International Journal of Heat and Mass Transfer 55 (2012) 1218–1225

Contents lists available at SciVerse ScienceDirect

International Journal of Heat and Mass Transfer

journal homepage: www.elsevier.com/locate/ijhmt

A molecular dynamics investigation on evaporation of thin liquid films

Jiapeng Yu, Hao Wang *

Lab of Heat and Mass Transport at Micro-Nano Scale, College of Engineering, Peking University, Beijing 100871, China

ARTICLE INFO

Article history:
Received 23 May 2011
Received in revised form 1 September 2011
Accepted 2 September 2011
Available online 21 October 2011

Keywords:
Evaporation
Thin liquid film
Disjoining pressure
MD

ABSTRACT

Understanding of evaporation process of thin liquid film is important for studying heat transfer near the triple-phase contact line. In the present work, equilibrium molecular dynamics (EMD) and non-equilibrium molecular dynamics (NEMD) were employed to investigate the evaporation of thin liquid films in a nanoscale triple-phase system. The simulation domain was a cuboid, which consisted of an upper and a lower platinum wall, with argon fluid in between. The solid walls were first set at the same temperature and EMD was employed to achieve an equilibrium state. Then the walls were set at different temperatures and NEMD was employed to simulate the non-equilibrium state. Evaporation and condensation of thin liquid films were observed. The evolution of the film thickness as well as the net mass flux was obtained for the evaporating thin film. Interfacial accommodation coefficient was analyzed and the measured evaporation mass flux was compared with Hertz–Knudsen–Schrage equation which is based on kinetic theory of gases (KTG). Non-evaporating layer due to strong intermolecular forces was obtained and compared with theoretical models.

1. Introduction

Due to fast developments in electronics and miniaturization technologies, thermal management concerning high heat flux in limited volume has become very popular. Tremendous studies have been conducted to understand microscale heat transfer mechanisms and develop practical heat transfer solutions [1–3]. Of all the relevant hot topics, the thin liquid film near a triple-phase contact line has received an increasing attention worldwide for its significance among various phase change processes.

When a liquid wets a solid wall, the extended meniscus can be divided into three regions: a non-evaporating region where a layer of liquid-like molecules is absorbed on the wall due to strong intermolecular forces; a transition region where effects of long-range molecular forces (disjoining pressure) are felt; and an intrinsic meniscus region where capillary forces dominate. "Thin films" represent liquid films near the contact line where the film thickness is micro- to nano-scale but high heat transfer coefficient is achieved. A thin film includes non-evaporating region, transition region and the beginning section of intrinsic meniscus. Thin-film heat transfer can be found in various cases wherever there is contact line and the liquid is wetting the solid surface [3,4]. It is often referred to as "micro layer" in boiling literatures [5]. Due to very low thermal resistance across the liquid film, the thin film region owns a high heat transfer rate and it could be a major contributor to the overall heat transfer from the extended meniscus. Moreover, since this thin film region's pivotal role resembles the boundary conditions of the whole extended meniscus, it is necessary to have a good understanding of the thin film region so as to describe the extended meniscus with more accuracy [4,6].

Due to the small scale, there lie several difficulties in accurate experimentation of thin films. Molecular dynamics (MD) simulation is an effective method to investigate microscopic problems. In many MD studies, intensive focus has been given on various issues in phase change process at a liquid–vapor interface. Ten Wolde and Frenkel [7] studied the homogeneous nucleation of liquid phase from vapor. Wang et al. [8] studied thermodynamic properties of liquid–vapor coexistent systems with liquid–vapor interfaces. A sharp peak and a small valley at the thin region outside the liquid–vapor interface were found which gives the evidence of the non-equilibrium state at the interface. Tsuruta and Nagayama [9] proposed a formulation of the condensation coefficient of vapor molecules at liquid–vapor interface based on MD studies and transition state theory. Furthermore, an inverted temperature profile in an evaporation–condensation system was found and explained. In thin liquid films, the difference between the normal component of pressure and the isotropic pressure in the bulk liquid with which the film is in contact is the disjoining pressure. For it is a representation of the intermolecular forces in the micro region, disjoining pressure attracts much attention. Bhatt et al. [10] and Carey and Wemhoff [11] investigated the disjoining pressure in Lennard-Jones liquid films by MD simulations and compared with thermodynamic theory. Both of their works were based on the Gibbs–Duhem equation, and MD simulations were

* Corresponding author. Tel.: +86 10 8252 9010.
E-mail address: hwang@coe.pku.edu.cn (H. Wang).

