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Molecular dynamics study of thermal resistance of solid-liquid interface in contact with single layer of nanoparticles

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ABSTRACT

In a nanoscale system, thermal resistance at a solid-liquid interface can be considerable. Nanoparticle deposition on the solid-liquid interface is able to influence the solid-liquid interfacial thermal resistance (ITR). The influence of the nanoparticle layer on the thermal resistance can be critical, for an example, in a heat transfer of nanoparticle suspension fluids. In the present study, by means of non-equilibrium classical molecular dynamics (MD) method, we numerically investigated changes in the solid-liquid ITR, liquid density near the interface, heat flux components through single layer of carbon nanoparticles. Under the present conditions, the carbon nanoparticle layers on the solid-liquid interface influenced the liquid density and the heat transport near the solid-liquid interface, and the ITR. The ITR decreased, as the liquid density in the nanoparticle layer increased. The change in the liquid density near the solid-liquid interface was one of significant factors that affected the ITR. When the nanoparticle layer significantly decreased the ITR, the energy transport from the wall to the nanoparticles was enhanced, and the energy transfer inside the nanoparticles increased. Therefore, the nanoparticles were able to extract and store the thermal energy from the solid wall, which decreased the ITR.

1. Introduction

Material interfaces significantly impact on thermal flow since energy carrier may scatter at the interfaces. Solid-liquid ITR, which was a discontinuous temperature across the interface, was experimentally observed when liquid helium was used. The solid-liquid ITR is now known as Kapitza resistance. In a nanoscale system, the solid-liquid interfaces can significantly influence the ITR since the interface regions are comparable with a characteristic length of the system. The interfacial thermal resistance changes with its interface geometry, and wettability. For an example, the interfacial thermal conductance of hydrophilic and hydrophobic interface, which were functionalized with a self-assembled monolayer (SAM), was experimentally measured with time-domain thermoreflectance [1]. The study showed that the ITR at hydrophobic interfaces were 2–3 times greater than that at hydrophilic interfaces. To investigate thermal energy transport across the interfaces, MD simulations have been performed so far. A previous MD study [2] numerically showed that the ITR decreased, as an intermolecular interaction between solid and liquid molecules, which was relevant to the wettability, increased. Pressure dependence on the solid-liquid ITR was numerically investigated [3]. It showed that the pressure dependence of the solid-liquid ITR strongly depended on the wettability of the solid surface. Nanostructured interfaces geometry affects the solid-liquid ITR. Other MD studies [4,5] employed nano slit structure on the solid-liquid interface, and showed that the nano slit was able to reduce the ITR depending on the nanostructure geometry. Surface modification also changes the ITR. For examples, previous MD studies showed a graphene sheet [6], and a SAM [7] on the solid-liquid interface influenced the ITR. In addition to the nanostructured surface, nanoparticle deposition onto the solid surface are able to influence the solid-liquid ITR. In the presence of nanoparticles in a heat transfer fluid, such as a nanofluid, the nanoparticles may deposit on the heat transfer surface. As a result, heat transfer from the solid wall can change. For thermal conductivity measurements of nanofluids, transient hot-wire method has been widely employed [8]. The method needs heating hot-wires in direct contact with the nanofluids. So far, the ITR between the wire surface and the nanofluids has been assumed negligible in evaluating thermal conductivity in the case of the transient hot-wire method. Yet, its influence on the solid-liquid ITR has not been quantified. Therefore, it is necessary to understand how and how much the nanoparticles on the solid-liquid interface change the ITR.

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Based on the background, we numerically investigated influences of a nanoparticle layer in contact with the solid surface on the ITR by means of classical non-equilibrium MD method. In the present study, the ITR were evaluated with changes in the intermolecular interaction parameters between the liquid molecules and the nanoparticle atoms, and that between the liquid molecules and the wall atoms, as well as the liquid pressure. Heat flux through the computation system was decomposed into each energy flux component. Based on this, we investigated the thermal energy transport mechanism that the nanoparticle layer changed.

2. Numerical method

Figs. 1 and 2 show calculation models of a nanoparticle deposited surface employed in the present study, including a molecular liquid region confined by the top and bottom walls, defined as a unit cell for the molecular dynamics simulations. We tested two types of calculation models. Unit cell sizes were $3.23 \times 3.23 \times 5.00\ \mathrm{nm}^3$ (model (a); see Fig. 1), and $4.44 \times 4.44 \times 5.00\ \mathrm{nm}^3$ (model (b); see Fig. 2). In model (a), the nanoparticles were in diameter of approximately 1.0 nm, and in a form of fullerene C_{60}. Nine nanoparticles formed a single layer on the solid wall. On the other hand, in model (b), the nanoparticles were in diameter of approximately 2.0 nm, and in an amorphous form consisting of 542 carbon atoms. Four nanoparticles formed a single layer on the solid wall. The carbon atoms were modeled with the Brenner potential for the interatomic potential [9]. The initial configurations of the amorphous nanoparticles were set up in a preliminary non-equilibrium molecular dynamics simulation. In the preliminary simulation, carbon atomic vapor was gradually cooled to form an atomic cluster by the velocity scaling method. A spherical region of 2.0 nm diameter was extracted from the formed carbon atomic cluster, and the extracted clusters were employed as the nanoparticles.

