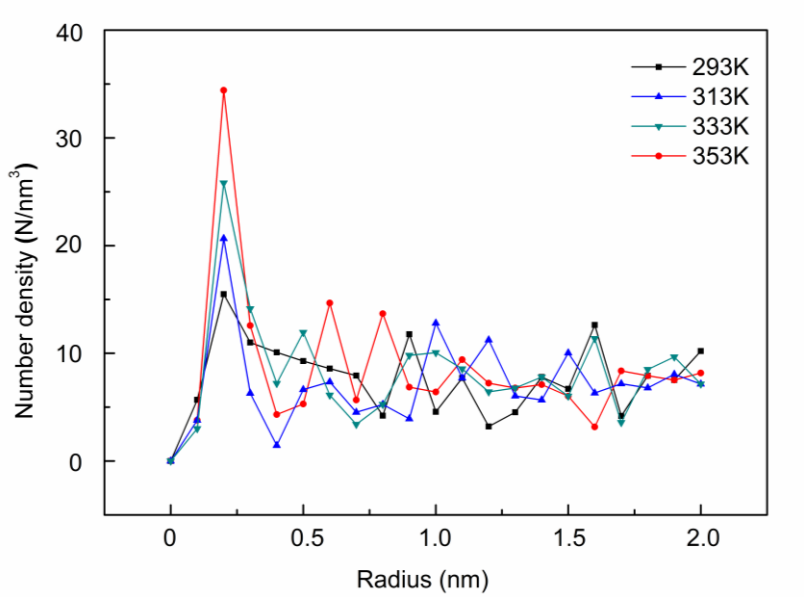


Fig.7 Absorption layer around nanoparticle at different temperature

Micro-motion of nanoparticles is also proposed to be an important reason for heat conduction augmentation in nanofluids. [24,25] Through MD simulation, the instantaneous velocity and position coordinates of each atom could be obtained. The translational and rotational velocity of nanoparticles could be acquired by defining a group for the Cu atoms within the nanoparticle. With commands provided by LAMMPS the time-averaged translational and rotational velocity of the atom group could be calculated and outputted. The influence of temperature for nanoparticle micro-motion has been studied in this work. Fig.8 shows the comparison between translational velocities of nanoparticle at different temperature. It could be found in the figure that the translational velocity of nanoparticles varies with temperature increase. The translational velocity of nanoparticle is larger at higher temperature. At 293K the average translational velocity of nanoparticle is about 3m/s, with peak value up to 7m/s; At 313K the average translational velocity of nanoparticle is about 4m/s, with peak value up to 9m/s; At 333K the average translational velocity of nanoparticle is about 5m/s, with peak value up to 9m/s; At 353K the average translational velocity of nanoparticle is about 7m/s, with peak value up to 12m/s. The translational velocity of nanoparticle varies between positive and negative values, which indicate that the translational movements of nanoparticle are mainly due to the scale-effect-induced random Brownian motion. Fig. 9 shows the comparison between rotational velocities of nanoparticle at different temperature. It could be found in the figure that the rotational velocity of nanoparticles varies with temperature increase. The rotational velocity of nanoparticle at higher temperature is also larger. At 293K the average rotational velocity of nanoparticle is about 2×10^9 rad/s, with peak value up to 8×10^9 rad/s; At 313K the average rotational velocity of nanoparticle is about 3×10^9 rad/s, with peak value up to 1×10^{10} rad/s; At 333K the average rotational velocity of nanoparticle is about 4×10^9 rad/s, with peak value up to 8×10^9 rad/s; At 353K the average rotational velocity of nanoparticle is about 6×10^9 rad/s, with peak value up to 1×10^{10} rad/s. The rotational velocity of nanoparticle also varies between positive and negative values, which also indicate that the rotational movements of nanoparticle are mainly due to the scale-effect-induced random Brownian motion.

