Modeling and simulation of the fluid–solid interaction in wetting

To cite this article: Fabiano G Wolf et al J. Stat. Mech. (2009) P06008

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Modeling and simulation of the fluid–solid interaction in wetting

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Received 3 November 2008
Accepted 5 March 2009
Published 15 June 2009

Abstract. In this work, a lattice-Boltzmann method based on field mediators is proposed for the simulation of wetting in a liquid–vapor system. The method includes the effect of long-range interactions between the particles and solid walls, and is implemented together with a liquid–vapor model which is known from the literature. The results that were obtained in the simulations show that the contact angle is strongly dependent on the long-range interactions. The initial formation of a film ahead of the macroscopic bulk liquid is observed below a specific contact angle. When rough solid surfaces were used, the simulations exhibit the contact angle hysteresis. The spreading dynamics on solid surfaces was studied and showed a considerable discrepancy from the expected value, which was attributed to the high resolution of the numerical simulations. It is shown that the initial conditions have a strong influence on the first steps of the droplet spreading, but they become negligible at long times.

Keywords: wetting (theory), microfluidics (theory), discrete fluid models, lattice Boltzmann methods
1. **Introduction**

The study of problems involving wettability has been a subject of great scientific and economic interest. The importance of this study is manifested by the many technological processes that include the direct application of fluids on surfaces of all kinds. Some examples are found in industries related to coating, lubrication, fabrics, advanced oil recovery, agriculture and ink-jet printers. Wettability is an important property when three phases coexist at the same place, like liquid–vapor–solid and liquid–liquid–solid systems, because it represents the macroscopic result of microscopic interactions between the fluid and solid molecules [1]. In a general sense, such a property measures the affinity of one particular liquid for the solid. Frequently, the wettability is quantified by using the definition of the contact angle, which is the angle formed between the tangent line to the solid and that tangent line to the liquid–vapor interface; a smaller contact angle for a given surface means a better wettability.

In the context of modeling of wetting on solid surfaces, a lattice-Boltzmann method is proposed for modeling the physical phenomena related to the fluid-solid interaction. It uses the concept of field mediators [2]–[5] to take the effect of attractive long-range interactions between the particles and solid walls into account. The role of field mediators is to mediate the long-range interactions among the particles located on a discrete lattice in such a way that the information on the field from the neighborhood sites is found locally at each site on the lattice. The purpose is to avoid the computing awkward searching procedure around a given site. To achieve this, liquid–vapor coexistence is required as a starting point and was done by modeling the fluid–fluid interaction using the Shan and Chen model [6, 7]. That model enables the simulation of phase equilibrium retrieving an equation of state similar to the van der Waals equation.

doi:10.1088/1742-5468/2009/06/P06008

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### Contents

<table>
<thead>
<tr>
<th>Section</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>1. Introduction</td>
<td>2</td>
</tr>
<tr>
<td>2. The lattice-Boltzmann method</td>
<td>3</td>
</tr>
<tr>
<td>2.1. Interaction forces between the fluid particles</td>
<td>4</td>
</tr>
<tr>
<td>2.2. Long-range attractive forces between fluid and solid particles</td>
<td>5</td>
</tr>
<tr>
<td>3. Results and discussion</td>
<td>8</td>
</tr>
<tr>
<td>3.1. Measuring the static contact angle</td>
<td>8</td>
</tr>
<tr>
<td>3.2. Relationship between the contact angle and microscopic parameters</td>
<td>9</td>
</tr>
<tr>
<td>3.3. Rough surfaces and hysteresis</td>
<td>10</td>
</tr>
<tr>
<td>3.4. Spreading dynamics</td>
<td>13</td>
</tr>
<tr>
<td>3.5. Influence of the initial conditions</td>
<td>16</td>
</tr>
<tr>
<td>4. Conclusions</td>
<td>19</td>
</tr>
<tr>
<td>Acknowledgments</td>
<td>20</td>
</tr>
<tr>
<td>References</td>
<td>20</td>
</tr>
</tbody>
</table>
For simplicity and computational convenience, only two-dimensional situations were considered, allowing us to explore different aspects of wetting under dynamic and static conditions. In such a way, the model was used to study the relationship between the contact angle and microscopic parameters, the influence of roughness on the measured contact angle, the droplet spreading dynamics and its dependence on the initial conditions.

