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## NORMAL AND EXPLOSIVE BOILING OF ARGON ON NANOSTRUCTURED COPPER SURFACE: A MOLECULAR DYNAMICS STUDY

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### ABSTRACT

Nonequilibrium molecular dynamics (NEMD) is carried out to investigate the normal and explosive boiling of thin film adsorbed on a metal substrate whose surface is structured by an array of nanoscale spherical copper particles. It is found that superheat degree and size of nanoparticles have significant influences on the location of atoms at multiple times and net evaporation rate. For the cases with nanostructure, liquid respond very quickly and evaporation rate increase with increasing the size of particles from 1 to 2 nm while it decreases for particles diameter of 3 nm.

### INTRODUCTION

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 for model molecular system. So far, most of molecular simulation studies of evaporation and boiling have focused on heterogeneous phase transition of liquid from a molecular level on a flat solid surface [1-4] and to the best of the authors' knowledge, the effect of spherical nanostructures on phase transition, especially explosive boiling, has not been studied. 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 NEMD simulations. The base fluid chosen for this study is argon while copper was used as solid material. Although the more applied working fluids such as water are more appropriate for study of boiling, nevertheless Argon yields good results for quantitative conclusion. Both normal and explosive boiling [6-8] 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.

### 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. 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 of saturation temperature of 90 K and the rest of domain was filled with vapor argon 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 i.e., 10, 20 and 30 nm. Figure 1 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/cm<sup>3</sup>. In the x- and y-directions, periodic boundary conditions are used while mirror boundary condition is used for top boundary.

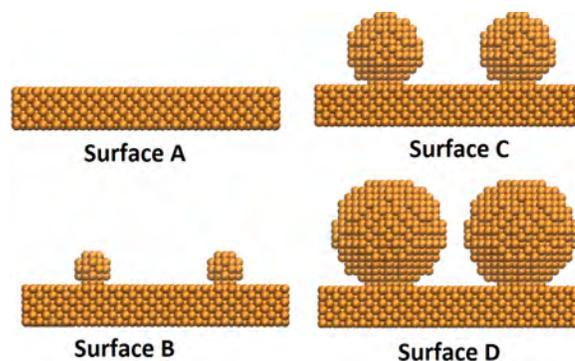


Fig. 1 Three dimensional view of different size of nanoparticles on the surface.

The well-known Lennard-Jones (L-J) 12-6 potential [1,4] with cut-off radius of  $4\sigma$  is used for interaction of atoms in both solid and liquid phases and time integration was carried out by the velocity Verlet algorithm with time step of 1 fs. The simulation contains three different steps. First using equilibrium molecular dynamics, the whole system was set at uniform temperature of 90 K under Langevin thermostat method for 500 ps (Stage I) then for the fluid domain, the Langevin thermostat was changed into NVE ensemble and the system was run for 1 ns while the temperatures of both nanoparticles and solid surface were still fixed at 90 K by the thermostat (Stage II). Finally, the temperature of solid wall was set at higher temperatures (290 K and 170 K) using NVT time integration via Nose/Hoover thermostat and simulations were run for enough steps to achieve equilibrium (Stage III).

## RESULTS AND DISCUSSIONS

The simulation results of temperature history of argon and wall material for high temperature case ( $T = 290$  K) correspond to explosive boiling are shown in Fig. 2. The numerical results show that during equilibration period the mean argon temperature is indeed 90 K. It can be seen that the wall respond very quickly to the temperature rise and reaches the equilibrium in less than 40 ps while at the start of boiling the argon temperature rises very rapidly and at some point it begin to decrease. It is worth mentioning that 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 liquid layer has high pressure which leads to pushing liquid above it and consequently separates it from the solid wall. As seen, in a specific interval of time, the temperature of liquid falls and then starts to increases. This is due to the separation of argon from the wall surface. In fact the vapor region near the wall has lower density and it prevents energy flow from hot wall to the separated liquid above the vapor region.

