Molecular Dynamics Simulation of Normal and Explosive Boiling on Nanostructured Surface

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1Introduction

With advances in ultrafine manufacturing technology, surface structures in nanoscale can be fabricated for different applications in engineering and science. Boiling on surfaces with and without nanostructures is an important phenomenon that is widely used in advanced micro/nano electromechanical systems, nanoelectronic cooling, and so on. Surface effects in nanoscale significantly change the behavior of boiling due to the high surface-to-volume ratio in micro/nanofluidic system. Although these effects cause enhancement of heat transfer [1–3] and change the wettability at liquid–solid interface [4–8], from a microscopic point of view, nanostructures are kinds of ultraflat surfaces because their roughness are in nanoscale. However, due to the complexity of physical mechanism in the nanoscale, the difference between heat transfer mechanisms at macroscale and nanoscale is not fully understood despite its general importance. Boiling on a flat surface with nanostructure is very complex and significantly bottlenecks the thermal management of the micro- and nanodevices.

Using nanotextured surfaces is one of the passive efficient methods for enhancing boiling heat transfer. Nanostructures are an attractive option for tackling the thermal management issues, particularly for cooling of high heat flux electronic devices. Recently, there is a certain tendency to investigate boiling processes in the framework of experimental study [9–16] or classical fluid mechanics and heat transfer with some adjustable model parameters. We believe that, however, considerations on the molecular scale are required for a full understanding and development of physically sound models of these processes.

With significant progresses in computer hardware and software, it is now possible to investigate transport phenomena in nanoscale via computer simulation. Molecular dynamics simulation is an attractive and powerful means for studying phenomena in nanoscale; hence, it is an ideal tool to study the behavior of the phase transition from liquid to vapor. Most existing molecular simulation studies of evaporation and boiling have focused on heterogeneous phase transition of liquid from a molecular level on a flat solid surface [17–21]. To the best of the authors’ knowledge, the effect of spherical nanostructures on phase transition, especially explosive boiling, has not been studied. Depending on the surface temperature, phase transition from a solid surface can occur in an explosive boiling or a normal boiling [10]. Explosive boiling is the process of rapid phase transition from highly superheated liquid to vapor [22–24]. This process is so violent and fast that it resembles an explosion; it can be triggered by laser heating [25] or when liquid droplets are suddenly and drastically heated by immersion in a hot medium.

Although it is possible to precisely control the nanoscale structure on a solid surface with the advanced ultrafine manufacturing technology, all solid surfaces have some levels of unevenness. Study of evaporation and boiling on nanostructures is of great importance because a better understanding of these phenomena can enhance energy efficiency in many engineering processes. Despite considerable efforts on modeling of flow boiling heat transfer in nanosystems, there are limited studies for the evaporation/boiling on nanoporous surfaces. Therefore, the theoretical work reported in the present study aims at developing a methodology for predicting the enhancement in net evaporation of thin film on a flat surface due to the addition of spherical nanostructures on the surface, using nonequilibrium molecular dynamics simulations. The base fluid chosen for this study is argon while copper was used as solid material. This combination was selected due to the ready availability of the potential functions required in the simulation. However, by applying pertinent potentials and other parameters in the simulation, the methodology can be extended to the case of any material combination. Both normal and explosive
boiling over a nanostructured surface will be investigated. The thickness of liquid was kept constant while the size of nanoparticles on the surface varies which result in different longitudinal and transverse distances between particles and height of nanostructures. Therefore, two different scenarios were studied: (i) height of nanostructure is less than liquid film thickness, and (ii) same height for nanostructure and liquid film.

2 Simulation Method

The simulation box is a cube with size of 7.2 nm (x) × 7.2 nm (y) × 80 nm (z), which contains liquid argon, vapor argon, and solid copper atoms as the wall material. In the x- and y-directions, periodic boundary conditions are used, so an infinite plane of evaporation is simulated. The solid atoms are arranged in the bottom of simulation box and liquid atoms initially placed on a face-centered cubic (FCC) lattice corresponding to density at saturation temperature of 90 K and the rest of domain was filled with vapor argon atoms. Figure 1 illustrates the initial configuration of liquid, vapor and solid atoms. The nanostructured surfaces formed by joining four equal-sized copper nanoparticles on the flat surface with diameters equal or less than the thickness of liquid film. To study the effects of particles size and interparticle distance on the evaporation and boiling, three different particle sizes (i.e., \( d = 1 \text{ nm}, d = 2 \text{ nm}, d = 3 \text{ nm} \)) were used to generate nanostructured surfaces.

