Capillary Filling in Nanochannels – Modeling, Fabrication, and Experiments

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ABSTRACT

While capillary filling in channels of micrometers scale is experimentally verified to obey Washburn's law well, the speed of capillary filling in nanochannels is noticeable lower than described by Washburn's formula. This article reports the theoretical and experimental results on capillary filling in open-end and closed-end nanochannels. Nanochannels of 45 nm and 80 nm depth, 10 μm width, were etched in silicon and bonded to a glass cover. Experiments on filling of non-electrolytic liquid in silicon nanochannels were carried out. The filling processes were observed and recorded. To estimate the influence of electrokinetics, a mathematical model to calculate the electroviscous effect was established. This model shows that contribution of electroviscous effect in the reduction of filling speed is small. This result also agrees well with previous theoretical work on the electroviscous effect. That means besides the electroviscous effect, there are other phenomena that contribute to the reduction of capillary filling speed in a nanochannel, such as air bubbles formation. Experimental investigation of capillary filling in open-end and closed-end nanochannels with different lengths was performed. The filling processes of ethanol and isopropanol and the behavior of the trapped air were recorded and evaluated. Analytical models based on the continuum assumption were used to evaluate the experimental data. We observed that the filling process consists of two stages. At the initial stage, experimental data agree well with the theoretical model, but with a higher apparent viscosity. In the final stage, condensation of the liquid phase and dissolution of the gas phase lead to total filling of the nanochannel. The observed phenomena are important for understanding the behavior of multiphase systems in nanochannels.

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INTRODUCTION

The increasing use of semiconductor devices and the increasing spatial density of these devices lead to the demand for new technologies for heat exchangers [1]. As air-cooling technology may soon become insufficient, liquid cooling promises to replace air-cooling technology in the near future [2]. The higher heat transfer rate is based on the fact that heat conductivity of liquid is generally higher than that of air. For instance, the heat conductivity of water of 0.58 W/m-K is more than 20 times of that of air (0.024 W/m-K). Thermal properties of liquid flow in microchannels and millichannels have been studied intensively in the past [3]. Weisberg et al. [4] numerically investigated a heat exchanger using microchannels. The authors also suggested a formulation to design such a heat exchanger. Peng et al. performed a series of works [5-8] on liquid flow and heat transfer in microchannels. Similar investigations of fluid flow and heat transfer in microchannels and their applications were discussed by other authors [9-14].

As the characteristic size of the channel gets smaller, the ratio between surface area and volume increases. Therefore, microscale phenomena are more favorable for heat transfer [15]. However, studies on heat transfer in nanochannels with characteristic dimensions ranging from several to hundreds of nanometers are still at an early stage. One of the difficulties in investigation of heat transfer in at the nanoscale is that the transport phenomena of fluid flow in nanostructure have not been fully understood.

In recent years, advances in micro/nanotechnologies allowed the fabrication of structures at nanoscale [16-21]. Various techniques and devices for transport, handling, and manipulating fluid in micro-/nanoscale were developed [22-24]. These technological advances allow more sophisticated investigation of transport phenomena in nanoscale. Many experiments on capillary filling in nanochannels have been carried out in recent years [25-30]. The experimental results revealed that capillary filling in nanochannel qualitatively follows Washburn's equation. However, the filling speed observed experimentally is usually lower than expected by the classic Washburn's formula, which is equivalent to an increase in apparent viscosity.

Several efforts were reported to explain the variation between experimental and theoretical results of capillary filling in nanochannels. Van Honschoten et al. [31] suggested a model of elastocapillary filling in deformable channels. The deformation of nanochannels due to high negative pressure across the meniscus may cause variation in apparent viscosity. Because of the high surface-to-volume ratio of the nanochannels, it is believed that surface effects such as electroviscous effect contribute to the increase in apparent viscosity [32-35]. Han et al. [26] investigated the filling kinetics of different liquids in nanochannels with rectangular cross section. Trapping of air bubbles during the filling processes was observed.