2. The lattice-Boltzmann method

The lattice-Boltzmann method (LBM) [8]–[10] is a mesoscopic method for the description of fluids, based on a mechanical system of particles. Although it has been, historically, originated from Cellular Automata models [11, 12], it has recently been shown [13]–[17] that LBM can be considered as based on a special discrete form of the continuous Boltzmann equation. The LBM is built on the mesoscopic scale, in which the system description is done using a single-particle distribution function, \( f_i(x, t) \), representing the number of particles with velocity \( c_i \) at the site \( x \) and time \( t \), where \( i = 0, \ldots, b \). In LBM, the particles are restricted to a discrete lattice in such a manner that each group of particles can only move along a finite number \( b \) of directions and with a limited number of velocities. Therefore, both the physical and the velocity spaces are discretized. In terms of \( f_i(x, t) \), the local macroscopic properties such as total mass density (the particle mass, \( m \), is assumed unitary) and total momentum can be obtained from \( f_i(x, t) \) by using

\[
n(x) = \sum_i f_i \quad \text{and} \quad n(x)u(x) = \sum_i f_i c_i,
\]

respectively.

As a result of collisions, the single-particle distribution function \( f_i(x, t) \) is modified on each site during the time interval \( \Delta t \). The evolution equation that describes this change is given by

\[
f_i(x + c_i \Delta t, t + \Delta t) - f_i(x, t) = \Omega_i,
\]

where \( \Omega_i \) is the collision operator. This last term must be written in such way as to preserve the total mass and momentum in collisions. In non-isothermal systems energy preservation is also required. The simplest form is given by the Bhatnagar, Gross and Krook model, which describes a collision as a relaxation process to a local equilibrium state, \( \Omega_i = -(\Delta t/\tau)(f_i - f_i^{(eq)}) \), where \( \tau \) is a relaxation time and \( f_i^{(eq)} \) is a single-particle local equilibrium distribution. Hence, the governing LBM mesoscopic equation will be

\[
f_i(x + c_i \Delta t, t + \Delta t) - f_i(x, t) = -\frac{\Delta t}{\tau}(f_i - f_i^{(eq)}).
\]

After the collision step, the evolution equation, equation (2), demands the local information at \( x \) and \( t \), \( f_i(x, t) \), to be transferred to the neighboring site \( x + c_i \Delta t \) at time \( t + \Delta t \) in the propagation step.

The macroscopic behavior of equation (2) is determined by a suitable choice of the equilibrium distribution, \( f_i^{(eq)} = f_i^{(eq)}(n, u) \). In the present case, one needs to recover the macroscopic dynamics of fluids ruled by the Navier–Stokes equations. For this purpose, the equilibrium distribution is chosen as [8]

\[
f_i^{(eq)}(x) = mw_i \left[ 1 + 3 (c_i \cdot u) + \frac{3}{2} (c_i \cdot u)^2 - \frac{3}{2} u_i^2 \right],
\]

where \( c = |c_i| \), \( w_0 = 4/9 \), \( w_1 = w_3 = w_5 = w_7 = 1/9 \) and \( w_2 = w_4 = w_6 = w_8 = 1/36 \) for a D2Q9 lattice. The \( w_i \)'s are weight factors chosen to guarantee macroscopic isotropy and Galilean invariance [8].
Using a multiscale method such as Chapman–Enskog [18], it is possible to show that the system described above recovers the Navier–Stokes equations in the limit of near incompressible flows [8,9,19,10]. Next to the solid surfaces, a halfway bounce-back boundary condition [10] is imposed for the particles leaving the fluid domain.