Figure 2 shows the configurations of nanostructure surfaces used in this study. The solid wall and nanoparticles were represented by layers of FCC (111) copper atom corresponding to its density of 8.96 g/cm³. Nine layers of copper atoms were constructed to form the bottom wall, and different layers have different functions. From the bottom to top, the first layer of atoms is fixed to avoid any migration of sample; the next two layers were set as heat source from which heat flux was generated; and the last six layers were set as solid walls through which heat is conducted to the fluid. The total number of atoms in the system is 10,200–13,716 for different nanostructured surfaces. The top boundary condition in the z-direction is mirror in which the argon atom is reflected back to the domain without any loss of momentum or energy, i.e., the top boundary is elastic and adiabatic. The size of simulation box in the z-direction was chosen to be sufficiently large so that the top boundary has no effect on evaporation.

In a molecular dynamics simulation choosing an appropriate intermolecular potential that describes the interaction between molecules is very crucial. In this work, the well-known Lennard-Jones (L-J) 12-6 potential is used for interaction of atoms in both solid and liquid phases

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\varphi(r_{ij}) = 4\epsilon \left[ \left( \frac{\sigma}{r_{ij}} \right)^{12} - \left( \frac{\sigma}{r_{ij}} \right)^{6} \right]
\]  

where \( r_{ij} \) is the distance between the two atoms, \( \epsilon \) is the potential well depth, and \( \sigma \) is the characteristic length that is a finite distance at which the interparticle potential become zero. The potential well depth and characteristic length for argon–argon and copper–copper as well as copper–argon were collected from Refs. [26–28]. In any MD simulations, calculation of the forces acting on atoms is the most time-consuming task. When the distance between the two atoms is greater than the cut-off distance, the interactive force between the two atoms is zero. Those atoms with distances longer than the cut-off distance should be excluded from the force calculation. To speed up the simulation, we chose cut-off radius equal to \( 4\sigma_{Ar-Ar} \).

For surface A, we compared the results for different time steps, i.e., 1, 2, and 5 fs and observed insignificant difference between them. Therefore, in this study, time integration was carried out by the velocity Verlet algorithm with time step of 5 fs. The simulation contains three different steps. First using equilibrium molecular dynamics, the entire system was set at a uniform temperature of 90 K under Langevin thermostat method for 500 ps (Stage I). Once the total energy and temperature in the simulation does not change anymore, the simulation enters to Stage II. The thermostat is removed for the fluid domain and the Langevin thermostat was changed into microcanonical (NVE) ensemble; the system was run for 1 ns. However, the temperatures of both nanoparticles and solid surface were still fixed at 90 K by the thermostat in Stage II. Empirically, if the temperature and potential energy are stable during the subsequent NVE ensemble stage just after the first
stage, the system can be treated as in the thermal equilibration state. In order to check whether the argon is in equilibrium state, we monitored the argon atoms’ density, pressure and temperature during the equilibration period. For the flat surface case the temperature and pressure fluctuate around 90 K and 1.35 bar and the density profile at the end of equilibrium has good agreement with the experimental data. Finally, the temperature of solid wall was set at higher temperatures (290 K and 170 K) using canonical (NVT) time integration via Nose/Hoover thermostat and simulations were run for enough steps to achieve equilibrium (Stage III). In the aforementioned ensembles, N denotes number of atoms, V is volume, T stands for temperature, and E means the total energy. In NVT ensemble, the volume and temperature will be constant, whereas in NVE ensemble, the volume and total energy will be constant.