The formation of air bubbles depends on the interaction between gas and liquid in nanoscale. However, most of the knowledge gained in the past was based on macroscopic investigation of porous materials. For instance, Shaw reported

experimental results of the evaporation process in porous materials [36]. This work set the foundation for subsequent research on modeling of drying processes in porous materials. Prat used the invasion percolation theory to model the drying process in porous material [37]. Yiotis et al. presented a theoretical model for the drying process in porous materials [38]. The results showed that the formation of a liquid film accelerates the drying process.

In this article, we report theoretical and experimental results on capillary filling in open-end and closed-end nanochannels. Capillary filling experiment in the open-end nanochannel were performed with non-electrolytic liquids (ethanol and isopropanol) to minimize possible electroviscous effects. A simple mathematical model to evaluate the contribution of electroviscous effect in the increase of apparent viscosity is also introduced. The results from the theoretical model are then compared to the experimental value above to estimate the general significance of electroviscous effect in the overall increase of apparent viscosity. Closed-end nanochannels offer an ideal platform for studying multiphase-related phenomena at the nanoscale, such as condensation. In our work, quantitative measurements of filling lengths versus time were obtained for isopropanol and ethanol. We also investigated the behavior of trapped air during the filling process. Under the high Laplace pressure, condensation of the liquid phase followed by dissolution of the gas phase are the main phenomena observed in the nanoscale confinement.

THEORY

Capillary filling in open-end and closed-end planar nanochannels with a height of h , a width of w , and an aspect ratio of $w/h \gg 1$ is considered. The driving force depends on the surface tension of the fluid and the contact angle between the fluid and the channel wall

$$F_s = 2\sigma w \cos \theta \quad (1)$$

where F_s is the capillary force at the contact line caused by surface tension σ , and θ is the contact angle between the fluid and the wall surface. In this investigation, the variation of surface energy due to accumulation of charged particles near the meniscus, as described in the Gibbs-Duhem equation, is ignored. It was reported in the literature that the liquid slip at the solid-liquid interface [39] and the interaction with electrokinetic effects [40] may lead to modification of effective contact angle. However, in capillary filling of nanochannels the shear stress and streaming voltage diminish quickly. Under such condition the change in effective contact angle is negligible. The velocity profile in capillary filling flow is considered the same as in pressure-driven flow. Due to low Reynolds number, the flow resistance caused by an entrance effect as discussed in recent investigation [41-44] is ignored. In the case of a planar nanochannel, the velocity profile has a parabolic form [45]. The viscous force is then given by

$$F_v = \left(\mu \frac{du}{dy} \Big|_{y=\frac{h}{2}} - \mu \frac{du}{dy} \Big|_{y=-\frac{h}{2}} \right) wx = -\frac{12\mu\bar{u}}{h} wx \quad (2)$$

where F_v is the viscosity force acting on the fluid, x is the filling length, $\bar{u} = dx/dt$ is the filling speed, and μ is the dynamic viscosity of the fluid. The filling process is considered to begin at the entrance of the channel; hence the initial condition $x=0$ at $t=0$ is applied to both closed-end and open-end nanochannels.

Capillary Filling in Open-End Nanochannels

Figure 1 a shows the model of capillary filling process in an open-end nanochannel, with consideration of electroviscous effect. Without any loss of generality, the wall of the channel is assumed to be positively charged, through the selective absorption of cations. The shaded areas in Figure 1 a represent the electrical double layer (EDL). The streaming voltage caused by the accumulation of anion at the meniscus generated a conductive current, as well as the electric force against the filling movement. The reduction of filling speed represents the increase in apparent viscosity [46].

In a nanochannel, the electric potential distribution and the charge density distribution is governed by the Poisson-Boltzmann (PB) equation [47, 48]:

$$\nabla^2 \Psi = -\frac{\rho_q}{\varepsilon_0} = -\frac{e}{\varepsilon_0} \sum_i z_i n_i^0 \exp\left(-\frac{z_i e \Psi}{kT}\right) \quad (3)$$

