

Free surface transition and momentum augmentation of liquid flow in Micro/Nano-scale channels with hydrophobic and hydrophilic surfaces[†]

Budiono¹, Doyoung Byun^{1,*}, Vu dat Nyugen¹, Jihoon Kim¹ and Han Seo Ko²

¹*Department of Aerospace and Information Engineering, Konkuk University,
1 Hwayang-dong, Gwangjin-gu, Seoul 143-701, Korea*

²*School of Mechanical Engineering, Sungkyunkwan University, Korea*

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Abstract

We propose a novel micro/nano-scale nozzle structure, featuring an interfacial line between the hydrophilic and the hydrophobic surfaces for a jetting system, such as an inkjet head or electrospray devices. This research will investigate the impact of the interfacial line on flow instability and momentum augmentation as the liquid meniscus moves across the line. The research methods used in this paper, in respect to micro- and nano-scale channels, are computational fluid dynamics (CFD) and non-equilibrium molecular dynamics (MD), respectively. With the growing interest in micro/nano electromechanical systems (MEMS/NEMS), many studies have been conducted to develop an advanced micro/nanofluidic system. However, until now, there have been few in-depth studies on passive flow control in micro and nano nozzles using the hydrophilic and hydrophobic surface characteristics. In this research, the sequential arrangement of hydrophilic and hydrophobic surfaces in the nozzle is presented along with an investigation into how flow instability and momentum augmentation are going to be applied to an efficient micro/nano jetting system. When a liquid meniscus arrives at the interfacial line between hydrophilic and hydrophobic surfaces, the meniscus shape changes from concave to convex and the fluid motion near the wall stops until the concave shape is fully converted. Because the momentum should be conserved, the lost momentum near the wall transfers to the center region, and therefore the liquid at the center region is accelerated as it crosses the line. If we use this nozzle structure and the augmentation of the momentum near the center, a tiny droplet can be easily generated.

Keywords: Molecular dynamics; Nano channel; Computational fluid dynamics; Micro channel; Flow instability; Hydrophobic surface; Hydrophilic surface

1. Introduction

Recently, we have witnessed the rapid development of microelectromechanical system (MEMS) technologies and applications, such as inkjet print heads, accelerometers for automobile air bags, micro sensors, micro actuators, bio-chips for medical diagnostics and micro-thrusters for spacecraft. Advanced fabrication technologies can be exploited to further miniaturize

electromechanical systems and thereby bring us into the regime of nanoelectromechanical systems (NEMS) [1]. Many chemical, biological, and biophysical processes in MEMS/NEMS devices are connected to handle liquid material. An understanding of the micro/nano fluidics, a core technology in a number of miniaturized systems for electrical, propulsion, and medical applications, is critical in designing a micro/nano fluidic system. An understanding of the physical properties and the dynamical behaviors of fluid flow in micro/nano channel is crucial to the theoretical study of fluid dynamics and engineering applications in physics, chemistry, medicine, and

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*Corresponding author. Tel.: +82 2 450 4195, Fax.: +82 2 444 6614

E-mail address: dybyun@konkuk.ac.kr

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electronics. In recent years, the behavior of fluid flow at the nanometer scale has frequently been a subject of interest for the NEMS. Fluid flow in micro/nano-scale channels has been investigated experimentally and numerically for the development of various devices and the advancement of knowledge [2].

Numerical simulation of micro/nano-scale thermo-fluidic transport has attracted considerable attention, with the aim of elucidating the characteristics of fluid flow in the micro/nano world. To analyze two-phase flow, the amorphous boundary between liquid and gas phases should be carefully taken into account. In the continuum fluid regime, there are two types of numerical approaches to solve the free surface problems in the context of incompressible flow. One approach is based on front tracking where the free surfaces are explicitly tracked. The second approach is based on front capturing. The capturing methods are based on a volume-of-fluid (VOF) formulation or a level set formulation to model and define the boundary between liquid and gas phases [3, 4].