3 Results and Discussions

3.1 High Superheat Degree Case. Simulation at higher superheat degree correspond to explosive boiling was first carried out for both flat surface and surfaces with nanostructure. The simulation results of temperature history of argon and wall material for high temperature case ($T = 290\text{ K}$) are shown in Fig. 3. It is noted that the argon temperature denote the temperature of all of the argon atoms, i.e., liquid and vapor argon. During equilibrium period, because the temperature of solid wall was set to be 90 K and the system is in thermodynamic equilibrium, it is expected that the temperature argon is 90 K. The numerical results show that during equilibration period the mean argon temperature is indeed 90 K. The wall responds very quickly to the temperature rise and reaches the equilibrium in less than 40 ps while at the onset of boiling the argon temperature rises very rapidly and at some point it begins to decrease. The quick rise of the wall temperature causes liquid argon near the copper wall to exceed the critical point temperature and vaporize while other layers are still in liquid phase. The vaporized argon has high pressure and pushes
Fig. 5 Number density—high temperature case 290 K

Fig. 6 Net evaporation number—high temperature case 290 K

Fig. 7 Net evaporation number at fast region of evaporation—high temperature case 290 K
the liquid above it; this results in separation of liquid from the solid wall. The temperature of liquid falls in a specific interval of time and then starts to increase again. This is due to the separation of argon from the wall surface. The vapor region near the wall has low density and it prevents energy flow from hot wall to the separated liquid above the vapor region.

It is clear from Fig. 3 that the nanostructures lead to higher argon temperature as results of increased solid–liquid interface area and interaction. Due to larger surface area of nanostructures compared with the flat surface, quick rise of wall temperature causes more energy transfer to the vapor atoms near the wall so the separation temperature increases due to the presence of nanoparticles on the surface. Furthermore, with increasing size of nanoparticles, the argon temperature increases further and it reaches to equilibrium sooner. The equilibration time for cases with the argon temperature increases further and it reaches to equilibrium on the surface. Furthermore, with increasing size of nanoparticles, more energy transfer to the vapor atoms near the wall so the separation temperature increases due to the presence of nanoparticles on the surface. The vapor region near the wall has low density and it prevents energy flow from hot wall to the separated liquid above the vapor region.

In order to give a molecular insight into the phase transition, the simulation system for each case at different times is shown in Fig. 4. As can be seen, before the start of boiling process, the interface between liquid argon and solid wall is clearly visible and the liquid argon shows a lower meniscus for surface D due to surface tension. For all cases the film thickness decreases during the period of simulation; the system reaches to near equilibrium condition for cases C and D but the rate of film thickness reduction and equilibrium time vary for these surfaces. For the flat surface case, because of transfer of energy is only from the bottom, the evaporation lasts longer and the system reaches at steady state after a longer time; this means that faster evaporation on nanostructured than that on the flat surface due to higher surface area in contact with fluid for nanostructured surfaces. As described earlier, at a specific time the liquid layers near the solid surface vaporizes and pushes the liquid above vapor to separate from the solid surface as a large cluster of liquid. The separation of liquid cluster starts at around 2.53 ns, 2.40 ns, 2.21 ns, and 2.10 ns for cases A, B, C, and D, respectively. For the flat surface case, the liquid layer above the surface separates from the solid as a large cluster of liquid as described earlier. For cases with nanostructures, with increasing size of nanoparticles on the surface, the surface area of solid material in contact with the liquid increases so that the temperature gradient in the liquid layer become less extensive than that of the flat surface. Therefore, a smaller cluster of liquid moves upward and the rest of liquid migrate as individual atoms in a well dispersed tiny cluster. It is worth to mention that due to higher heating area in the cases with larger nanoparticles, the volume of liquid layer increases and it moves upward; the separation starts from liquid just above the nanoparticles.

Figure 5 shows the number density profile in the z-direction for different times and surfaces. The computational domain along the z-direction is divided into 190 slices and the number of atoms in each slice is calculated to obtain the average density of each slice. It is clear that a cluster of liquid moves away from the flat surface at different times. The region of high density peaks appearing in the curve show the locations of floating liquid argon. For example, at $t = 2.2$ ns, the liquid cluster is between 25 and 30 nm for the flat surface case, 30–35 nm for surface B and 41–46 nm for surface C; this means that with increasing the size of nanoparticles from 1 to 2 nm, the liquid cluster travels faster. For surface D, however, due to small number of liquid atoms and larger heat area and vaporization, there is not any large liquid cluster. Another important result is that the number density gradually flattens after and before each peak which show the disappearance of the vapor–liquid phase interface.

Figures 6 and 7 show the net evaporation molecular number in the simulation period. The evaporation rate was calculated by counting the change of argon atoms in the vapor region. It can be seen that after the start of evaporation/boiling, the number of evaporation molecules increases linearly as the simulation time proceeds. As seen for high temperature case, the number of evaporation molecules for nanostructured surface C and D is almost constant after $t > 1.8$ ns while for surfaces A and B it increases with time. This means that for surfaces C and D, there is not much significant evaporation after $t = 1.8$ ns.