where Ψ is the electrostatic potential, ρ_q is the charge density, $e = 1.6021 \times 10^{-19} \text{C}$ is the elementary charge, ε is the relative permittivity of the fluid, $\varepsilon_0 = 8.854 \times 10^{-12} \text{CV}^{-1}\text{m}^{-1}$ is the permittivity of vacuum, z_i is the charge number of ionic species i , n_i^0 is the bulk concentration of ionic species i , $k = 1.381 \times 10^{-23} \text{JK}^{-1}$ is Boltzmann's constant, and T is the temperature. Applying Debye-Hückel approximation of low potential for binary electrolyte (i.e. $z_1 = z_2 = z$), the potential distribution is given by [48]

$$\Psi(y) = \zeta \frac{\cosh(-\kappa y)}{\cosh(\kappa h/2)} \quad (4)$$

and the charge density distribution is given by

$$\rho_q = -\varepsilon_0 \frac{d^2 \Psi}{dy^2} = -\varepsilon_0 \zeta \kappa^2 \frac{\cosh(\kappa y)}{\cosh(\kappa h/2)} \quad (5)$$

where κ is the Debye parameter and $\kappa^2 = \frac{2e^2 z^2 n^0}{\varepsilon_0 kT}$ (n^0 is the bulk concentration of the solute). The Debye-Hückel approximation is only valid for ζ potential below 26 mV [49]. Studies in reported literature describe the solution of the PB equations in

other conditions [32, 34, 35, 50-59]. These results can be applied in our model by substituting the Eq. (4) accordingly. In such cases, the final equations should be more difficult to be solved.

Considering the redistribution of charge density, the average conductivity of the monovalent electrolyte, such as NaCl solution, in nanochannels is given by

$$\bar{\lambda} = n^0 \frac{\Lambda_m}{N_A} \left(1 + \frac{\zeta e}{kT} \frac{v_- - v_+}{v_+ + v_-} \frac{2 \tanh\left(\frac{\kappa h}{2}\right)}{\kappa h} \right) \quad (6)$$

where v_i is the electrical mobility of ionic species i and $\Lambda_m = \sum_i e N_A v_i$ is the molar conductivity of a monovalent electrolyte.

The charge balance is considered with streaming current, conductive current and accumulated charge. The charge balance is then expressed by

$$-\frac{q_a \bar{\lambda}}{\varepsilon_0} + \int_{-\frac{h}{2}}^{\frac{h}{2}} u \rho_q w dy = \frac{d}{dt} q_a \quad (7)$$

The electric charge accumulated at the front of the fluid column produces an electric field, namely the streaming potential field. Such an electric field in turn causes electrostatic force acting on the entire liquid column. Assuming a constant electric field along the channel, the electric force is

$$F_e = -x \int_{A_c} \frac{U_s}{x} \rho_q dA_c = -xw \int_{-\frac{h}{2}}^{\frac{h}{2}} \frac{U_s}{x} \rho_q dy \quad (8)$$

where U_s is the streaming potential, which is the electric potential difference between the meniscus and the entrance of the channel.

The force balance on the moving liquid column is expressed according to Newton's second law [60]

$$F_s + F_v + F_e = \frac{d}{dt} (\rho_m w h x \bar{u}) \quad (9)$$

where F_e is the electric force and ρ_m is the mass density of the fluid.

From the force balance and the charge balance, the equation system for the capillary filling effect in a nanochannel is expressed as

$$\left\{ \begin{aligned} 2\sigma w \cos \theta - \frac{12\mu\bar{u}}{h} wx - \frac{q_a x}{h\mathcal{E}_0} \int_{-\frac{h}{2}}^{\frac{h}{2}} \rho_q dy &= \frac{d}{dt}(\rho_m whx\bar{u}) \\ -\frac{q_a \bar{\lambda}}{\mathcal{E}_0} + \bar{u}w \int_{-\frac{h}{2}}^{\frac{h}{2}} \left(\frac{3}{2} - 6\frac{y^2}{h^2} \right) \rho_q dy &= \frac{d}{dt} q_a \end{aligned} \right. \quad (10)$$