However, theories and techniques based on the continuum hypothesis cannot be used to investigate nano scale systems. When the system length reduces to the nano scale, molecular dynamics (MD) simulation, based on the statistical mechanics of non-equilibrium liquids [5], is an effective means to describe the fluid flow at the nano scale. In classical fluid mechanics, the assumption of no-slip flow at a solid surface has successfully accounted for experimental observations where the velocity vanishes at a solid surface. However, for the flows at nano scales, this assumption may no longer be applied. Using MD, fluid-solid surface interactions have been investigated, such as hydrophobic and hydrophilic surface characteristics. It is found that the velocity profile, in nano channels, deviates from that of the classical fluid flow, due to the slip boundary at the wall [6-8]. Recently, the non-equilibrium MD (NEMD) was developed to simulate the non-equilibrium physical problems, such as nano jetting and surface wettability phenomena. Droplet sizes and jet break-up characteristics follow the Rayleigh break-up theory, and higher liquid temperature is preferred to achieve faster nanojet break-up with shorter break-up length [9, 10]. Nagayama and Chen [11] studied the effects of the surface wettability on a pressure-driven flow in a nano channel with a width of 16 molecules and showed plug flow velocity profiles for a hydrophobic surface. The results show that the hydrodynamic boundary condition

at the solid-liquid contact line depends on both the wettability and the magnitude of the driving force. When the driving force exceeds a critical value that overcomes the interfacial resistance, the no-slip boundary condition may break down. They also found that the slip length is not consistent but depends on the driving force. Barrat and Bocquet [12] showed that when the contact angle is large enough, the boundary condition can dramatically differ from a no-slip condition. Moreover, the slip velocity at the stationary wall surface increases with the applied gravity force or the shear rate [13]. The slip length sharply decreases at small shear rate, and then slightly decreases with the applied shear rate.

Fluid-solid surface interactions on the hydrophobic or hydrophilic surfaces have been widely investigated. However, to date, there have been few in-depth studies on the transient flow characteristics in micro/nano channels with sequential hydrophilic and hydrophobic surface regions, such as flow instability and morphological change of meniscus.

In particular, liquid droplet, or spray injection, is an interesting research area in the fields of bioengineering, electrical engineering and aerospace engineering. Its importance can be seen in its capability to deliver a gene or drug directly into biological cells, its applicability to new display fabrication technology and its implementation as propulsion systems for nano-satellite [14-16]. A new idea of the nozzle design for a tiny droplet jetting device, featuring hydrophilic and hydrophobic surfaces inside, was proposed by Lee et al. [17]. The flow instability across the interfacial line of the hydrophilic and hydrophobic surfaces may augment momentum at the center region, where fine droplets can be produced more effectively.

In this paper, using a non-equilibrium MD simulation as well as computational fluid dynamics (CFD) methods, we investigate the effects of the interfacial line between a low-affinity region (hydrophobic surface) and a high-affinity region (hydrophilic surface) on flow instability as the liquid meniscus moves across the line. We have carried out a non-equilibrium molecular dynamic simulation of argon fluid subject to external forces in a platinum nano channel. Multi-layers of platinum molecules are used to simulate the solid walls and argon molecules are employed as a Lennard Jones (LJ) fluid. Also, the water flow in the micro-scale channel with the interfacial line is simulated to investigate the effects of the surface wettability. After these simulations, we pro-

posed a comprehensive concept of a micro/nano-scale nozzle structure, featuring an interfacial line between hydrophobic surface region and hydrophilic surface region.

2. Numerical method

2.1 Continuum approach for liquid flow in micro-scale channel

To describe the change of the liquid meniscus shape in the micro-scale channel, several models have been implemented with continuity, momentum equations, and volume fraction transport equation in the Cartesian coordinate system [4]. The flow is considered to be laminar incompressible Newtonian and isothermal.

$$\text{div}(\vec{V}) = 0 \quad (1)$$

$$\frac{\partial(\rho u)}{\partial t} + \text{div}(\rho u \vec{V}) = -\frac{\partial p}{\partial x} + \text{div}(\mu \text{grad} u) + S_{Mx} \quad (2)$$

$$\frac{\partial(\rho v)}{\partial t} + \text{div}(\rho v \vec{V}) = -\frac{\partial p}{\partial y} + \text{div}(\mu \text{grad} v) + S_{My} \quad (3)$$

Where u and v stand for velocity components of velocity vector, \vec{V} , ρ for density, p for pressure, μ for viscosity, S_{Mx} for surface tension force along x direction, and S_{My} for surface tension force along y direction, respectively.