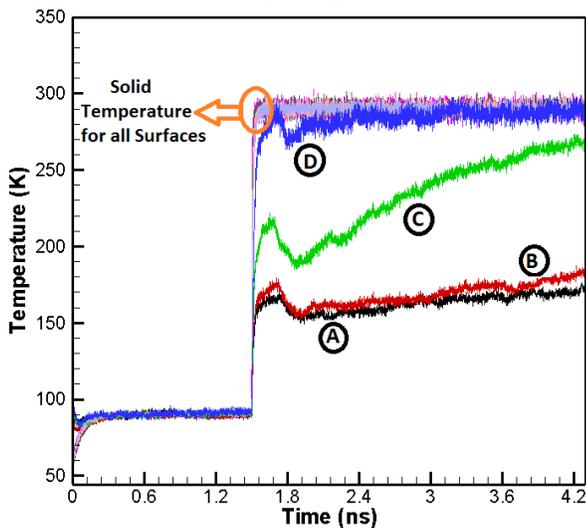


Fig. 2 Temperature history of argon and solid wall - high temperature case 290 K.

From Fig. 2, it is clear 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 the size of nanoparticles, the argon temperature increases more and it reaches to equilibrium sooner. The equilibration time for cases with  $d = 30$  nm is around 0.4 ns while the other two cases and also the flat surface case do not reach equilibrium within the simulation time.

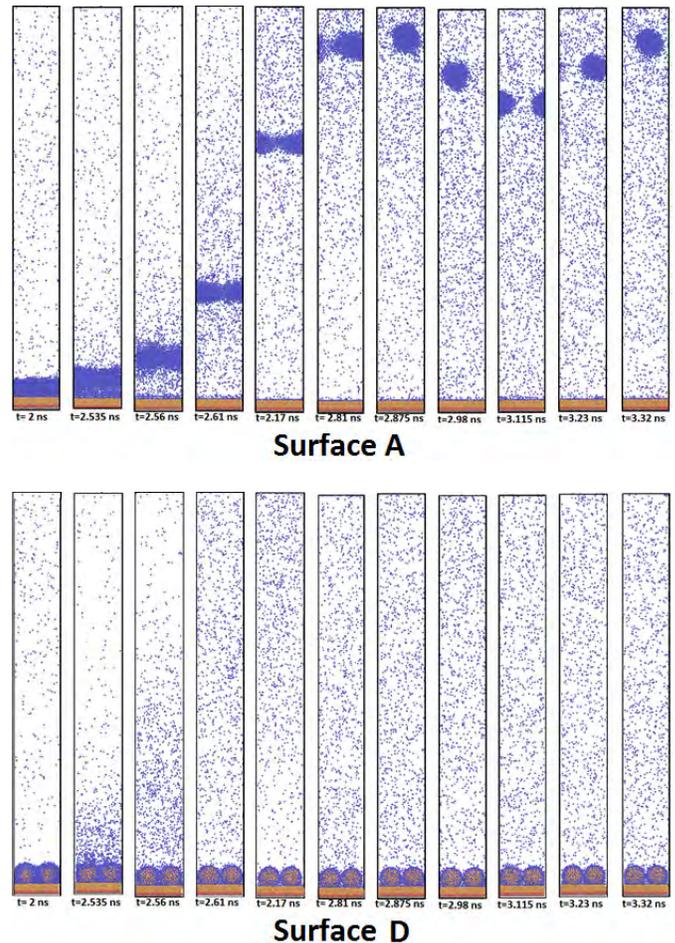


Fig. 3 Location of atoms at multiple times for different nanostructure surfaces- high temperature case 290 K.

In order to give a molecular insight into the phase transition, the simulation system for cases A and D at different times is shown in Fig. 3. 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 both cases the film thickness decreases during the period of simulation and

