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Original Research Paper

Modeling and simulations of nanofluids using classical molecular dynamics: Particle size and temperature effects on thermal conductivity

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ABSTRACT

We use molecular dynamics simulations to investigate the thermal conductivity of argon-based nanofluid with copper nanoparticles through the Green-Kubo formalism. To describe the interaction between argon-argon atoms, we used the well-known Lennard-Jones (L-J) potential, while the copper–copper interactions are modeled using the embedded atom method (EAM) potential that takes the metallic bonding into account. The thermal conductivity of the pure argon liquid obtained in the present simulation agreed with available experimental results. In the case of nanofluid, our simulation predicted thermal conductivity values larger than those found by the existing analytical models, but in a good accordance with experimental results. This implies that our simulation is more adequate, to describe the thermal conductivity of nanofluids than the previous analytical models. The efficiency of nanofluids is improved and the thermal conductivity enhancement is appeared when the particle size and temperature increase.

1. Introduction

Nanofluids are a new class of Nano technological materials, which are prepared by suspension of nanoparticles, nanotubes, or nanofibers with length on the order of 1–50 nm in conventional fluids [1–3]. Furthermore, many works [4,5] have shown that nanofluids exhibit very high thermal conductivity even for low concentrations of suspended nanoparticles. In this context, many experimental and theoretical studies on the thermal conductivity of solid-particles suspensions have been carried out since the classic works of Maxwell [6]. However, these works have been largely restricted to suspensions with millimeter- or micronized particles. The proposed approach in engineering fluids with heat transfer properties, based on the emerging field of nanotechnology, has recently been the subject of many researchers. Lee et al. [4] have measured thermal conductivity of nanofluids containing (CuO) and (Al_2O) nanoparticles in ethylene glycol and water, using the volume fractions of 1 to 6% to obtain a significant enhancement of thermal conductivity, they found that not only particle shape, but size is deemed to be controlling in improving the thermal conductivity of nanofluids. Another, work carried out by Choi et al. [7,8], has reported that a small amount of copper nanoparticles or carbon nanotubes dispersed in oil and/or ethylene glycol increases the inherently poor thermal conductivity of the liquid.

So, to understand the mechanisms responsible for the enhancement of thermal conductivity in nanofluids, Keblinski et al. [13] have suggested four potential mechanisms: effects of nanoparticles clustering, molecular-level layering of the liquid at the nanoparticle surface, nature of heat transport in nanoparticles, and Brownian motion of nanoparticles. To specify these mechanisms, the modeling and simulation techniques can be investigated to understand many properties at the microscopic scale. Molecular dynamics (MD) simulations are generally applied in many areas to obtain structural and thermal properties with very good accuracy [9–12]. In this context, Brownian dynamics simulations, based on equilibrium Green–Kubo approach [14], have been interested in calculating the effective thermal conductivity of nanofluids and confirmed the success of this approach to reproduce the experimental results [15]. Barrat and Vladkov [16] have used molecular dynamics simulation to calculate thermal conductivity of nanofluids through the classical Maxwell Garnet equation model [17], they

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found that there is an absence of collective effects. Xue et al. [18] have studied the effect of the liquid–solid interface on the interfacial thermal resistance using non-equilibrium molecular dyanmics simulations and they concluded that the simple monoatomic liquid around the solid particle do not have any influence on the thermal transport neither normal nor parallel to the surface.

In this work, we have investigated the thermal conductivity of a nanofluid (Ar-Cu) consisting of argon-based fluid and suspended copper nanoparticles using the Green–Kubo formalism to compute the thermal conductivity of Ar-Cu Nanofluid using equilibrium molecular dynamics simulation. So, to show the volume fraction and temperature effect on thermal conductivity, we investigated the thermal conductivity corresponding to several volume fractions at the same temperature around 86 K, while, the effect of temperature has been shown within a temperature range from 86 K to 102 K. The obtained results show that thermal conductivity of nanofluids depends on both temperature and volume fraction of nanoparticles.

The remainder of this paper is structured as follows. Section 2 presents a brief description of the interatomic potential used together with the simulation technique and explains the methods for preparing the nanofluids and for determining the thermal conductivity. Section 3, presents and discusses the detailed results about the fraction volume and the temperature effects on the thermal conductivity. The last section summarizes the findings of this work.

