FORMATION OF LIQUID MENISCUS IN FLEXIBLE NANOCHANNELS

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ABSTRACT

This paper describes the elasto-capillary formation of menisci at the liquid-air interface in nanochannels that are covered with flexible capping membranes. The equilibrium between the capillary pressure in the fluid and the membrane bending results in a very peculiar shape of the meniscus. We present an analytical description of these meniscus shapes and show that the protrusion length of the meniscus along the channel is an accurate measure for the deflection of the nanochannels.

KEYWORDS: nanotechnology, nanofluidics, capillarity, fluid interfaces

INTRODUCTION

In nanofluidics, capillary action is very prominent due to the large surface-to-volume ratios [1,2,3]. A clear example is the filling of nanochannels by capillarity [4,5,6], where the wetting properties of channel walls play a crucial role. The specific phenomena associated with fluid behaviour in nanochannels are of considerable interest to manipulate the transport of molecular or ionic species and (bio)chemical analysis [7,8]. For these applications the study of fluids in nanoconfinement is crucial. Especially the shape of the meniscus of a liquid that wets a solid and related elastocapillary effects [9,10], are subjects of large interest in nanofluidics.

We present an analytical model to describe the shape of the meniscus in deformating nanochannels, and propose a new method to quantify the channel deformation based on observation of the so-called protrusion length of the meniscus. To this purpose, we study micromachined hydrophilic nanochannels of 79 nm height and different widths capped by a thin membrane, that are filled with ethanol. The capping layers are easily deformed due to the induced negative Laplace-pressure in the liquid, and the in-plane shape of the meniscus is substantially modified [3]. The meniscus curvature changes from purely concave into convex-concave. See Figs. 1 and 2.

THEORY

Assume the channels to be directed along the z-axis, with their width w in the x-direction and height h along the y-axis. We describe the fluid surface by \( z = f(x,y) \) and write its orthogonal curvatures in terms of \( \nabla f = (\partial_x f, \partial_y f) \). The Young-Laplace equation, relating the sum of these curvatures to the Laplace-pressure, then yields

\[
-\hat{\nabla} \left[ \frac{\nabla f}{\sqrt{1 + (\nabla f)^2}} \right] = \frac{p^{(l)} - p^{(g)}}{\gamma}
\]

where \( p^{(l)} \) and \( p^{(g)} \) represent the pressures in the liquid and in the gas, respectively. Eq. (1) has to be solved for \( f \), together with the boundary condition that the fluid sur-
Figure 1. (left:) Microscopic image (top view) of an array of nanochannels of varying width (3.5 - 7.5 µm) partially filled with ethanol, showing the influence of the channel width (and therefore the membrane deflection) on the meniscus shapes. (right:) Image of the ethanol-air interface in the 5.4 µm wide channel, together with the theoretically calculated curve.

The rectangular channel is covered by an elastic membrane of thickness $t$, that is pulled downward due to the negative pressure inside the liquid. This downward deflection can be written as $u(x, z)$, and it obeys the equation of elasticity

$$\kappa \Delta^2 u(x, z) = \begin{cases} 0 & \text{membrane – air} \\ P^{(g)} - P^{(w)} & \text{membrane – liquid} \end{cases}$$

where $\Delta = \partial_x^2 + \partial_y^2$, $\kappa = E t^3/12(1-\nu^2)$, $E$ is the effective Young's modulus and $\nu$ the Poisson ratio of the membrane composite. A solution of the elasticity equation, Eq.(2), can in good approximation be written as

$$u = \delta (1 - \xi^2)^2$$

with $\delta$ the centre deflection and $\xi = 1 - 2x/w$ the normalized distance. The maximal deflection, $\delta$, is proportional to the pressure difference $\Delta p = P^{(g)} - P^{(w)}$. This makes the equations (1) and (2) coupled. If the deflection of the channel is not too large ($\delta/h < 0.11$), a solution for $f$ can be found analytically. It can be written in the form

$$f(\xi) = \frac{w}{2} \int \frac{P(\xi')}{\sqrt{1 - P(\xi')}} d\xi'$$

with

$$P(\xi) = \frac{w \Delta p}{2 \gamma} \xi + \frac{w \cos \theta}{4 h \xi_1 \sqrt{\delta}} \left( \frac{2 \xi_1}{\xi_2} \arctan \frac{\xi}{\xi_2} + \ln \frac{\xi + \xi_2}{\xi + \xi_1} \right), \quad \xi_{1,2} = 1/\sqrt{\delta} \pm 1$$

EXPERIMENTS AND RESULTS

Hydrophilic nanochannels of 79 nm height and widths ranging from 3.5 to 7.5 µm were fabricated as described recently [11] and filled with ethanol by capillary action. The membranes were scanned using an AFM and observed under an optical microscope: Fig. 1 shows the menisci at the fluid-air-interfaces of the partially filled channels. By digital contrast interpretation of the obtained grayscale images the meniscus shapes were composed (Fig.2), showing good correspondence of the shapes with the
Figure 2. Meniscus shapes determined from microscopic images (dots) and calculated curves according to the model (solid lines) for channel widths $w=3.41 \mu m$, $3.88 \mu m$, $4.41 \mu m$ and $4.88 \mu m$ (Fig.2a, b, c, and d, resp.) Both axes have been normalized by the channel width $w$.

model. Defining the protrusion length of the meniscus $l_p$ as the distance between the front of the meniscus and its attachment point at the channel wall, we can investigate its dependence on the membrane bending. Figure 3 shows the measurement results, together with the theoretical prediction. Clearly, the protrusion length is a very sensitive measure of the membrane deflection.

CONCLUSIONS

We obtained an analytical model to describe the fluid menisci that were seen to adopt a very peculiar shape in the deformed nanochannels. It was found that the proposed model is in good correspondence with the experimental results. Also, the protrusion length of the meniscus in the flexible nanoconfinement was in good agreement with the model predictions. Moreover, this length is found to be a very sensitive and convenient measure for the membrane deflection of the nanochannels.

REFERENCES