0017-9310/$ - see front matter © 2011 Elsevier Ltd. All rights reserved.
doi:10.1016/j.ijheatmasstransfer.2011.09.035

Nomenclature

A	Hamaker constant (J)
h_{fg}	latent heat of evaporation (J/kg)
ke	kinetic energy (J)
k_B	Boltzmann constant (J/K)
m''	mass flux (kg/m^2 s)
m''_c	condensation mass flux (kg/m^2 s)
m''_e	evaporation mass flux (kg/m^2 s)
$\overline{M}$	molecular weight (kg/kmol)
N_A	Avogadro's number (mol^{-1})
N_{all}	number of argon atoms
P	pressure (N/m^2)
P_c	capillary pressure (N/m^2)
P_d	disjoining pressure (N/m^2)
P_{d0}	disjoining pressure (N/m^2)
P_l	liquid pressure (N/m^2)
P_{sat}	saturation pressure (N/m^2)
$P_{sat\text{-}ref}$	reference saturation pressure (N/m^2)
P_{sat}	saturation pressure (N/m^2)
P_V	vapor pressure (N/m^2)
P_{v_equ}	equilibrium pressure (N/m^2)
r	distance between molecules
$\overline{R}$	universal gas constant (J/kmol K)
S	area of film (m^2)
T	temperature (K)
T_{lv}	liquid–vapor interface temperature (K)
T_W	solid wall temperature (K)
T_V	vapor temperature (K)
ε	energy parameter of L-J potential
ρ_{gn}	gas number density (m^{-3})
ρ_l	liquid density (kg/m^3)
ρ_{ln}	liquid number density (m^{-3})
σ	length parameter of L–J potential
ϕ_r	potential energy

Subscripts

c	condensation
e	evaporation
equ	equilibrium
g	gas
l	liquid
lv	liquid–vapor interface
sat	saturated
v	vapor
w	solid wall

used to simulate the different state of the liquid/vapor system, so as to calculate the disjoining pressure indirectly.

A triple-phase system with evaporating and condensing processes has been studied. Maruyama and Kimura [12] measured the thermal resistance over a solid–liquid interface, and it was found to strongly depend on the wettability. Nagayama et al. [13] investigated the evaporation and condensation processes between hot and cold liquid films in contact with different nanostructured surfaces, and it revealed that the evaporation rate of the ultra-thin liquid film manifested a larger value than that of the flat surface. Ji and Yan's [14] study shows that there is a non-evaporating liquid film with thickness in nanometers, existing on the heated solid surface. The thickness of such an ultra-thin liquid film varies only slightly with the number of argon molecules but decreases with a rise in heating substrate temperature.

In the present work, a comprehensive MD and theoretical investigation on the thin film evaporation/condensation process was conducted. With EMD simulation, the influence of thin film on equilibrium vapor pressure was seen and the interfacial accommodation coefficient was analyzed. In NEMD simulation, the evaporative mass fluxes driven by the different temperature gradients were obtained and compared with the theoretical results. In addition, the thicknesses of non-evaporating layers were measured and compared with the calculated results based on the disjoining pressure theory. The comparisons proved the validity of the theories but also indicated the room for further improvement.

2. Simulation system and method

Argon atoms were used to generate liquid and vapor domain while platinum atoms were used to generate solid walls. The well-known L-J potential was employed to calculate the intermolecular forces, for its quantitatively reasonable description of non-polar atoms:

$$\phi_r = 4\varepsilon\left\{\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^{6}\right\} \quad (1)$$

The parameters are listed in Table 1. LAMMPS code was used to construct the simulation system.

Table 1
Parameters for LJ potential.

Parameters	ε/(kcal/mol)	σ/Å
Platinum–Platinum	12.0181	2.475
Argon–Argon	0.2404	3.405
Platinum–Argon	1.6996	2.940

As shown in Fig. 1(a), two similar cases, named Cases A and B, were constructed in this work. Both of them were cubic boxes with periodic boundary conditions applied in x and y directions. The solid walls were represented by layers of FCC (face-centered cubic) platinum atoms. In Z direction, argon atoms were sandwiched between the two solid walls. The difference between Cases A and B was only the temperature of the solid walls (detailed in Table 2). The time step for the simulations was 5 fs, and the cut-off radius was set as 3.5 σ beyond which the potential was just truncated without switching or shifting. The parameters were common in similar investigations [12,14].

In both cases, 9 layers of platinum atoms were constructed to form each of the two solid walls, and different layers match with different functions. From outside to inside, the first layer of platinum atoms stayed still as a boundary wall to keep the volume of the system constant; the following inside three layers were set as heat source or heat sink from which heat flux was generated or dissipated; the last five layers were set as solid walls through which heat conducted to argon fluids.

In EMD simulation, the equilibrium states of Cases A and B at 101 K and 110 K respectively were studied. Two steps were taken to realize the simulation. First using constant NVT (i.e. constant atom number, constant domain volume and temperature-canonical ensemble) time integration via Nose/Hoover thermostat method (simple Nose/Hoover thermostat), the whole system was set at a uniform temperature 100 K. After 500,000 steps of integration, a solid–liquid–vapor system of 100 K was achieved. Then for the fluid domain, the NVT ensemble was changed into NVE ensemble (certain number, volume and energy-micro-canonical ensemble) and was run for 1,000,000 steps while the temperature of the