The boundary conditions on the wall are the no-slip conditions. Constant velocity of the liquid is applied as the boundary condition on the inlet. Since information of flow is rarely known at the outlet boundary, these boundaries should be as far downstream of the region of interest as possible. To find approximations at the boundary, extrapolation is usually used along grid lines from the interior to the boundary. However, here we use a simple first order zero gradient of the derivative. The surface wettability can be described by the contact angle of the contact line of water and air. Initially, zero velocity is assumed in the micro-channel, where only air exists and water moves through the inlet.

The meniscus is modeled with the volume of fluid (VOF) technique, which was proposed by Hirt and Nichols [4]. The VOF method has been widely used to track the contact line of two immiscible fluids, such as water and air. The advantage of the VOF method is the absence of topological constraint. The distribution of the liquid in the grid systems is ac-

counted for by using a single scalar field variable, F , which defines the fraction of the volume of liquid (0:air, 1:water). The volume fraction distribution can be determined by solving the passive transport equation where \vec{V} is velocity vector.

$$\frac{\partial F}{\partial t} + \nabla \cdot (\vec{V} F) = 0 \quad (4)$$

The numerical simulation of free-surface flow is composed of two immiscible fluids which involves two tasks: (1) resolving the flow field and (2) updating the position of the interfacial line. The liquid is assumed as pure water. Navier-Stokes equations are solved by using CFD-ACE commercial software (CFD Research Corporation), which is the pressure-based finite volume method. In the pressure-based method, the pressure-density-velocity coupling is handled by a Poisson-type pressure or a pressure correction equation derived from the continuity equation, momentum equation, and equation of state [18]. For simplicity purposes, the movement of the meniscus is simulated in a 2-dimensional geometry.

2.2 Molecular dynamic simulation for liquid flow in nano-scale channel

The MD method is suitable for simulating the liquid flow in a very small geometry, with dimensions on the order of 100 nm and time intervals of a few nano seconds. In order to describe the interaction between atoms or molecules, a potential energy function has to be specified. In this study, the intermolecular interactions are described by the well-known Lennard-Jones (LJ) 12-6 potential models for simple fluids:

$$\Phi(r_{ij}) = 4\epsilon \left[\left(\frac{\sigma}{r_{ij}} \right)^{12} - \left(\frac{\sigma}{r_{ij}} \right)^6 \right] \quad (5)$$

where r_{ij} is the distance between two interacting

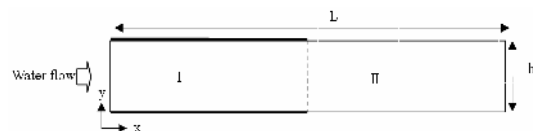


Fig. 1. Schematic of the nozzle with $L = 1\text{mm}$ and $h = 100\ \mu\text{m}$. Regions I and II have different wettability at the walls.