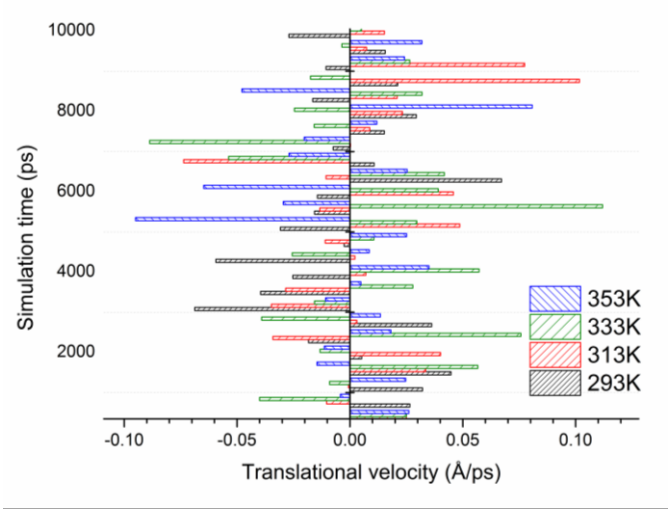


Fig.8 Comparison between translational velocities of nanoparticle at different temperature

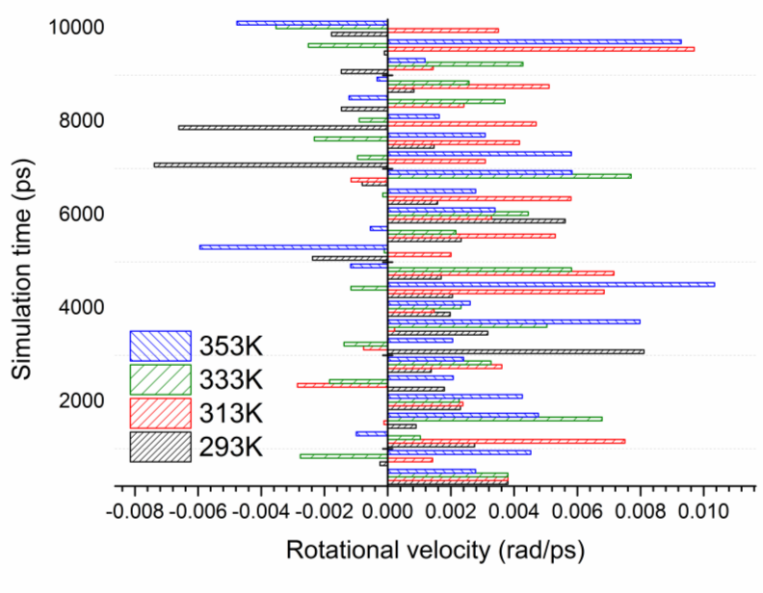


Fig.9 Comparison between rotational velocities of nanoparticle at different temperature

3.2 Changed flow boundary layer by nanoparticles

The present work further examined the influence of nanoparticles for flow boundary layer of nanofluids. The instantaneous velocity and position coordinates of each atom could be obtained through MD simulation. It is found that the adding of nanoparticles provides additional perturbation for the boundary layer flow. Fig.10 shows the desired moving direction of surface atoms in nanoparticle in a transient simulation time. It could be found in the figure that the surface atoms in nanoparticles are intended to move outwards. However, due to the strong interactions between Cu atoms, the nanoparticle still keeps its original shape. Yet, the trend of nanoparticle motions could impact its surrounding fluidic molecules. Fig.11 illustrates the snapshot for the holistic desired moving direction of nanoparticles in nanofluid boundary-layer flow. The laminar sub-layer of nanofluids is filled with irregularly moving nanoparticles. Therefore the flow boundary layer is destroyed. This phenomenon is conducive to heat conduction through the laminar sub-layer, and is beneficial to convective heat transfer in nanofluids. Fig.12 illustrates the snapshot for boundary layer flow of nanofluids during MD simulation. It could be found that the entire nanoparticles move along with the base fluid along the main flow direction, despite

