A mesoscopic approach for multi-phase flows in nano-corrugated channels

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An approach based on a lattice version of the Boltzmann kinetic equation for describing multi-phase flows in nano- and micro-corrugated devices is proposed. We specialize it to describe the wetting/dewetting transition of fluids in presence of nanoscopic grooves etched on the boundaries. This approach permits to retain the essential supra-molecular details of fluid-solid interactions without surrendering -actually boosting- the computational efficiency of continuum methods. The mesoscopic method is first validated quantitatively against Molecular Dynamics (MD) results of Cottin-Bizonne et al. [Nature Mater. 2 237 (2003)] and then applied to more complex situations which are hardly accessible to MD simulations. The resulting analysis confirms that surface roughness and capillary effects may conspire to promote a counter-intuitive but significant reduction of the flow drag with substantial enhancement in the mass flow rates and slip-lengths in the micrometric range for highly hydrophobic surfaces.

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The motion of fluids at the micro and nanoscale is controlled by the competition of dissipative effects and pressure drive. The weakness of inertia in the microworld implies that it is increasingly difficult to push fluids across micro/nanoconfined geometries, as their surface/volume ratio is made larger and larger. An obvious consequence is that the dynamics of microflows is crucially affected by the interaction of the fluid with the confining solid boundaries. Information on these interactions is usually conveyed into the formulation of proper boundary conditions for the fluid flow. In particular, slippage properties (see [2] for a recent review) have been reported in experiments and in molecular dynamics simulations, depending on the thermodynamical and wetting properties of the boundary (contact angle) and on the surface geometry. A fundamental question arises as to whether the fluid really slips over the surface, or rather the indirect measurements based on pressure/mass flow rate relations or surface force apparatus reflect an apparent slip arising from surface inhomogeneities or complex interface with additional physics. Indeed, it has been argued that a gas layer at the interface would alter the fluid dynamics in the bulk, leading to a mass flow rate increase even in the presence of pure no-slip [10, 11, 12, 22, 23]. This hypothesis is supported by the observation of nanobubbles trapped on the surface and by a decreasing apparent slip length as the fluid is degased [14].

The aim of this article is to discuss the complex effects, at the hydrodynamical scales, induced by the surface wetting properties in presence of complex geometries in micro- and nano-devices. The results can be summarized in two main points. First, we provide neat evidence that the physics of the boundary conditions is quantitatively reproduced by modeling the fluid at mesoscopic level, by means of a minimal version of the Boltzmann equation, i.e. the Lattice Boltzmann Equation (LBE) [15, 16]. This result is obtained by performing a quantitative comparison of a “finite-volume” dewetting transition against recent Molecular Dynamics simulations (MD) [17]. Far from being a mere technicality, this result opens the way to numerical investigations at spatial and time scales much larger than those currently available in most MD simulations. Second, we extend the MD results by investigating the critical dependency of the mass flow rate on the degree of roughness at constant bulk pressure. The simplest LBE reads as follows [10]:

\[
\begin{align*}
    f_i(x + c_i \Delta t, t + \Delta t) - f_i(x, t) = -\omega \Delta t [f_i(x, t) - f_i^{eq}(x, t)] + F_i \Delta t,
\end{align*}
\]

where \(f_i(x, t)\) is the probability of finding a particle at site \(x\) at time \(t\), moving along one of the \(b\)-th lattice direction defined by the discrete speed \(c_i\) with \(i = 1, \ldots, b\) and \(\Delta t\) is the time unit. The left-hand side of (1) stands for molecular free-streaming, whereas the right-hand side represents molecular collisions. These are expressed through a simple relaxation towards local Maxwellian equilibrium \(f_i^{eq}\) in a time lapse of the order of \(\tau = \omega^{-1}\). Finally the term \(F_i\) represents a volumetric body-force, which can be tailored to produce highly non-trivial macroscopic effects, such as phase-transitions. Non-ideal effects, leading to two-phase flows, are modeled through a self-consistent force term:

\[
    F(x, t) = \mathcal{G}_b \sum_i w_i \psi(x, t) \psi(x + c_i \Delta t, t) c_i.
\]
Here, $\psi(x)$ is a phenomenological pseudo-potential (generalized density), $\psi(x,t) = \psi(\rho(x,t))$, first introduced by Shan and Chen \cite{22}, $w_l$ are normalization weights and $G_b$ tunes the molecule-molecule interaction, i.e. it plays the role of the normalized inverse temperature, $\epsilon/KT$, with $\epsilon$ the molecular interaction, $K$ The Boltzmann constant and $T$ the system temperature. Here we choose the standard form $\psi = \sqrt{\rho_0}\{1 - \exp(-\rho/\rho_0)\}$, with the reference density $\rho_0 = 1$, in lattice units.

In spite of its simplicity, the Shan-Chen approach provides two crucial ingredients of non-ideal fluid behavior: a non-ideal equation of state and a non-zero liquid-vapor surface tension, $\sigma_{lv}$. Both features are encoded in the expression of the non-ideal momentum flux tensor $P_{kj}$. In the hydrodynamic limit, the LBE equations \cite{11, 22} can be shown to evolve according to the Navier-Stokes equations \cite{18} with a pressure tensor $\mathbf{P}$:

$$
P_{kj} = \left[ c_s^2 \rho + \frac{1}{2} c_s^2 G_b (\psi^2_0 + \frac{\tau}{4} \nabla^2 \psi_0^2) + \frac{\tau}{2} c_s^4 \nabla^2 \psi_0 \right] \delta_{kj} - \frac{1}{2} c_s^4 \nabla \psi_0 \nabla \psi, \tag{3}
$$