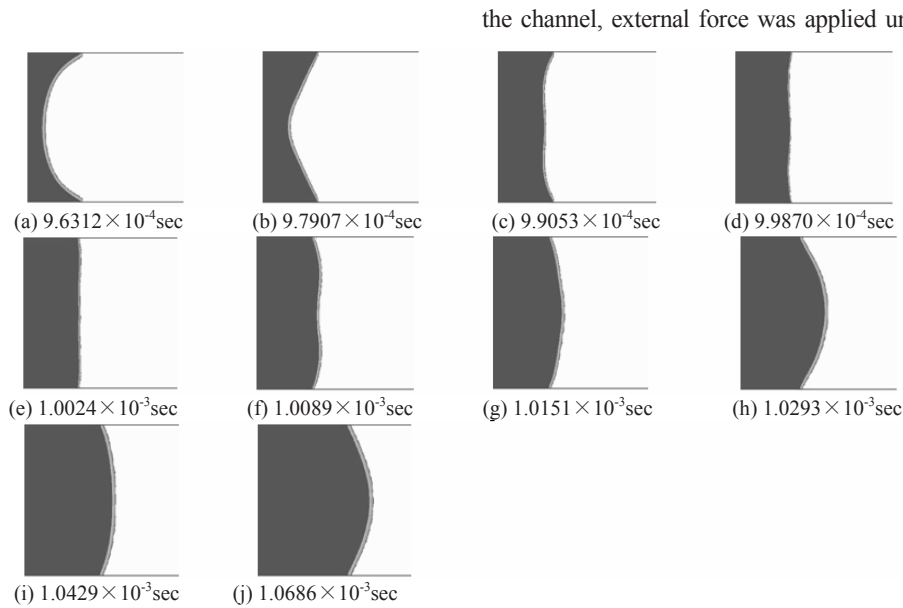


Fig. 2. Sequential transition of meniscus shape across the interface between hydrophilic and hydrophobic surfaces.

molecules, i and j . Here, ε and σ are characteristic energy and length scales, which are selected based on the polarizability and van der Waals radius of the simulated atoms. The first term in Eq. (5) represents a short-range repulsive force, which prevents overlapping of the atoms, while the second term represents an attractive polarization interaction. For liquid-liquid interactions and solid-solid interactions, respectively, the characteristic energy (ε_l) and length scales (σ_l), and characteristic energy (ε_s) and length scales (σ_s) are specified. The potentials for liquid-solid molecules interactions are also represented by the LJ potential function with σ_{sl} and ε_{sl} being given by $\sigma_{sl} = (\sigma_l + \sigma_s)/2$ and $\varepsilon_{sl} = \sqrt{\varepsilon_l \varepsilon_s}$ based on Lorentz-Berthelot combining rule [11].

Molecules move according to the imposed intermolecular forces based on the potential function, and these movements can be described by the classical momentum equations.

$$m \frac{d^2 r_i}{dt^2} = F_i = - \sum_{j \neq i} \frac{\partial \Phi(r_{ij})}{\partial r_i} + f_i \quad (6)$$

Where r_i is the position vector of i th molecule from the coordinate origin. The non-equilibrium MD simulations are performed by using the well-known GROMACS software [19]. Here f_i represents the arbitrary external force. To derive the liquid flow in

the channel, external force was applied uniformly to

each liquid molecule, similar to that of a gravity-driven flow [11, 20].

3. Results and discussions

3.1 Flows in micro-scale channel

The microchannel, as shown in Fig. 1, is assumed to have 2-dimensional geometry, with the size of $100 \mu\text{m} \times 1 \text{mm}$, with an interfacial line in the middle of the channel. Half of the channel wall surface, as shown in Fig. 1, is hydrophilic so that the concave meniscus is entering from the inlet, and the other half is hydrophobic. The spatial grid system used is $(N_x \times N_y) = (400 \times 60)$. The liquid is considered as water and the surface wettability is specified by the contact angle of the liquid meniscus. The contact angles on the hydrophilic and hydrophobic surfaces are assumed to be 30 and 120 degrees, respectively. The liquid is assumed to be supplied into the microchannel with uniform velocity through the inlet. Initially, only air exists in the microchannel with zero velocity. Once the liquid enters the microchannel, the meniscus shape of liquid is traced by the volume fraction of liquid in each grid cell. If the volume fraction in any grid cell has a value of one, it is considered to be liquid.