nanoparticles located at different positions move with different velocities. Nanoparticles that are farther from the fixed wall move faster. In addition, the nanoparticles also move along the normal direction of fixed wall, which could be considered as the inducement of secondary flow. This movement behavior is very conducive to enhance heat transfer through the laminar sub-layer. Fig.13 shows the calculated translational velocity of a nanoparticle in the flow field. It could be found in the figure that the nanoparticle moves with main flow along y axis. The translational velocity along main flow direction is approximately 4m/s. Along the other two directions the nanoparticle also has quite a large kinematic velocity. The translational velocity of nanoparticle along these directions varies between positive and negative values. And the absolute value of translational velocity of nanoparticle along these two directions reaches 2m/s. Furthermore, the nanoparticles also rotate at a very high speed in the base fluid, as shown in Fig.14. The rotational velocity of nanoparticle varies between positive and negative values. And the absolute value of rotational velocity of nanoparticle in three directions could reach 4×10^9 rad/s. The total movements of nanoparticles in flow boundary layer completely change the flow behavior of nanofluids, destroy the flow boundary layer, and therefore make heat conduction through laminar sub-layer easier. The changed flow boundary layer by nanoparticles is another reason for explaining the convective heat transfer augmentation by nanofluids.

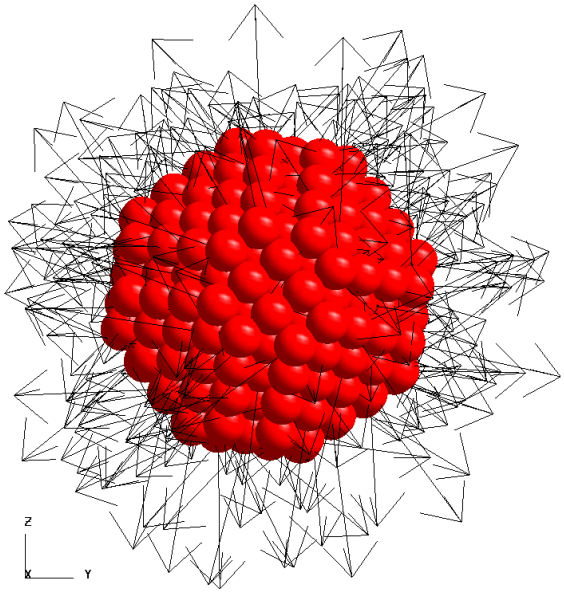


Fig.10 Desired moving direction of surface atoms in nanoparticle

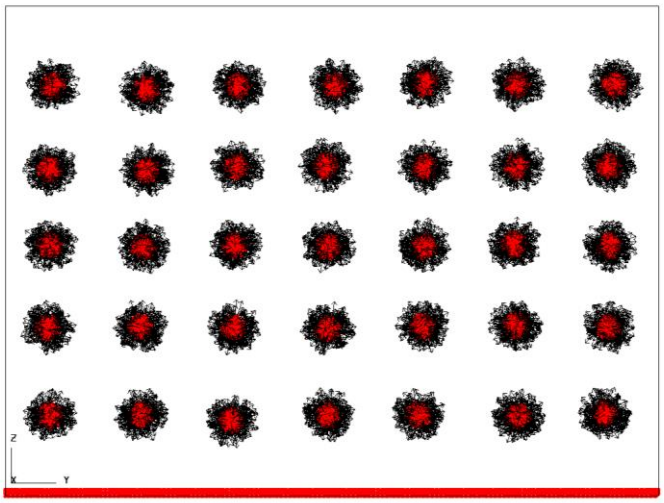
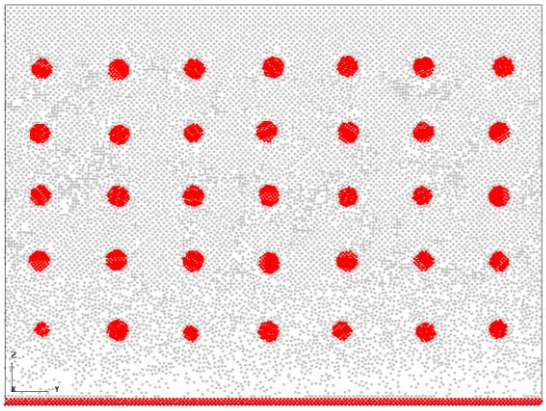
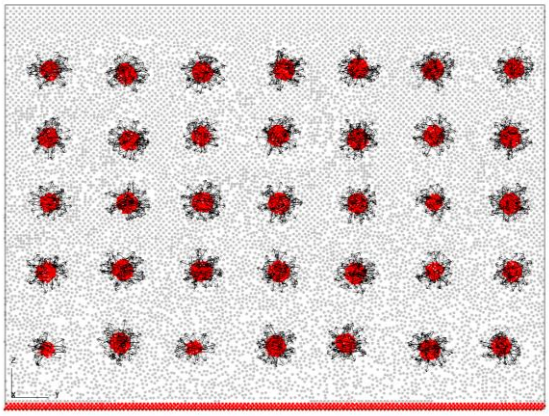