The system of nonlinear ordinary differential equations (10) cannot be solved analytically. However, an asymptotic solution can be found. The asymptotic solution is expressed in the form similar to Washburn's formula, with apparent viscosity μ_a ,

$$x = \left[\frac{\sigma \cos \theta h}{3\mu_a} t \right]^{1/2} \quad (11)$$

where the relative viscosity change is defined as

$$\frac{\Delta\mu}{\mu} = \frac{\mu_a - \mu}{\mu} = \eta \frac{\tanh\left(\frac{\kappa h}{2}\right) \left[\kappa h - 2 \tanh\left(\frac{\kappa h}{2}\right) \right]}{\left[1 + \frac{\zeta e}{kT} \frac{v_- - v_+}{v_- + v_+} \frac{2 \tanh(\kappa h/2)}{\kappa h} \right] (\kappa h)^2} \times 100\% \quad (12)$$

where $\eta = \frac{4\mathcal{E}_0 N_A \zeta^2 e^2}{\mu \Lambda_m kT}$ is a dimensionless parameter.

With solutions in which two species of ions have comparable diffusivities (i.e. NaCl), Eq. (12) can be further simplified to

$$\frac{\Delta\mu}{\mu} = \eta \frac{\tanh\left(\frac{\kappa h}{2}\right) \left[\kappa h - 2 \tanh\left(\frac{\kappa h}{2}\right) \right]}{(\kappa h)^2} \times 100\% \quad (13)$$

Figure 2 presents the relative viscosity change predicted by Eq. (13), in comparison to the values reported by Mortensen and Kristensen [33], under the same condition.

Capillary Filling in Closed-End Nanochannels

Figure 1 b illustrates the capillary filling process in a closed-end nanochannel. Both trapped air and filling liquid are considered as continua. Condensation of the liquid phase and dissolution of the gas phase are neglected. The forces governing the filling process of a closed-end nanochannel are the capillary force F_s , the viscous force F_v , and the resistance force of the trapped and compressed air,

$$F_p = -hwp_0x/(L-x) \quad (14)$$

where L is the total length of the nanochannel and p_0 is the initial pressure of the trapped air. The electroviscous effect is not considered in this model. Since inertia force is negligible in nanoscale, the force balance $F_s + F_v + F_p = 0$ leads to

$$\frac{dx}{dt} = \frac{h\sigma \cos \theta}{6\mu x} - \frac{h^2 p_0}{12\mu(L-x)} \quad (15)$$

The parameters of Eq. (15) are made dimensionless as $x^* = x/L$, $t^* = t/2\tau$ and $\alpha = hp_0/2\sigma \cos \theta$, where α represents the ratio of force due to the trapped air pressure over the capillary force, and $\tau = 3\mu L^2/h\sigma \cos \theta$ is the required time to fill an open-end channel of the same length L . The governing equation (15) has then the dimensionless form

$$\frac{dx^*}{dt^*} = \frac{1}{x^*} - \frac{\alpha}{1-x^*} \quad (16)$$

Integrating the preceding equation with the initial condition $t^*(0) = 0$ for any $x_0^* < 1/(1+\alpha)$ leads to the relationship:

$$t^*(x^*) = \frac{x^{*2}}{2(1+\alpha)} - \frac{\alpha x^*}{(1+\alpha)^2} - \frac{\alpha}{(1+\alpha)^3} \ln[1 - (1+\alpha)x^*] \quad (17)$$

The final position of the liquid column can be derived from Eq. (17) as:

$$\lim_{t^* \rightarrow \infty} x^* = \frac{1}{1+\alpha} \quad (18)$$

If condensation and dissolution are neglected, the filling length x of the liquid cannot exceed $L/(1+\alpha)$. At this final length, the magnitude of the resistance force of the trapped air is equal to that of the capillary force, $F_s + F_p = 0$. For the case of 45-nm-height nanochannels, the Laplace pressure caused by ethanol is approximately 9.96 bars.