Fig. 2 shows the transition of liquid-free surface as

the meniscus moves across the interfacial line between the hydrophilic and the hydrophobic surface regions. In Fig. 2(a), the concave meniscus of the liquid is fully developed and moved through hydrophilic micro-channel. In Fig. 5(b)-(j), the transitions of the meniscus shape at the interfacial line are illustrated. The concave meniscus in the hydrophilic region should be changed into the convex shape in the hydrophobic surface. The details of the transition process can be found from these results. When the tip of the concave meniscus arrives at the interfacial line, the fluid near the wall stops for a while until the concave shape disappears. Since momentum is conserved, the lost momentum near the wall transfers to the center region. As a result, the liquid at the center region is accelerated as it crosses the interfacial line. Therefore,

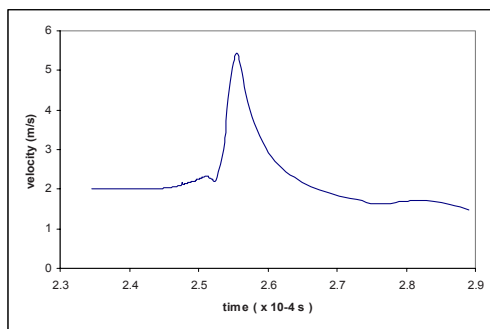


Fig. 3. Variations of the meniscus velocity at center point in the micro-channel.

the transition of meniscus shape from concave to convex can augment the momentum near the center of the channel.

Fig. 3 presents variations of the velocity at the center point of the moving liquid meniscus, which shows the momentum increases up to twice its original value in the upstream during the transition time from concave to convex shape. Therefore, if we use this momentum augmentation at the center region based on the surface wettability characteristics, we can passively control and handle the fluid flow in the micro-channel. Typically, we can apply this type of nozzle structure to the inkjet-head or spray system.

In contrast to the case in Fig. 2, Fig. 4 shows the transition of the meniscus shape in the case of hydrophobic and hydrophilic surfaces arrangement. When the meniscus reaches the interfacial line, the velocity near the center begins to decrease and the meniscus slip velocity on the wall increases.

3.2 Flows in nano-scale channel

To investigate the effects of the interfacial line between hydrophilic and hydrophobic regions in a nano-scale nozzle on flow instability as the liquid meniscus moves across the interfacial line, molecular dynamics simulations are performed by moving argon liquid molecules through a nano-scale nozzle, which is bounded by two parallel solid walls, as shown in Fig. 5. The solid surface consists of three layers of plati-

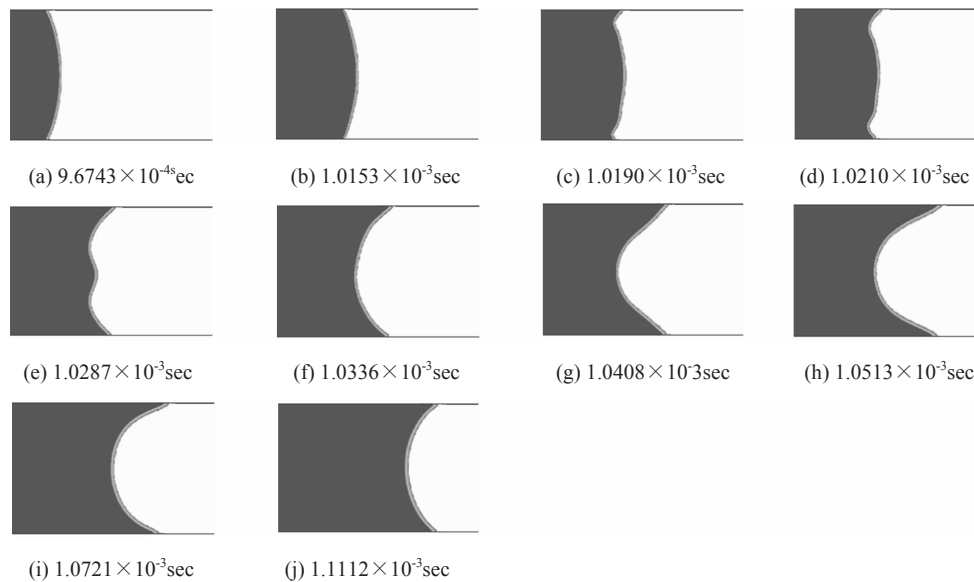


Fig. 4. Sequential transition of meniscus shape across the interface between hydrophilic and hydrophobic surfaces.