2.1. Interaction forces between the fluid particles

In order to take into account the physical ingredients found in the microscopic interactions among the molecules of a fluid, Shan and Chen [6,7] inserted an interaction potential among the particles in the LBM. In this way, the model is able to reproduce the liquid–vapor phase transition and coexistence of two distinct phases of a single component in equilibrium, in which the density distribution allows us to identify both phases. The authors used the following nearest-neighbor interaction potential between a pair of particle groups localized at the sites $x$ and $y$:

$$V(x, y) = \varphi(x)G_i(x, y)\varphi(y),$$

where, for a D2Q9 lattice [20], $G_i(x, y) = G$ for $|c_i| = \sqrt{2}$, $G_i(x, y) = 4G$ for $|c_i| = 1$ and $G_i(x, y) = 0$ for two sites which are not nearest-neighboring sites. The function $\varphi(x) \equiv \varphi[n(x)]$ will be chosen in a way to establish a correspondence between the LBM and an isotherm in a $PVT$ system. The strength of the interaction potential among the particles is given by $G$, while its sign defines if the potential is attractive or repulsive. The resulting interaction force due to this potential acting on the particles located at the site $x$ at time $t$ is then

$$F_\sigma(x, t) = -\varphi(x)\sum_i G_i\varphi(x + c_i\Delta t)c_i.$$  \hspace{1cm} (5)

In the LBM, the local velocity, $u(x)$, in the equilibrium distribution, equation (3), is modified due to the force $F_\sigma$ in the following way:

$$u_{eq} = u + \frac{\tau}{n}F_\sigma.$$  \hspace{1cm} (6)

This type of interaction among the particles does not preserve the local momentum during the collisional process because the force $F_\sigma$ behaves as an external force acting on the site. Nevertheless, Shan and Chen [6,7] showed that the total momentum of the system is preserved and no net momentum is inserted in the system by the above interaction. Redefining the fluid momentum, $nv$, as an arithmetic average for the states before and after the collision step, $n\mathbf{v} = n\mathbf{u} + \frac{1}{2}F_\sigma$, the method of Chapman–Enskog [18] allows us to determine the macroscopic behavior of the referred microscopic system. The resulting macroscopic equation describes a non-ideal fluid whose equation of state is written for a D2Q9 lattice as

$$P = \frac{n}{3} + 6G\varphi^2(n).$$  \hspace{1cm} (7)

The correct choice of the function $\varphi(n)$ enables the modeling of a wide variety of fluids, including the liquid–vapor phase transition. Supposing the equation of state, equation (7), to correspond to an isotherm in the $PVT$ system, Shan and Chen [7] determined a specific
coexistence curve, concluding that if \( \varphi(n) \) is chosen to be

\[
\varphi(n) = \varphi_0 \exp(-n_0/n),
\]

where \( n_0 \) and \( \varphi_0 \) are arbitrary constants, the LBM will be consistent with that of an isothermal process \([7]\). Further on the functional, equation (8), the authors showed that the model \( \varphi(n) = \varphi_0[1 - \exp(-n/n_0)] \) can also be used, because the resulting coexistence curve exhibits notable discrepancies with respect to the functional, equation (8), only when the temperature parameter goes far below the critical value \([7]\).

2.2. Long-range attractive forces between fluid and solid particles

When there are \( n(x) \) molecules located at point \( x \) of the fluid and \( n_s(y) \) sources of attraction located at \( y \), if the shape and size of molecules are not considered, an approximated form for the potential energy will be

\[
\Phi = n(x)n_s(y)\phi(|x - y|),
\]

where \( \phi(|x - y|) \) is a function dependent on the distance separating \( x \) and \( y \). Therefore, the potential energy related to the intermolecular interaction between the fluid molecules at a position \( x \) and the solid attractive sources at a point \( y \) in the solid phase can be written as

\[
\Phi^{(n)}(|x - y|) = n(x)n_s(y)\omega(|x - y|),
\]

where \( \omega \) is a parameter related to the potential strength at the surface and \( \alpha \) is a decaying function, which will be written as

\[
\alpha(|x - y|) = \exp(-|x - y|/\kappa),
\]

where \( \kappa \) is a constant related to the potential interaction length.