2. Modeling and computational methods

2.1. Interatomic potentials

In the present work, we have used MD simulations to investigate Argon-Copper nanofluids, where the nanoparticles are suspended in the liquid argon. The interatomic potentials between (Ar-Cu) and (Ar-Ar) are modeled by the L-J potential Eq. (1), which is sufficient and physically meaningful for many relevant applications [19–21].

$$U(r_{ij}) = 4\varepsilon\left[\left(\frac{\sigma}{r_{ij}}\right)^{12} - \left(\frac{\sigma}{r_{ij}}\right)^{6}\right] \tag{1}$$

where ε is the depth of the L-J potential well,σ is the finite distance at which the inter-particle L-J potential is zero, and r_{ij} is the distance between atoms i and j. The L-J parameters for Argon are: $\sigma_{Ar} = 0.3405$ nm and $\varepsilon_{Ar} = 1.66 * 10^{-21}$ J [22] and for Cooper is: $\sigma_{Cu} = 0.2337$ nm and $\varepsilon_{Cu} = 65.625 * 10^{-21}$ J [23]. The Lorentz Berthelot mixing rule have been used to compute the interaction between argon and copper [24], which is presented as follows:

$$\varepsilon_{Ar-Cu} = \sqrt{\varepsilon_{Ar} * \varepsilon_{Cu}} \tag{2}$$

$$\sigma_{Ar-Cu} = \frac{\sigma_{Ar-Ar} + \sigma_{Cu+Cu}}{2} \tag{3}$$

From this, the pairwise force field coefficients are: $\varepsilon_{Ar-Cu} = 10.421 * 10^{-21}$ J and $\sigma_{Ar-Cu} = 2.871$ nm. The cut-off distance beyond which the interatomic potential vanishes has been chosen to be 2.5 σ. For the interaction of copper–copper (Cu-Cu) atoms, the embedded atom method (EAM) potential is used [9]. In this formalism, the total potential energy E_i of a system of atoms is given by [25]:

$$E_i = F_i\left(\sum_{i\neq j}\rho_i(r_{ij})\right) + \frac{1}{2}\sum_{i\neq j}\varphi_{ij}(r_{ij}), \tag{4}$$

where F_i is the embedding energy of atom *i*, $r_{ij}$is the interatomic separation distance between pairs atoms, ρ_i is the atomic electron density and φ_{ij} is a short-range pair potential interaction between the atoms *i* and *j*.

2.2. Determination of the thermal conductivity

One of the most fascinating formalisms to calculate the thermal conductivity of nanofluids is the Green–Kubo (G–K) [26–28], representing an equilibrium molecular dynamics simulation. So, the thermal conductivity from Green–Kubo formula [29,30] obtained as follows:

$$K = \frac{1}{3k_B VT^2}\int_0^\infty < J(0)J(t) > dt \tag{5}$$

where *J* is heat flux vector

$$J = -kgrad(T) \tag{6}$$

In the case of a homogeneous system the thermal conductivity is reduced to

$$K = \frac{1}{k_B VT^2}\int_0^\infty < J_x(0)J_x(t) > dt, \tag{7}$$

where T is the system temperature, k_B is the Boltzmann constant, V is the volume of the simulation box. When the thermal conductivity is determined using average over three components, the heat current J is expressed as follows [31]:

$$J = \sum_i^N v_i e_i + \frac{1}{2}\sum_i^N\sum_{j\neq i}^N r_{ij}(f_{ij}.v_i) - \sum_i^N v_i h_\alpha, \tag{8}$$

where v_i is the velocity of particle *i*, e_i is per-atom kinetic and potential energies, h_α is the average partial enthalpy of specie α. The two parameters r_{ij} and f_{ij} are the displacement and interacting forces between particles *i* and *j*, respectively. N is the total number of particles. Here, the nanofluid setup model is obtained using a simulation box, which consisted initially of argon atoms arranged into a face-centered cubic (fcc) with a lattice parameter equals 5.72 Å. Firstly, the spherical central part of the box is emptied and then replaced by a spherical nanoparticle of copper atoms to create the suspension situation of (copper, solid) - (argon, liquid). All simulations have been performed with a time step of 4 *fs* under periodic boundary conditions, using the LAMMPS package [32]. The entire sample is then relaxed at T = 300 K using an NVT ensemble during 10^6 time steps, ensuring the presence of liquid argon phase. In the second step the system is cooled down to the desired temperature using an NPT ensemble. The visualization of the obtained structures has been carried out using OVITO [33].

Fig. 1 shows a snapshot of the structure obtained with different values of nanoparticle radii. The liquid phase of the Ar was checked by both visualization and using radial distribution functions. On the other hand, to investigate temperature and volume fraction effects on the enhancement of thermal conductivity, the thermal conductivity has been computed for a volume fraction ranging from 0.19% to 7.66% for temperatures of 86 K, 90 K, 94 K, 98 K and 102 K.

So, to compute the thermal Conductivity of liquid argon, the simulation boxes are relaxed using the canonical ensemble (NVT) during 1.1×10^6 time steps. Finally, the production run consists of another relaxation during 8000 steps using (NVE) micro canonical ensemble.

To validate our results, the calculated thermal conductivity of argon liquid is compared to the available numerical and experimental data, which are previously reported by Kang [34], Lee et al. [35] and Loulijat et al. [36]. Our simulation shows that the thermal conductivity of argon liquid is 0.135 W/m·K at a temperature of 86 K.

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