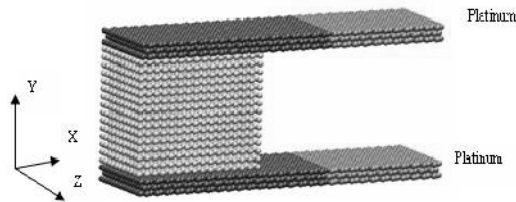


Fig. 5. Schematic of nano scale nozzle and initial state of argon liquid.

num atoms whose lattice is FCC lattice and $\langle 111 \rangle$, and whose surface is in contact with the liquid argon atoms. The simulation domain has a size of $13.94 \times 6.96 \times 3.88 \text{ nm}^3$ and the distance between these two walls is 5.41 nm. There are 5222 platinum molecules and 2640 argon molecules inside the nozzle and the initial temperature of the system and the density of the argon are assumed as 85 K and saturated liquid density. The left region of the platinum wall is assumed as the hydrophilic surface and the right region is assumed as the hydrophobic surface. Because the molecular dynamic simulation should be done in a 3-dimensional geometry, the model is contrary to the 2-dimensional microchannel mentioned in the previous section.

For the liquid-liquid interaction, the LJ potential is applied to argon with the characteristic length scale $\sigma_l = 3.405 \text{ \AA}$ and the energy scale $\epsilon_l = 1.67 \times 10^{-21} \text{ J}$. The LJ potential is also applied for the solid-solid interaction with $\sigma_s = 2.475 \text{ \AA}$ and $\epsilon_s = 1.67 \times 10^{-21} \text{ J}$. In every simulation, the time step used is 1 femtosecond. Periodic boundary conditions with respect to the number of molecules are applied along the x- and z-directions. In the y-direction, on the outside of each solid wall of the simulation cell, three layers of phantom solid molecules are anchored by their lattice are located to model a semi-infinite potential field of the solid wall [11]. External forces are applied uniformly to the argon atoms to initiate flow along the x direction, similar to that of a gravity-driven flow [20].

When considering the free surface, in order to simulate the liquid flow in the nano-channel, we have to specify the contact angle of the liquid argon meniscus on the platinum surfaces. To define the contact angle between liquid argon and the platinum wall, the change of contact angles is investigated by varying the characteristic scales, such as characteristic length and the energy scale [21-23]. The LJ potential in Eq. (5) may also be written in the following form:

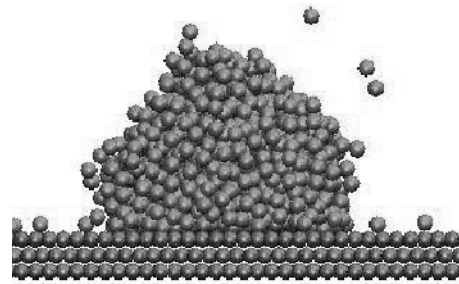


Fig. 6. Snapshot of a liquid droplet on the solid surface.

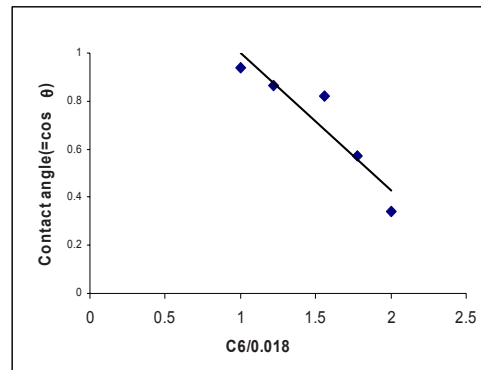


Fig. 7. Contact angles of the liquid argon droplet on the platinum surface according to the potential scale, C6.

$$\Phi(r_{ij}) = \frac{C12}{r_{ij}^{12}} - \frac{C6}{r_{ij}^6} \tag{7}$$

Where C12 and C6 are same as $4\epsilon\sigma^{12}$ and $4\epsilon\sigma^6$ in Eq. (7), respectively.