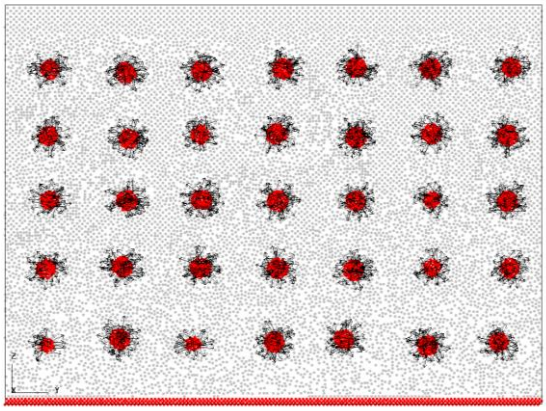
Fig.11 Snapshot for the holistic desired moving direction of nanoparticles in nanofluids



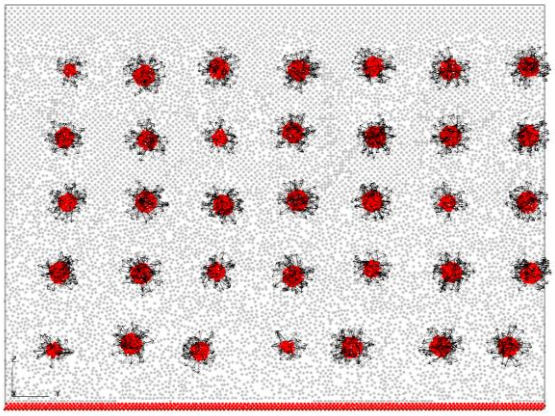
(a) 0 ps



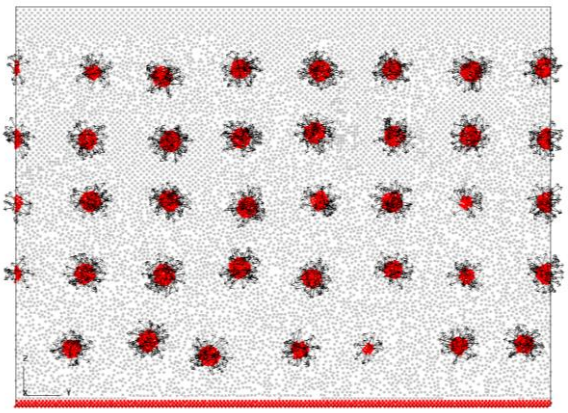
(b) 320 ps



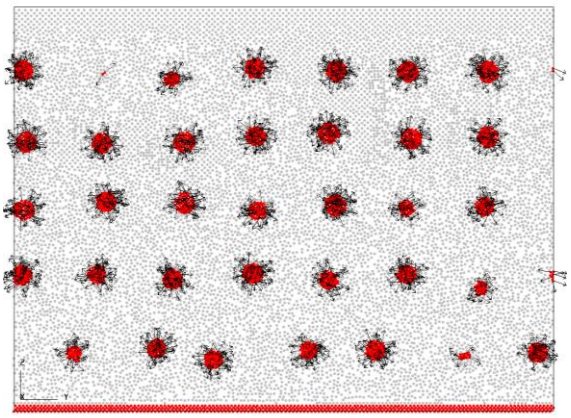
(c) 640 ps



(d) 960 ps



(e) 1280 ps



(f) 1600 ps

Fig.12 Snapshot for boundary layer flow of nanofluids

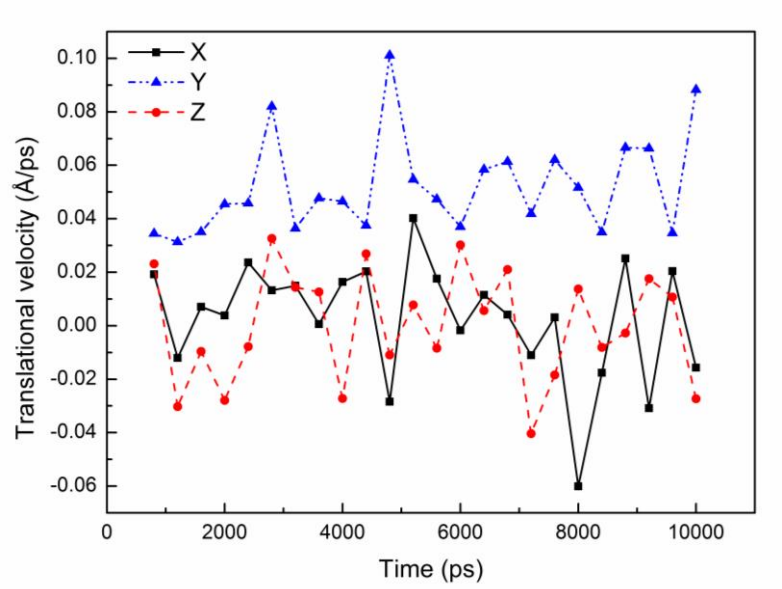


Fig. 13 Translational velocity of nanoparticle in boundary-layer flow

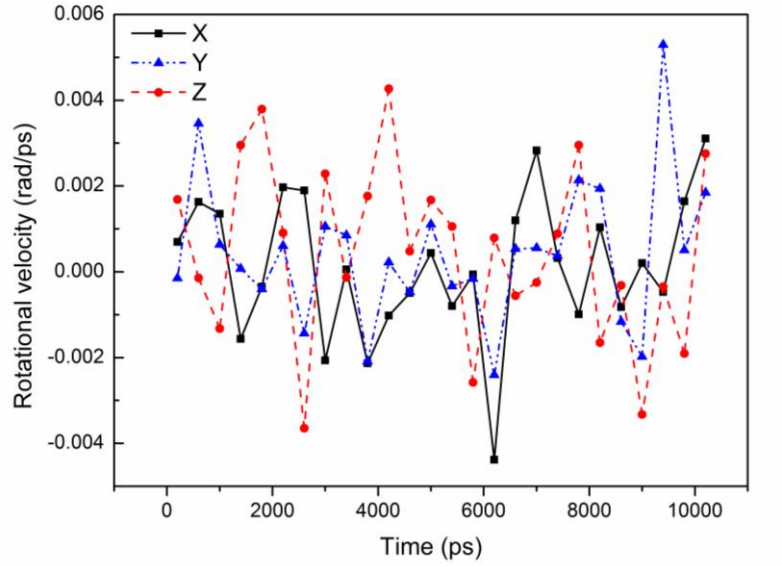


Fig. 14 Angular velocity of nanoparticle in boundary-layer flow

4. Conclusions

The present work is intended to reveal the microscopic mechanisms behind convective heat transfer augmentation with nanofluids. MD simulations for thermal conductivity calculation and boundary-layer flow of nanofluids have been performed. The following conclusions have been obtained:

(1) Abnormal increased thermal conductivity of nanofluids is one key reason for convective heat transfer enhancement in nanofluids. Thermal conductivity of nanofluids is calculated to be markedly increased by MD simulation. Thermal conductivity of nanofluids increases with increased temperature. The influence of temperature for absorption layer and micro-motions of nanoparticles are examined.

(2) The changed flow boundary layer by nanoparticles is another key reason for convective heat transfer enhancement in nanofluids. Through MD simulation, it is found that the adding of nanoparticles provides additional perturbation for the boundary layer flow. Especially

for the laminar sub-layer, the micro-motions of nanoparticles completely change the flow behavior of nanofluids, destroy the flow boundary layer, and therefore make heat conduction through laminar sub-layer easier.

Acknowledgments

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