EXPERIMENTS

Fabrication and Experimental Setup

Figure 3 depicts the schematic designs of the open-end and closed-end nanochannels under investigation. Nanochannels of 10 μm width and 45 nm and 80 nm depth were fabricated in silicon by reactive ion etching (RIE). The length of closed-end nanochannels ranges from 10 μm to 8 mm. For each channel dimension, a set of 10 channels were designed for obtaining good statistics in later experiments. Microchannels with a cross section of 8 $\mu\text{m} \times 100 \mu\text{m}$, which connected to the nanochannels, were realized by deep reactive ion etching (DRIE). Both ends of the microchannels are accessible through 1-mm-diameter reservoirs etched through the silicon wafer using a long DRIE process. Lastly, a Pyrex glass plate was anodically

bonded to the silicon wafer to seal the nanochannels. Bonding was performed at 350 °C and 400 V. Figure 4 a shows the fabricated channel network consisting of the access hole, the microchannel and the nanochannels. A typical filling process of open-end nanochannels is shown in Figure 4b.

In our experiments, nanochannels with reservoirs facing up were observed and recorded with an inverted microscope (Zeiss Observer D1) and a camera (EMCCD AndorIQ, Andor Technology PLC, Northern Ireland). The liquid (ethanol or isopropanol) was introduced into one of the reservoirs and filled the microchannel and nanochannels by capillary force. To keep ethanol from wetting the outer surface of the chip, a spacer made of polydimethylsiloxane (PDMS) was placed above the reservoir. The data were recorded as 16-bit gray-scale multi-images uncompressed tagged image files (TIF) with a size of 1004 pixels×1002 pixels. The temperature and relative humidity were measured before each experiment. The surface tension and viscosity were derived from the temperature, using a formula provided by Yaws [61]. The environmental temperature during the experiments for both cases of with ethanol and isopropanol is measured as 26 ± 1 °C. The relative humidity of the environment was 48%. According to the formula provided by [61], the relative variation in surface tension and viscosity is less than 1%, which is considered to be negligible. Macroscopic contact angle measurement revealed that both liquids under investigation totally wet silicon and glass ($\theta = 0^\circ$).

In the experiment with closed-end nanochannels, the filling processes were observed with different magnifications and time scales. The experiment consists of two steps. In the first step, we recorded the filling process from the entrance of the nanochannels until the end of the 3-mm-long channel set. Objective lens with a magnification of $2.5\times$ was used for this purpose. The field of view was approximately 3 mm × 3 mm. The exposure time was 10 ms. Images were recorded with a frame rate of 17 fps. In the second step, one of the closed ends of the nanochannels was chosen to investigate the behavior of the advancing meniscus and of the trapped air. A magnification of $20\times$ and a frame rate of 5 fps were used for this experiment. The images were recorded to observe the slow process of condensation and dissolution until the nanochannels were fully filled.

Experiment Result

The square of filling distance versus time characteristics, as shown in Figure 5, reveals that capillary filling in open-end nanochannels qualitatively follows Washburn's formula

$$x = \left[\frac{\sigma \cos \theta h}{3\mu} t \right]^{1/2} \quad (19)$$

However, quantitative analysis shows that the Washburn coefficient,

$$a = \sqrt{\frac{\sigma \cos \theta h}{3\mu}} \quad (20)$$

is lower than predicted by the classical Washburn's theory, which is equivalent to an increase in apparent viscosity. Table 1 lists the measured relative viscosity change of ethanol and isopropanol. It is reported in recent investigations that electroviscous effect is the cause of reduction of speed in pressure-driven flow and even capillary filling [32-35]. However, as derived from Eq. (13), which indicates that the dimensionless parameter η is on the order of unity for most of electrolytic solutions (i.e., $\eta \approx 0.5$ for NaCl 0.01M solution), the maximum increase in apparent viscosity is not more than 5%. In our capillary filling experiment, non-electrolytic liquids that exhibit negligible electroviscous effect also encounter a relatively high viscosity increase ratio, as shown in Table 1. Therefore, there are other phenomena besides the electroviscous effect that cause the increase in apparent viscosity of liquid in nanochannels. There are reports on the formation of air bubbles during the filling process [8-11]. A fraction of energy is stored in term of surface energy of the air bubbles. Hence, less energy transfers into kinetic energy of the fluid column, leading to the reduction in filling speed. Additionally, the presence of bubbles modifies the flow profile and may lead to an increase in viscous dissipation, and thus to a slower filling.