Atoms that consisted of the solid walls were the Lennard-Jones (LJ) particles that had mass of Pt (atomic weight: 195 g/mol). Liquid atoms were the LJ particles that had mass of Ar (atomic weight: 39.9 g/mol). Atoms that consisted of the nanoparticles were the particles that had mass of C (atomic weight: 12.0 g/mol). The 12-6 LJ potential were employed to understand the energy transfer mechanism for the simply shaped liquid molecules, which had three translational degrees of freedom. The interaction between the liquid molecules and the wall atoms, and that between the liquid molecules and the nanoparticle atoms were expressed by the 12-6 LJ potential function. The standard parameters of σ and ε were determined by the Lorentz-Berthelot combining rules [10]. The potential parameter ε between the liquid molecules and the solid wall atoms, and that between the liquid molecules and the nanoparticle atoms were expressed as the relative value, by respectively multiplying α_{wl} and α_{nl}. Those parameters employed in the present study were shown in Table 1. If the value of α is high, the interaction intensity is strong, and thus droplet contact angle is small. The potential energy parameter $\alpha_{\mathrm{wl}}\varepsilon_{\mathrm{wl}}$ varied from 6.88×10^{-22} to 1.73×10^{-21} J, and the parameter $\alpha_{\mathrm{nl}}\varepsilon_{\mathrm{nl}}$ varied from 3.45×10^{-22} to 1.38×10^{-21} J, as shown in Table 1. The potential energy between the liquid molecules and the solid wall atoms, ϕ_{wl}, and that between the liquid molecules and the nanoparticle atoms, ϕ_{nl}, are expressed as follows:

$$\phi_{\mathrm{wl}} = 4\alpha_{\mathrm{wl}}\varepsilon_{\mathrm{wl}}\left\{\left(\frac{\sigma_{\mathrm{wl}}}{r}\right)^{12} - \left(\frac{\sigma_{\mathrm{wl}}}{r}\right)^{6}\right\} \tag{1}$$

$$\phi_{\mathrm{nl}} = 4\alpha_{\mathrm{nl}}\varepsilon_{\mathrm{nl}}\left\{\left(\frac{\sigma_{\mathrm{nl}}}{r}\right)^{12} - \left(\frac{\sigma_{\mathrm{nl}}}{r}\right)^{6}\right\}, \tag{2}$$

where r is the distance between atoms and molecules. When α_{wl} was equal to 0.0510 or 0.128, the corresponding droplet contact angle was approximately 95° or 30° [11]. In the present study, we employed that the cut-off distance for the 12-6 LJ potential was 3σ, and that for the Brenner potential was 0.20 nm. With the present parameter set, the nanoparticles stayed on the solid-liquid interface.

Newton's equations for the solid atoms and the liquid molecules were numerically integrated by the leapfrog method with the time interval of 0.25 fs. Periodic boundary conditions were employed in the x and y directions. Solid walls consisted of four layers of the solid particles. Its outermost layers were fixed. Temperatures of the second layers from the outermost layers of the walls, which were phantom atom layers, were controlled constant by Langevin method [7]. Temperature gradient was formed in the molecular liquid region by controlling the temperatures of the top and bottom walls to be 115 K and 85 K, respectively.

The liquid region was divided into 10 slabs in z direction, and the temperature profile was calculated. By extrapolating the liquid temperature profile to the solid-liquid interface, each interface temperature was evaluated (see Fig. 3). We linearly extrapolated temperature profile of the liquid region slabs where $z = 0.0$–0.5, 0.5–1.0, 1.0–1.5 nm, and evaluated the solid wall surface temperature at $z = 0.0$. The temperature difference between the solid wall surface temperature and the nearest nanoparticle region temperature was denoted by ΔT_{b}. The ITR, R_{b}, was calculated as follows:

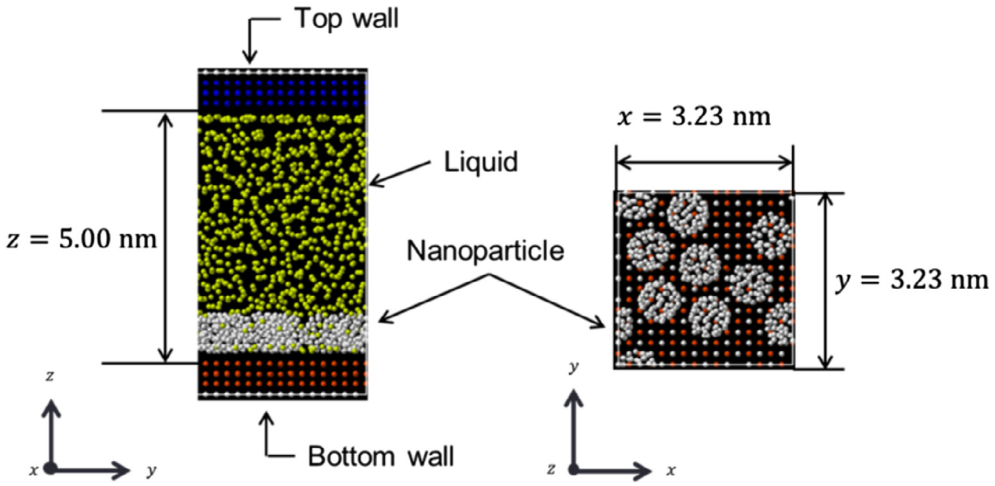

Fig. 1. Calculation model: model (a).