Supporting Information for

On Interfacial Viscosity in Nanochannels

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S1. Applying Boundary Condition for the Governing Equations of Capillary Flow

Applying Navier-Stokes equation to both bulk liquid and the interfacial layer yields

$$\mu_1 \frac{\partial^2 u_1}{\partial z^2} - \frac{dp}{dx} = 0 \tag{1a}$$

$$\mu_2 \frac{\partial^2 u_2}{\partial z^2} - \frac{dp}{dx} = 0 \tag{1b}$$

where subscripts 1 and 2 represent the bulk and the interfacial layer properties, respectively. The follwing boundary conditions must be satisfied:

$$\begin{aligned} &\left(\frac{\partial u_1}{\partial z}\right)_{z=0} = 0 \\ &(u_2)_{z=\frac{h}{2}} = 0 \\ &(u_1)_{z=l_s} = (u_2)_{z=l_s} \\ &(\tau_1)_{z=l_s} = (\tau_2)_{z=l_s} \end{aligned} \tag{2}$$

where $z = 0$ marks the middle of the channel and, $z = h/2$ is the location of the nanochannel wall. Also, τ is the shear stress in the liquid. Integrating both sides of Eqs. (1a) and (1b) twice and applying boundary conditions (2), yields the flow velocities in bulk liquid u_1, and within the interfacial layer u_2:

$$u_1 = \frac{1}{\mu_1}\left(\frac{dp}{dx}\right)\frac{z^2}{2} - \frac{1}{\mu_2}\left(\frac{dp}{dx}\right)\frac{h^2}{8} - \left(\frac{dp}{dx}\right)\left(\frac{1}{\mu_1} - \frac{1}{\mu_2}\right)\left(\frac{l_s^2}{2}\right)$$

$$u_2 = \frac{1}{\mu_2}\left(\frac{dp}{dx}\right)\frac{z^2}{2} - \frac{1}{\mu_2}\left(\frac{dp}{dx}\right)\frac{h^2}{8}$$

S2. Nanochannel Fabrication

The fabrication process consists of four main steps including patterning, etching, thermal oxide growing and anodic bonding presented in Figure S1. First, a positive photoresist was uniformly dispersed on a silicon wafer by using a spin coater (Brewer Cee 200) and then backed on a hot plate at 130°C for 90s. In order to create an optical mask, the geometric pattern of nanochannels was designed in SolidWorks (Dassault Systemes) and transferred onto a chrome mask with a resolution of 1 μm. The optical mask was utilized to pattern the photoresist by exposing it to UV irradiation through mask aligner (ABM). Afterward, the sample was immersed into a photoresist developer, washed with deionized water and dried with nitrogen flow. Our structure includes 9 sets of nanochannels bridging two micro-reservoirs. Each set of nanochannels consists of 11 individual nanochannels. The width and length of each nanochannel is 5 μm and 500 μm, respectively. The nanochannels' patterns were etched into the silicon wafer with different depths using controlled reactive ion etching (Oxford Plasma Lab ICP 180 RIE). To remove the photoresist after etching, the sample was washed with acetone, alcohol and deionized water, and then dried with nitrogen flow. The channels height is in the range of 10 nm to 500 nm, which can be defined by the etching recipe of silicon. All the steps of standard photolithography and reactive ion etching were repeated to pattern and etch two micro-reservoirs perpendicularly located at the two ends of nanochannels. The micro-reservoirs are squares with length of 4 mm and height of 20 μm. In the next step, 300 nm oxide was thermally grown on the developed micro-nanochannels. This thermal oxide acts as an insulating layer which decreases the high electro-static forces in nanometer distances between silicon and glass and prevents channel collapse during anodic bonding. The last step of nanofluidic device fabrication is sealing the developed micro-nanochannels with borosilicate glass through anodic bonding process. Initially, two holes were drilled on the glass in order to facilitate the introduction of liquid to the micro-reservoirs. Next, the silicon wafer and borosilicate glass were dipped into piranha solution (H_2SO_4: H_2O_2=3:1) for 15 min. After cleaning, the silicon wafer and borosilicate glass were bonded together by anodic bonding process carried out for 10 min at 400°C with a voltage of 300V.