From a physical point of view, the electrostatic field generated by the atoms on the surface of a solid is affected by the electrical properties of the fluid molecules in its neighborhood. Indeed, an electrical field can induce or modify the dipolar moments of these molecules, in accordance with their polarizability. The interaction between a fluid and a solid surface will, thus, simultaneously depend on the surface itself and on the electrical polarizability of the fluid molecules. In the present paper, the parameter \( \omega \) is used to describe the surface, giving the potential strength, i.e. the potential very close to this surface. In addition to being dependent on the solid itself, the potential strength \( \omega \) is dependent on the electrical properties of the fluid molecules, since the electrical field around these molecules affects the potential at the solid sites. On the other hand, the parameter \( \kappa \) in the decaying law, equation (11), is directly related to the way the fluid feels the solid wall, i.e. to its polarizability: when the fluid molecules have a low polarizability, the surface potential have a strong decay and the fluid behaves as if it was free.

Since only attractive fields were considered for simulating the potential energy between any two particles, when the above expression, equation (11), is used to calculate the potential energy between two sites in the fluid phase, it produces a mass collapse in the sites, since LBM particles are supposed without volume and each site becomes an increasingly attractive sink of particles as the number density of particles increases in this site. This is the main tactical reason that prevented the use of this potential form for

doi:10.1088/1742-5468/2009/06/P06008
calculating the potential energy among fluid particles in the last section. In the present case, although the sites in the solid phase act as attractive sinks of fluid particles, the bounce-back boundary condition used for the particles that reach the solid surface prevents the accumulation of these particles on the boundary. In physical terms, a density increase of the liquid in the immediate neighborhood of the solid is to be expected, but the correct prediction of this mass accumulation is a difficult and open question and would require a more detailed analysis, taking the correct fluid–solid interaction laws into account. The detailed description of this solid–fluid transition layer is beyond the purposes of this work.

In a discrete space, the potential energy at \( x \) due to the interaction with the solid wall \( S \) will be

\[
\Phi^{(s)}(x) = \sum_{y \in S} n(x)n_s(y)\omega(\|x - y\|). \tag{12}
\]

From a numerical point of view, the above calculation is extremely cumbersome because it requires, for each time step and for each point \( x \), a searching step along all the neighborhood sites of \( x \) that belong to the solid phase.

In this work, a lattice-Boltzmann method is proposed for modeling the physical phenomena related to the fluid–solid interaction. It uses the concept of field mediators [2]–[5] to take the effect of attractive long-range interactions between the particles and solid walls into account. The role of the field mediators is to mediate the long-range interactions among the particles located on a discrete lattice in such a way that the information on the field from the neighborhood sites is found locally at each site on the lattice. The purpose is to avoid the computing awkward searching procedure around a given site. To get such an effect, some particles with null mass, called mediators, are emitted from the solid sites at each time step. Each mediator is able to carry with it, the information on the number density of attractive sources on the solid site from where it was emitted. As a consequence, each fluid site next to the wall gets this information after a few time steps and this information is used to modify the local momentum of the fluid particles. Since a field travels at the speed of light, this delay in field information was investigated by Santos, Facin and Philippi [3] in the study of capillary waves and no meaningful effect was observed. For the simulation of the fluid–solid interaction using field mediators, the following procedure has been adopted. At each time step, field mediators are emitted from the solid sites with the information on the number density of the attractive sources \( n_s \) and on the potential strength \( \omega_i \):

\[
M_i^{(s)} = n_s\omega_i, \tag{13}
\]

where, for a D2Q9 lattice [20], \( \omega_i = \omega \) for \( |c_i| = \sqrt{2} \), \( \omega_i = 4\omega \) for \( |c_i| = 1 \) and \( \omega_i = 0 \) for two sites which are not nearest-neighboring sites.