Figure 6a shows the measured filling length of isopropanol in closed-end nanochannels with different lengths. The graph shows an identical filling pattern for all nanochannels with different lengths in the first stage. When the meniscus of the liquid approaches the end of the channel, the filling process begins to slow down. Figure 6b depicts the normalized filling length as a function of normalized time in comparison to the theoretical behavior predicted by Eq. (17) with the bulk value of viscosity. The results show that the filling speed in the first stage is actually lower than predicted by theory. This behavior was also observed in previous works on capillary filling of open-end nanochannels. The phenomenon is described by introducing an apparent viscosity, whose value is higher than the bulk value. Figure 6c presents a comparison to amended theory using modified apparent viscosity, obtained by fitting the first stage of filling to the Washburn's formula. Corresponding results for ethanol are presented in Figure 7.

The results show that the filling process consists of two major stages. In the initial stage, the length of the liquid column follows the theory well, but with a higher apparent viscosity. In this stage, where $t^* \ll \tau$, the pressure force is negligible and the filling process can be approximated by Washburn's law. This condition is used to calculate the experimental relative viscosity change, as presented in

Table 2. The relative viscosity change in closed-end nanochannels is noticeably higher than those in the open-end nanochannel. The difference is more significant for shorter channels. This effect can be explained by the non-ideality of the assumptions used in the mathematical model. In this model, an isothermal condition is used. However, if the dissipation of heat is not fast enough, the compression of air during the filling process may cause heating of air, and hence increase the pressure force reducing the filling speed. In the second stage, the meniscus does not stop as assumed with the theory. The liquid column slowly advances toward the end of the nanochannel and finally fills it completely. We observed that in the second stage, the filling process consists of a combination of condensation of the liquid phase and dissolution of the gas phase.

A larger noise was detected in the second stage of the filling process because of the irregular shape of filled areas. To analyze this second stage, we evaluated the remaining unfilled area of the nanochannel. Images of the end of the nanochannels recorded at a higher magnification of $20\times$ were analyzed. Figure 8 a shows the measured surface area of the unfilled part in a 2-mm-long nanochannel as a function of time. The measured data were filtered by Savitzky-Golay filter of second order to reduce the noises. These two stages can be observed clearly in Figure 8a. In the second stage the amount of unfilled area reduces linearly with time. This linear behavior is confirmed by Figure 8b, which plots the rate of change of the unfilled area as a function of time. The inserts in Figure 8a show that during the first stage, the meniscus of the liquid advances more quickly and the shape of the meniscus is more defined. In the second stage, the filling speed is much slower and the meniscus does not have a defined shape. We observed that liquid islands were formed and air bubbles were trapped in the confined space. While the liquid islands spread, the encircled bubbles reduced their size and gradually disappeared. This phenomenon can be considered as a combination of gas dissolution and liquid condensation and probably is enhanced by film flows and corner flows [25]. The advance movement of the meniscus in the second stage is governed by the dissolution of gas into liquid and the diffusion of the gas molecules out of the nanochannels. Due to the high pressure of the trapped gas, which is approximately equal to the Laplace pressure, the gas molecules dissolve into the liquid phase, and the concentration of gas molecules at the meniscus in the liquid phase is determined by the Henry's law. Due to the difference in gas molecules concentration between the meniscus and the entrance of the nanochannel, the gas molecules diffuse out of the channel. The diffusion rate of the gas molecules determines the advancing rate of the meniscus. A detailed theoretical analysis on this phenomenon is beyond the scope of this article and is considered in our future works. The evaporation and condensation of liquids is properly due to thermal fluctuation. These phenomena lead to the formation of liquid islands and air bubbles, which cause local instability of the meniscus in the second stage. However, because the diffusion process only depends on the global concentration gradient in the nanochannel, local instability caused by evaporation and condensation should not affect the average advancing rate of the meniscus.