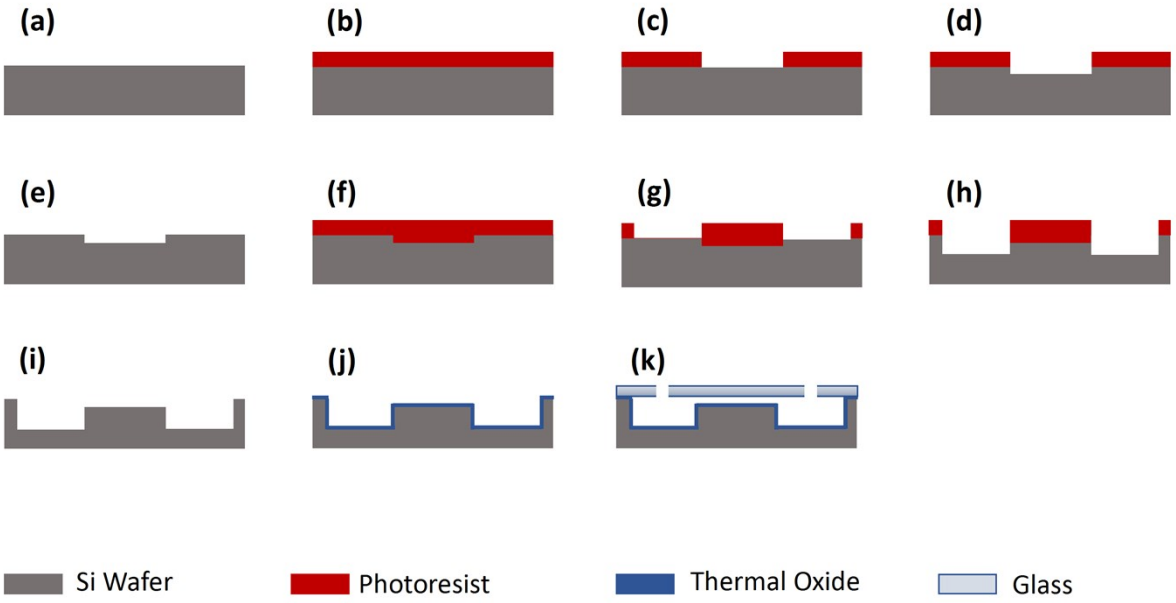

Figure S1. Fabrication process of Nanofluidic device. (a) Silicon wafer, (b) Spin coated photoresist on top of the Silicon wafer, (c) Nanochannels pattern on the positive photoresist through standard photolithography, (d) Etched Nanochannels on the silicon wafer through RIE, (e) Washed sample after etching, (f) Spin coated photoresist for a reservoir fabrication, (g) Micro-reservoirs pattern through standard photolithography, (h) Etched micro-reservoirs on the silicon wafer through RIE, (i) Washed sample after etching, (j) Growth of thermal oxide on the fabricated micro-nanochannels, (k) Sealed micro-nanochannel with glass through anodic bonding.