In the propagation step, the mediators are moved from \( y \) to the nearest-neighbor sites \( y + c_i\Delta t \), suffering an attenuation, in accordance with the following relation:

\[
M_i^{(s)}(y + c_i\Delta t, t + \Delta t) = \alpha(r)M_i^{(s)}(y, t), \tag{14}
\]

where \( r \) is unity, because the mediators are only moved to the nearest-neighboring sites. After this step the new variable \( M_i^{(s)} \) contains the information: (i) on the number density of attractive sources at the point \( y \) of the solid surface; (ii) on the potential strength and (iii) on the distance traveled by the mediator. Figure 1 illustrates this numerical scheme.
The fluid–solid interaction force, $F_s(x, t)$, on the particles located at the site $x$ at time $t$, due to the field mediators coming from the solid wall, can thus be written as

$$F_s(x, t) = -n(x) \sum_i M_i^{(s)}(x, t)c_i. \quad (15)$$

The fluid–solid force, equation (15), is imposed on the particles through a change in their momentum, in the same way as explained in section 2.1.

In comparison with other models, the present model includes explicitly the effect of long-range forces among the particles, allowing the interaction length to be easily controlled by only changing a single parameter. Such an attribute had not been considered in previous models, such as the ones by Martin and Chen [21] and Raiskinmaki et al [22], based on nearest-neighbor interactions. In the work by Zhang and Kwok [23] a distance-dependent fluid–solid potential was considered explicitly in describing the fluid–wall interaction. However, the methodology adopted by Zhang and Kwok assumes the use of a simple wall geometry—a flat plate—for which the net force on the fluid particles at a given site is previously known and only depends on the height $z$. The interaction field in the neighborhood of more complex solid surfaces, such as rough surfaces, depends on the coordinates $x$ and $y$, in addition to $z$, requiring previous knowledge of the specific fluid–wall potential. In the present model, it is only necessary to know the inter-particle potential itself, the field information being carried out from the solid sites to the fluid sites and among fluid sites by mediators, avoiding the use of simplified $z$-dependent decaying laws. In contrast, more complex solid surfaces demand lattices with higher numbers of lattice directions, since the emission and propagation steps are carried out according to the nearest-neighbor sites. Higher-order lattices would thus imply a better representation of the net force on a given site. Benzi et al [24] discussed a generalization of the Shan and Chen model [6, 7] which includes the fluid–wall interaction. They also showed an extension of the model with long-range forces by using a distance-dependent potential similar to the one used by Zhang and Kwok [23].

doi:10.1088/1742-5468/2009/06/P06008
3. Results and discussion

3.1. Measuring the static contact angle

In this work, the measurement of the static contact angle was carried out by considering two different techniques, both of them putting in evidence the contact angle as a macroscopic property in essence. The first method is based on geometrical relations which are grounded on a cylindrical bidimensional droplet shape when gravity is missing. Due to the field mediators, the long-range force length, $\ell_{FS}$, is larger than one lattice spacing, which induces the deformation of the liquid–vapor interface curvature near the triphasic contact line, the use of cylindrical-shape-based relations becoming imprecise in the measurement of the contact angle. The mentioned inaccuracy can be avoided if the angle, $\theta(y)$, is measured far away from the contact line, at a specific height $y$ from the solid surface, and after that the static contact angle is obtained by extrapolating the cylindrical shape up to the solid surface, $y = 0$. This is illustrated in figure 2. Therefore, the static contact angle is a function of geometrical parameters such as the droplet base radius at height $y$, $r(y)$, the droplet height, $H$, and $\theta(y)$, and can be calculated by

$$\theta_s = \arccos \left[ 1 - \frac{H}{r(y)} \sin\theta(y) \right],$$

where $\theta(y) = 2\arctan[(H - y)/r(y)]$. In all the cases considered here, $y = 4.5$ lattice spacings is chosen. The determination of droplet dimensions, as $r(y)$ and $H$, was done through the density field by considering the abrupt variation of local densities, which occurs near the interface region. From the calculation of the density gradient in a specific direction, for instance $y$, $dn/dy \approx [n(y + \Delta y) - n(y - \Delta y)]/2\Delta y$, the interface position can be determined by noticing that $|dn/dy|$ is a maximum at the liquid–vapor interface. Considering a specific coordinate system, as shown in figure 3, all droplet dimensions are available.