In order to specify the contact angle on the hydrophilic or hydrophobic surface, the potential scales of C12 and C6 should be determined [11]. The contact angles, between argon liquid droplet and the platinum surface, are examined regarding the variation of C6. The surface is modeled by three layers of a rigid $140 \times 139 \text{ \AA}$ platinum $\langle 111 \rangle$ sheet fixed in a parallel staggered configuration, which consists of 9018 platinum molecules, as shown in Fig. 6. The argon droplet is located on the platinum surface and then the contact angle is measured by assuming a line from a 3-phase contact point, which connects the three or four liquid molecules. Even if the result reveals little information of the value of contact angle on the solid surface, we can analyze the change of wettability according to the variation of C6.

Fig. 7 shows the variation of the contact angle of a liquid droplet according to the C6 potential scale,

which shows how well the results agree with the results by Maruyama et al. [21]. The cosine of the contact angle, of the liquid argon droplet, inversely depends on the strength of the surface potential. For the macroscopic contact angle, the well known Young's equation relates the contact angle to the balance of surface energies such as:

$$\cos \theta = \frac{\gamma_{SG} - \gamma_{SL}}{\gamma_{LG}} \quad (8)$$

Where γ_{SG} , γ_{SL} and γ_{LG} are surface energies between solid-gas, solid-liquid, and liquid-gas. When we change the energy parameter between liquid and solid molecules, γ_{LG} is kept constant and γ_{SG} is negligible compared to γ_{SL} , because the vapor density is much lower than the liquid density. With the increase of $C6$, γ_{SL} should also increase proportionally because of the gain in potential energy in the liquid-solid contact line. Then $(\gamma_{SG} - \gamma_{SL})$ should decrease and Young's equation predicts the decrease in $\cos \theta$. Hence, the linear relation in Fig. 7 can be explained by the analogy in Young's equation. Because $C6$ can describe the interaction between the liquid argon molecule and the solid platinum molecule, we can specify the wettability on the wall by changing the values of $C6$. However, we cannot define $C6$ as

the physical value to represent the real wettability on the hydrophobic or hydrophilic surfaces. Therefore, it is a limitation of this study and may require further investigation. In this paper, we observe just the meniscus change with respect to changes of the wettability on the wall in the nano-channel assuming an arbitrary contact angle.

Based on the variation of contact angle, we can also describe the nano-scale nozzle structure, which is characterized by the interfacial line between the hydrophobic surface and the hydrophilic surface. In the hydrophilic region, we use 0.005 as $C6$ for the interaction between liquid argon molecules and solid platinum molecules, while in the hydrophobic region, we use the value 0.018 as $C6$ to describe the hydrophobic contact angle. Under the initial conditions, the liquid starts moving from left to right due to the external force applied uniformly to each liquid argon molecule.

The liquid meniscus passes across the interfacial line between the hydrophilic and hydrophobic regions within approximately 300 picoseconds. Fig. 8 depicts the sequential movement of the liquid meniscus crossing the interfacial line, and it also shows the meniscus shape's transition from concave to convex. In Fig. 8(a), the concave meniscus shape of the liquid flow is maintained before arriving at the interfacial