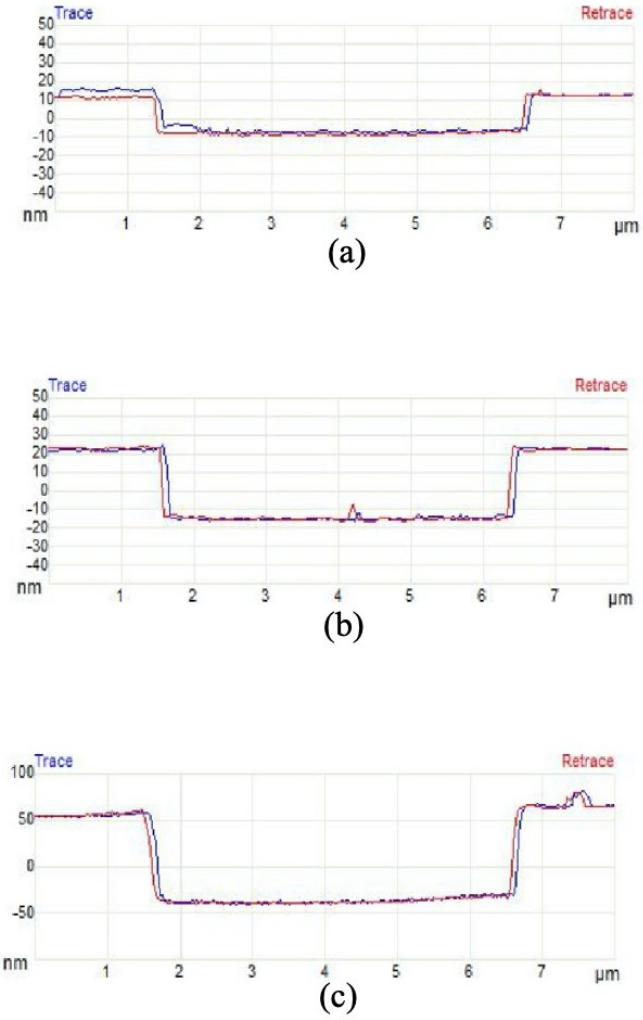

Figure S2. Height profile analysis of nanochannels obtained by Scanning Probe Microscopy. Height uniformity across the nanochannels with heights of (a) 20 nm, (b) 40 nm and (c) 80 nm is confirmed.

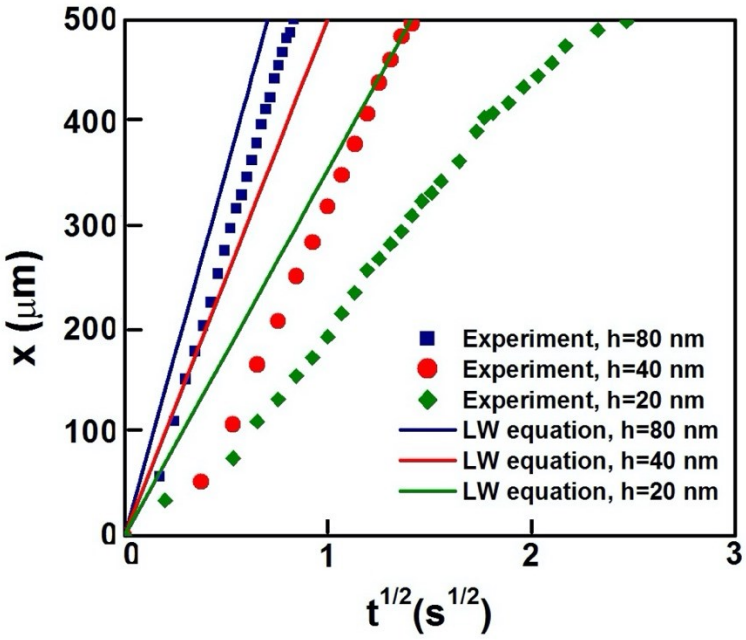

Figure S3. The deviation of the LW model prediction from the experimental observation for imbibition of ethanol in nanochannels with heights of 80 nm, 40 nm, and 20 nm.

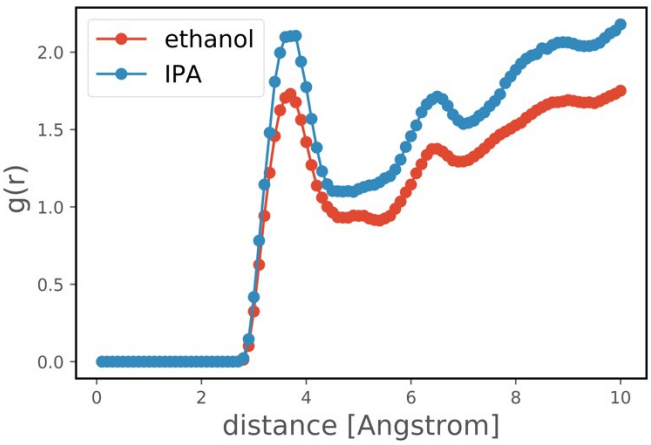

Figure S4. Radial distribution function of the oxygen atoms in the hydroxyl groups of ethanol and IPA molecules with respect to silicon atoms of the wall.