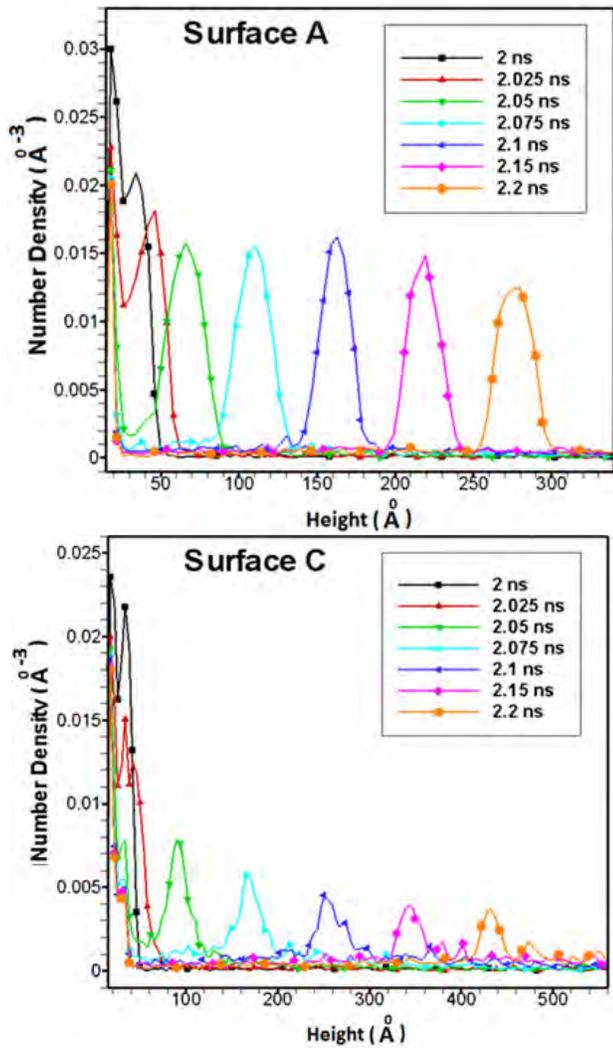


Fig. 4 Number density- high temperature case 290 K.

the system reaches to near equilibrium condition for case D but the rate of film thickness reduction and equilibrium time vary for each surface. For flat surface case, because of transfer of energy is only from the bottom, the evaporation last longer and the system reach at steady state after a longer time; this means that faster evaporation on nanostructure than that on the flat surface because of lower solid-liquid interface resistance for nanostructured surfaces. As described earlier, at a specific time the liquid layers near the solid surface vaporize, which lead to pushing the liquid above vapor to separate from the solid as a large spherical 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 liquid increases so 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.

The number-density profile in the z-direction for different times and two surface of A and C is shown in Fig. 4. It is clear that a cluster of liquid moves away from the flat surface at different times. It is worth to mention that, 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 and 41-46 nm for surface C; this means that nanoparticles on the surface leads to the liquid cluster travels faster. 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. Figure 5 illustrate the net evaporation molecular number during the simulation period. The net evaporation number 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 net evaporation number of 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.

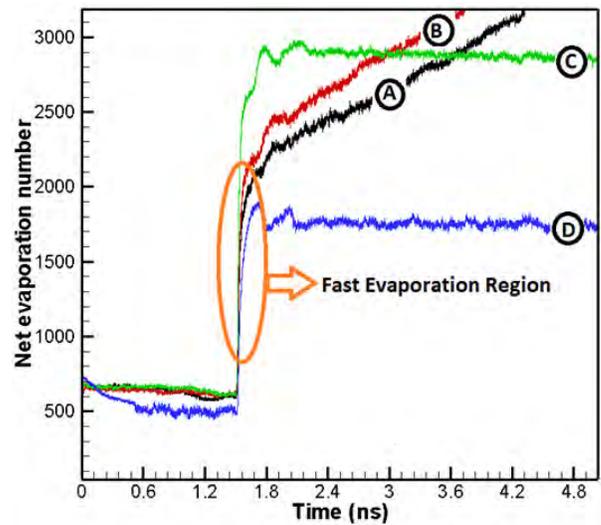
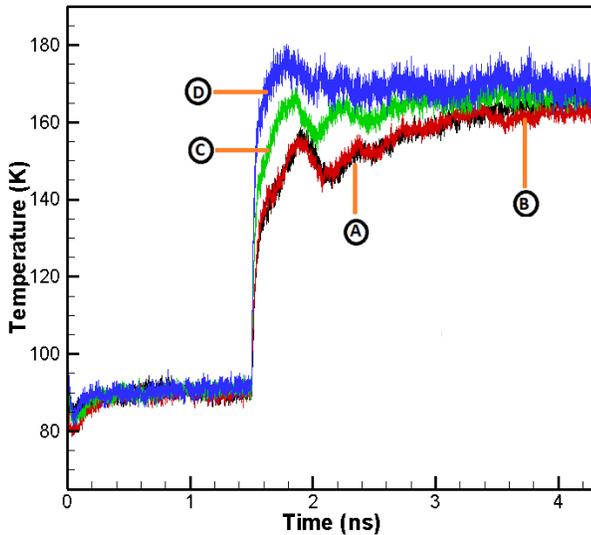