CONCLUSIONS

This article reports the theoretical and experimental results on capillary filling in open-end and closed-end nanochannels. The nanochannels of 45 nm and 80 nm depths and 10 μm width were fabricated by a standard plasma etching process in silicon wafer. A Pyrex glass cover was anodically bonded to the silicon wafer to seal the nanochannels. The nanochannels were then filled with non-electrolytic liquid. The filling processes were observed and recorded. A mathematical model to calculate the electroviscous effect was established. This model shows that contribution of electroviscous effect in the reduction of filling speed is small. The result agrees well with previous theoretical works on the electroviscous effect. Consequently, there exists other phenomena involve in the variation in Washburn's coefficient, such as the formation of air bubbles. The capillary filling processes of closed-end nanochannels were also recorded and compared to a simple theoretical model that neglects the condensation of the liquid and the dissolution of gas. The data show clearly two stages of the filling process. In the first stage, the experimental data agree well with the theory, but with a higher apparent viscosity. However, in the second stage, the menisci do not stop at the critical position predicted by the theory, but continue to fill until the end of the nanochannels. A quantitative analysis of this stage shows a linear relationship between the filling area and the time. High-magnification images reveal that in this stage liquid islands form, while gas bubbles trapped in the nanochannels dissolve into the liquid. The air bubbles reduce their size before totally disappearing. This phenomenon suggests that condensation of liquid and dissolution of gas are important processes at the nanoscale, where the Laplace pressure is relatively high. The observed condensation and dissolution phenomena can affect transport processes of liquid in nanochannels.

NOMENCLATURES

a	Washburn's coefficient
A	Area of trapped air
A_c	Cross-sectional area of the channel
DRIE	Deep reactive ion etching
e	Elementary charge, $1.305 \times 10^{-19} \text{C}$
EDL	Electric double layer
F_e	Electrical force
F_s	Surface force, capillary force
F_v	Viscous drag force
F_p	Pressurized force of trapped air

h	Channel height
k	Boltzmann's constant, $1.381 \times 10^{-23} \text{ JK}^{-1}$
L	Channel length
n	Local concentration
n^0	Bulk concentration
N_A	Avogadro's number, 6.0221415×10^{23}
p_0	Pressure
p_0	Initial (atmospheric) pressure
q_a	Accumulated charge
t	Time variable
t^*	Dimensionless time
T	Room temperature
u	Fluid velocity
U_s	Streaming potential
w	Channel width
x	Capillary filling length
x^*	Dimensionless filling length
x_0^*	Initial dimensionless filling length
y	Coordinate across the channel height
z	Charge number

Greek Symbols

α	Ratio between initial pressure and Laplace pressure
$\Delta\mu$	Increase in dynamic viscosity
ε	Relative permittivity of the fluid
ε_0	Permittivity of free space, $8.854 \times 10^{-12} \text{ CV}^{-1} \text{ m}^{-1}$
ζ	Zeta potential
η	Material dimensionless parameter
θ	Contact angle

κ	Inverse of Debye thickness
λ	Conductivity of the fluid
Λ_m	Molar conductivity of solution
μ	Dynamic viscosity
μ_a	Apparent dynamic viscosity
σ	Surface tension
ρ_m	Mass density of the fluid
ρ_q	Charge density
τ	Characteristic filling time
ν	Ion mobility
Ψ	Electrostatic potential across the channel's height

Subscripts

0	initial condition
+	positive ion
-	negative ion
a	apparent, accumulated
c	cross section
E	electric
m	molar, mass
p	pressurized
q	charge
s	surface
S	streaming
v	viscous

Superscripts

—	Average value across the height
^	Divide by channel width w
i	Belong to ion species i

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Table 1 Relative viscosity change of different fluids and channel geometries.

Fluid	Channel height (nm)	Relative viscosity change (%)	
		Mean	Std dev
Isopropanol	45	28.0	6.10
Isopropanol	80	9.70	1.00
Ethanol	45	21.5	2.00
Ethanol	80	9.30	3.60

Table 2 Ratio between the increase in viscosity to standard value for different fluids and channel geometries during the first stage of filling in closed-end nanochannel (45nm in height).

Fluid	Channel length (mm)	Relative viscosity change (%)
Isopropanol	0.5	140
Isopropanol	1	136
Isopropanol	1.5	92.0
Isopropanol	2	81.0
Isopropanol	2.5	77.0
Ethanol	0.5	149
Ethanol	1	115
Ethanol	1.5	108
Ethanol	2	108
Ethanol	2.5	103
Ethanol	3	95.0

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