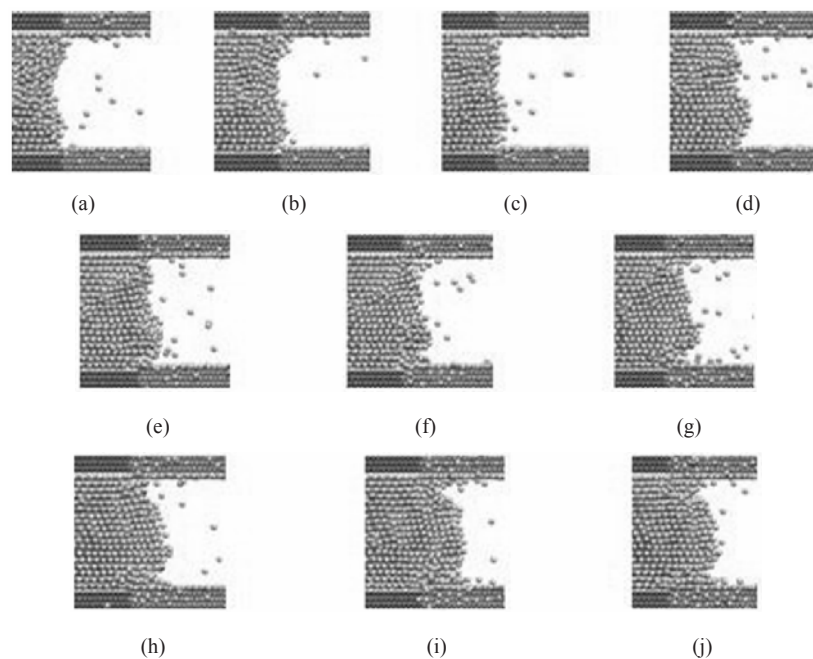


Fig. 8. Sequential movement of meniscus across the interface in nano scale nozzle.

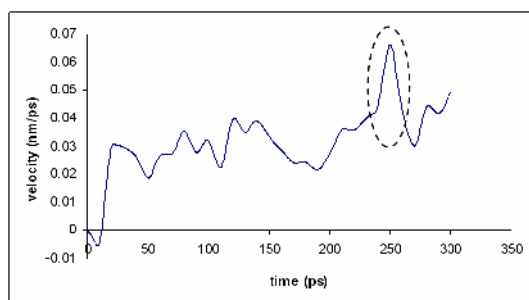


Fig. 9. Variation of velocity in the vicinity of the meniscus center.

line. In Figs. 8(b)-(j), the process of the transition of the meniscus shape at the interfacial line is illustrated. This change in meniscus shape is due to the hydrophobic, which is similar to the observation in the microscale channel. The concave meniscus in the hydrophilic region changes into a convex shape. When the meniscus arrives at the interfacial line, the fluid near the wall ceases moving for a period of time until the concave shape disappears. Because momentum is conserved, the lost momentum near the wall transfers to the center region, and thus the liquid at the center region is accelerated as it crosses the interfacial line.

Fig. 9 presents the velocity profile of the liquid meniscus along the longitudinal direction (x-direction). The profile is obtained by selecting 100 argon molecules in the vicinity of the center region near the meniscus, and then taking an average velocity. Velocity fluctuation exists since the atoms always move with different velocities due to interactions with neighboring atoms. During the transition of the meniscus shape from concave to convex, velocities near the center region suddenly increase almost by two-fold before arriving at the interfacial line. Therefore, by manipulating the surface characteristics, we can passively control and easily handle the fluid flow even in a nano-scale nozzle.

4. Conclusions

A novel structure of the hydrophilic and hydrophobic surfaces is proposed to passively control the liquid flow and efficiently produce droplets for a micro/nano scale jetting system. This is done by using the sudden augmentation of the momentum in the vicinity of the center region of the meniscus. With the computational fluid dynamics (CFD) method and the non-equilibrium molecular dynamics (MD) method in

micro- and nano-scale channels, we investigate the effects of the interfacial line on flow instability, meniscus transition, and momentum augmentation as the liquid meniscus moves across the interfacial line. When the liquid meniscus arrives at the interfacial line between hydrophilic and hydrophobic surfaces, a transition of the meniscus shape occurs from concave to convex, and the fluid motion near the wall stops until the concave shape is fully converted. Because momentum is conserved, the lost momentum near the wall transfers to the center region and, as a result, the liquid at the center region is accelerated more as it crosses the interfacial line.

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Doyoung Byun received the B.S., M.S, and Ph.D. degrees in school of mechanical and aerospace engineering from the Korea Advanced Institute of Science and Technology (KAIST), Taejon, Korea, in 1994, 1996, and 2000, respectively. From 2000 to 2002, he was in the Korea Institute of Science and Technology Evaluation and Planning as a Senior Researcher. In 2003, he joined the faculty of the School of Mechanical and Aerospace Engineering, Konkuk University, Seoul, Korea. His current research topics are development of electrohydrodynamic inkjet head, microfluidic devices, and biomimetic robot systems. His research interests include microfluidics, MEMS, and biomimetics.