Fig. 5 Net evaporation number- high temperature case 290 K

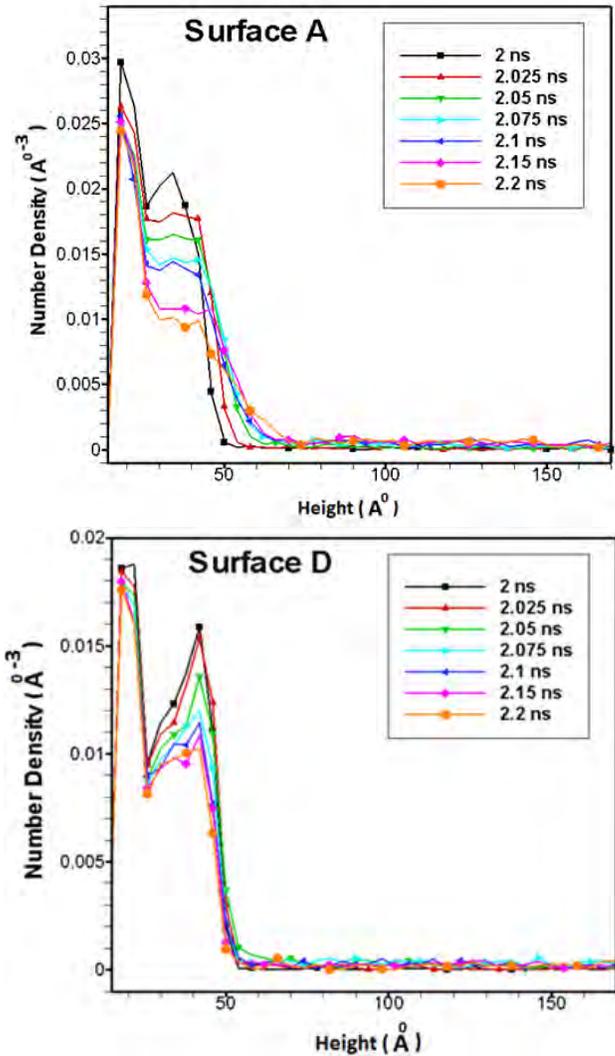
The temperature history of argon for all surfaces for low-temperature case ( $T=170$  K) correspond to normal boiling are shown in Fig. 6. Unlike the high temperature case, the temperatures for all surfaces reach equilibrium very soon. This is due to the fact that in lower temperature case the liquid 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.



**Fig. 6** Temperature history of argon-low temperature case 170K

Density profile of the argon atoms in the direction normal to the nanostructures A and D for different time steps is shown in Fig. 7. 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, 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 production 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 non-evaporating liquid atoms increases. The evaporation number for the low temperature boiling case is shown in Fig 8. 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 due to the smaller number of argon molecules in the system for surface D.



**Fig. 7** Number density- high temperature case 170 K

## CONCLUSIONS

A molecular dynamics simulation has been carried out in this paper 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. It is noticed that for high temperature case the separation temperature depends on the size of nanostructure and there is not any non-evaporative 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.

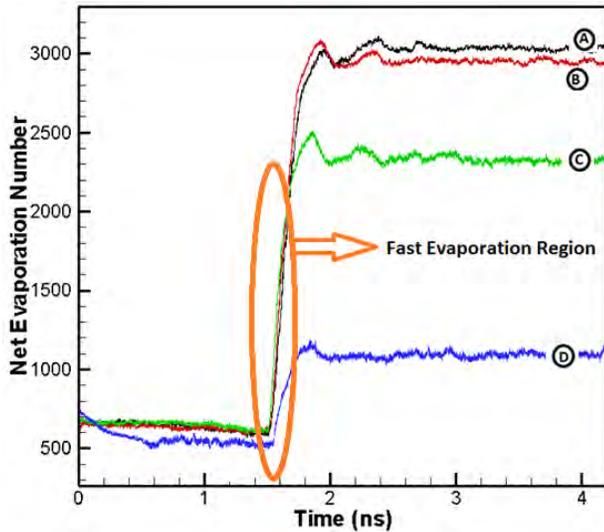


Fig. 8 Net evaporation number- high temperature case 170 K

#### ACKNOWLEDGMENTS

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