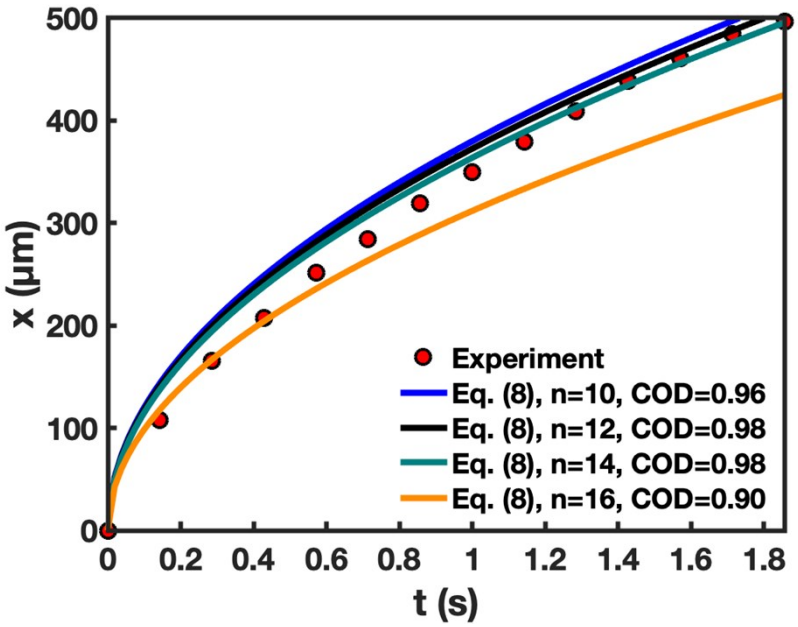

Figure S5. The COD values show that n=12 yields the closest match between our model and the experimental data for imbibition of ethanol in 40 nm channel.

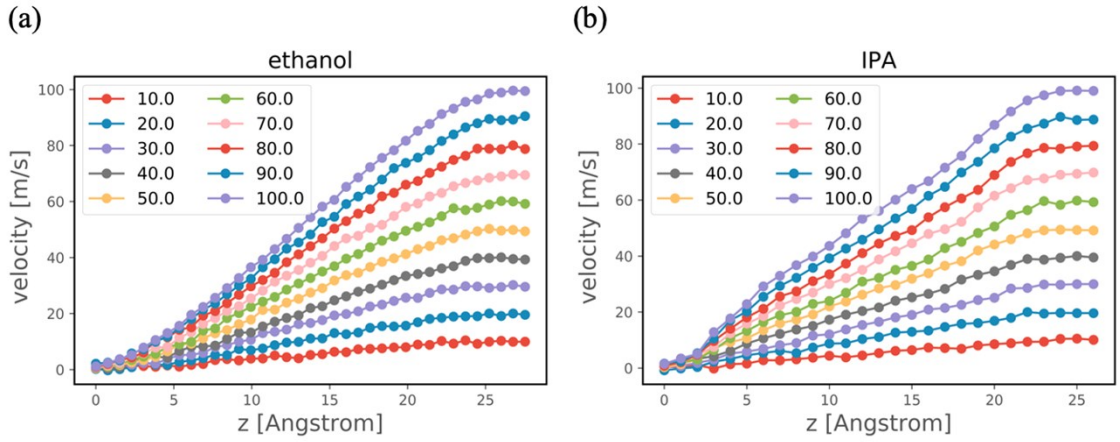

Figure S6. Velocity profiles of (a) ethanol and (b) IPA confined in silicon nanochannels under different shear velocities.