3.2 Low Superheat Degree Cases. In this section, we present the results for the low temperature case ($T = 170$ K) when the initial temperature difference between argon and hot wall was more moderate. The temperature histories of argon and solid wall for all surfaces are shown in Figs. 8 and 9. Similar to previous cases, at the end of equilibrium, the liquid argon atoms cover nanostructures and create a meniscus for all nanostructured surfaces. Unlike the high temperature case, however, the temperatures for all surfaces reach equilibrium very soon. This is due to the fact that in low temperature case the liquid atoms escape into the vapor region from top layer as individual atoms or as very tiny cluster while in higher temperature case a large droplet causes that the
simulation needs more time to equilibrate. For the cases with nanostructure on the surface the distance between top liquid layer and solid atoms decreases so the liquid responds faster with presence of nanostructure. Furthermore, the nanostructures cause enhancement in interaction between solid and liquid atoms which result in quicker energy transfer from solid wall to the liquid atoms and energy transfer enhances with increasing nanoparticle size.

Figure 10 shows the density profile of the argon atoms in the direction normal to the nanostructures for different time steps. The position of the peak point that separates the vapor and liquid regions indicates the nonevaporative liquid film on the surfaces. It can be seen that for all surfaces, there is a high density region near the wall that slightly changes with time; this implies that the thickness of liquid region changes very slowly and generation of vapor is low. It is important to note that for the cases with nanostructures, a few molecular layers of argon absorbed on surface of nanoparticles result in number of nonevaporating liquid atoms increases.

Figures 11 and 12 show the evaporation number for the low temperature boiling cases. It can be seen that the evaporation numbers for all the surfaces are constants after 2 ns. In both cases without and with nanostructure, there is a region after start of
boiling with high evaporation rate. The results also indicate that net evaporation numbers of surface B and C are higher than that of the flat surface case due to large heating areas of nanostructures or reduced thermal resistance at the liquid–solid interface. On the contrary, surface D shows lower evaporation number with respect to flat case. There are two reasons for this behavior, (i) smaller number of argon molecules in the system for surface D compared with other cases, and (ii) because of higher heating area of surface D, more argon atoms attached to solid surface due to strong intermolecular forces between solid and liquid. Therefore, the size of liquid droplet and number of evaporated atoms decrease as the size of nanostructures increases. At this point, it is worth to mention that the attachment of liquid atoms to the surface formed an ultra-thin layer of argon atoms named nonevaporating layer [29,30] which has higher density with respect to other regions of liquid film. The structure of these atoms on the surface is crystal-like with obvious periodicity.

4 Conclusion

A molecular dynamics simulation has been carried out to predict the enhancement of evaporation and boiling heat transfer of argon on a flat surface with nanostructure. From the study of the temperature histories and net evaporation numbers for both flat surface and nanostructure surfaces cases, it is found that the nanostructure surfaces lead to enhancement in heat transfer from solid surface and liquid due to increase of surface area or size of nanoparticles. Furthermore, the nanostructured surfaces with particle size of 1 and 2 nm cause higher evaporation rate compared to that of the flat surface but for the case of 3 nm nanoparticle the net evaporation is lower than that of the flat case due to small number of liquid atoms in the system. For high temperature case the separation temperature depends on the size of nanostructure and there is not any nonevaporative thin liquid film on any surface. On the other hand, liquid atoms above the solid surface go into explosive boiling and droplet of a liquid move upward. The size of droplet depends on the size of nanostructures and for larger nanoparticles the liquid atoms move individually or as tiny droplets instead of large droplet. For low temperature cases, the temperatures for all surfaces reach equilibrium very soon and the liquid atoms escape into the vapor region from top layer as individual atoms or as very tiny cluster.

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Nomenclature

- $d$ = diameter of nanoparticles
- $r_{ij}$ = distance between molecule $i$ and $j$
- $t$ = time
- $T$ = temperature (K)
- $x$ = coordinate in $x$-direction
- $y$ = coordinate in $y$-direction
- $z$ = coordinate in $z$-direction

Greek Symbols

- $\varepsilon$ = energy parameter of LJ potential
- $\varphi$ = potential energy
- $\sigma$ = length parameter of LJ potential

References