The equation of state in the bulk is $P = c_s^2 \rho + \frac{1}{2} c_s^2 G_b \psi^2$ where we recognize a non-ideal contribution on top of the ideal equation of state $P = \rho c_s^2$ with $c_s^2$ the sound speed velocity. This equation of state supports a phase-transition at a critical density $\rho_c/\rho_0 = \ln(2)$, whenever the coupling strength exceeds (in magnitude) the critical value $G_b = -4.0$. Concerning the velocity field, following \cite{18}, we set it to be zero at the boundary by using bounce-back boundary conditions while, for the case when slippage properties are a priori imposed, different boundary conditions should be used \cite{25, 26}. Next, the

$$
P_{cap} = \frac{2 \sigma_{lv} \cos(\theta)}{L_x - a} \tag{4}
$$

In Fig. 2 we validate the LBE by a direct comparison with the MD results published in \cite{11, 17} with the same geometry and with comparable contact angle. In particular, we show the Pressure drop between the bulk liquid and vapor phases $\Delta P_{lv} = P_l - P_v$, at changing the distance $d$ (see Fig. 1) at fixed total mass, i.e. at changing the average density. As one can clearly see, the agreement between LBE and MD is quantitative.
the two bulk phases, is shown as a function of the normal-
pressure, \(\Delta P_{lv}/h/\sigma_{lv}\) between
the two insets represent the density configura-
tion at the onset of the wetting/dewetting transition (right)
and for a wetted configuration (left). The plateaux in the
pressure curve defines the capillary pressure, \(P_{cap}\).

observed for \(\Delta P_{lv}\) in the range 0.9 < \(d/L_x\) < 1.05 corre-
sponds to the pressure/density values at which the fluid is
invading the corrugation, forming an interface which does
not yet touch the bottom of the groove. This corresponds
to the capillary pressure, \(P_{cap}\). Reducing further \(d\), i.e.
increasing the average density, a change of concavity for
0.8 < \(d/L_x\) < 0.9, is observed. This range corresponds
to values such that the interface starts to touch the bot-
tom of the groove, adjusting its pressure/density in such
way as to minimize the free energy in the presence of the
new liquid-vapor-solid interface. The agreement of
LBE with the capillary pressure \(P_{cap}\) is checked in detail
in Fig. 3, where we report the change of the pressure
diagram with a changing distance \(d\), for three different
corrugation values, \(L_x - a\). In the inset of Fig. 3 we
extract the value of \(\sigma_{lv}\cos(\theta)\) from the slope of the
observed plateaux vs \(\Delta x/(L_x - a)\). The value of \(\cos(\theta)\)
is then obtained by estimating the surface tension, \(\sigma_{lv}\),
through Laplace’s law for a droplet in equilibrium with
its saturated vapor. The agreement of the contact angle
measured in this way, \(\theta = 158^\circ \pm 6^\circ\), with the analytical
estimate, \(\theta = 160^\circ\), obtained in \(18\) by imposing the me-
chanical equilibrium condition of the contact line is very
satisfactory.

The comparison shown in Fig. 2 and Fig. 3 demonstrates
the first result discussed in this paper, namely that the
model introduced in \(18\) captures the correct interplay
between roughness and wetting effects.

Even more complex behavior is observed for heteroge-
neous nano-corrugations, with the simplest case shown
in the right panel of Fig. 1. In this case, one has
two characteristic groove sizes, \(H_1\) and \(H_2\), and hence
two corresponding critical capillary pressures. The pres-
sure/density diagram for this heterogenous corrugation is
shown in Fig. 1 where the two plateaux corresponding
to the two capillary pressures coexist. This device may
be considered a “smart” two-state surface, whose wetting
properties and mass throughput (under the application of
a pressure gradient ) may be tuned by changing the
bulk pressure.

The dynamical response of the micro-channel is investi-
gated by applying a constant pressure gradient. In Fig. 4
we show the mass flow rate as a function of the degree of
roughness, \(a/(L_x - a)\), for a fixed groove depth \(h\) and at a
given normalized pressure drop, \(\Delta P_{lv}/h/\sigma_{lv} = 0.75\). The
main result here is the presence of a transition at a criti-
ical roughness, \(a/(L_x - a)\), where the mass flow rate starts
to increase with respect to the perfect wetting situations
reaching as much as 100% gain. The strong dynamical
effect of the gas-layer can be quantified in more detail by
inspecting the velocity and momentum profiles along the
vertical direction without the vapor layer (fully-wetted
configuration) and with a thin vapor layer starting to
accumulate close to the bottom of the groove. This is
shown in the inset of Fig. 4 where the local slip length
is also depicted by extrapolation of the bulk profile in-
side the wall. As soon as a vapor layer is formed, the
local slip-length ramps-up, reaching values of the order
of the channel height \(45\Delta x \sim 15nm\). Even larger values
can be measured close to the dewetting transition, where
a well-developed vapor layer is formed inside the groove
(the so-called super-hydrophobic regime).

Summarizing, we have shown that an extension of lat-
Boltzmann equation for non-ideal fluids, can quantitatively account for the concerted effects between wetting phenomena and geometrical irregularities. In particular, the presence of nano/micro-irregularities in the flow geometries leads to sizeable effects with respect to the infinite volume liquid-gas transitions, as well as to a significant reduction of mechanical drag on the flowing fluid. The consequence of our results is twofold: from a theoretical perspective, it indicates that drag-reduction via geometry-induced wetting transitions is a non-specific phenomenon. On the practical side, the present LBE approach offers the opportunity to perform very efficient numerical simulations of complex micro/nanofluidic phenomena at scales of direct experimental relevance, which are hardly accessible to atomistic simulations. This will open the way to the systematic optimization of microfluidic devices via computer simulation. Useful discussions with J.-L. Barrat, X. Shan and S. Troian are kindly acknowledged.

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