In these simulations there was observed the detachment of a precursor film ahead of the macroscopic bulk liquid, when the contact angle is smaller than around $34^\circ$ ($\omega = 0.3$ and $\kappa = 0.5$) (see figure 4). When the film movement ceases, it has something similar to a
‘tongue’ ahead of the droplet as cited by Kavehpour et al [25]. The presence of a precursor film in physical systems, whose thickness is normally below the micron range [26], has been reported by several authors in the literature (see, e.g., [27]–[29], [26, 30, 25]). That film exhibits a peculiar dynamics and it will be not explored here, because the main objective of this work is to study the wettability from a macroscopic point of view by comparing simulation results with experiments available in the literature.

A second technique for measuring the contact angle was used due to the presence of the mentioned precursor film, which breaks down the supposition of a cylindrical droplet shape, particularly for small contact angles. An alternative way to measure the contact angle is given in terms of the inflection point. The existence of an inflection point is a requirement for droplets with small apparent contact angles and provides a connection between the inner region near the ‘tongue’ and the outer macroscopic bulk liquid region [25], as is illustrated in figure 3. The determination of the inflection point is based on the knowledge of the droplet profile, \( h(x) \), and can be obtained using the numerical procedures which incorporate the maximum density gradient method as described above. The droplet profile is built from the set of all interface positions on a given coordinate system. From the droplet profile, the macroscopic contact angle is conventionally defined as \( \theta_a = \max [\arctan(dh/dx)] \) [31, 25], which is the slope of the liquid–vapor interface at the inflection point.

3.2. Relationship between the contact angle and microscopic parameters

The method proposed in this work allows the interactions occurring between the lattice sites separated by greater distances than one lattice spacing. Thus, it is possible to investigate the static contact angle, \( \theta_s \), as a function of the microscopic parameters related to the long-range fluid–solid potential, given by the strength and length of the fluid–solid
interaction, ω and κ, respectively. For this purpose, a lattice D2Q9 with 500 × 78 sites was initialized with the parameters τ = 1.0 and G = −0.15, resulting in a liquid–vapor density ratio, n_L/n_V ≃ 20. For simplicity and computational convenience, only two-dimensional simulations were considered. The initial condition was based on a liquid droplet (n_L = 2.261) with an initial diameter, R_0, equal to 35 lattice spacings in contact with a solid surface (n_S = 1.0). In all domain boundaries periodic boundary conditions were imposed. The dependence between θ_s and the parameters ω and κ is shown in figures 4 and 5.

In figure 5, the obtained results show that the static contact angle is strongly dependent on the interaction potential defined by the microscopic parameters ω and κ. By varying the strength of the fluid–solid interaction, it was verified that any contact angle between 0° and 180° can be simulated and that an increase in the adhesion forces leads to a decrease in the contact angle. This is in agreement with simulation results based on molecular dynamics methods [32,33]. The dependence between θ_s and ω seems to be approximately linear, similar to the results related by Zhang and Kwok [23] who used the mean-field free-energy lattice-Boltzmann approach. The parameter κ controls the fluid–solid interaction length of the field mediators and allows us to modulate the influence on the triphasic contact line from the particles that are located at sites far from the solid surface, 5(b).

3.3. Rough surfaces and hysteresis

Most of the experimental results available in the literature [34]–[36] are based on the apparent contact angle, θ_ap, and not on the actual contact angle [35], θ_s, which is a very
Figure 5. Relation between the static contact angle and controlling parameters of the long-range fluid–solid interactions ($\kappa = 0.25$, $\omega_{\text{max}} - \omega_{\text{min}} = 2.6$; $\kappa = 0.50$, $\omega_{\text{max}} - \omega_{\text{min}} = 0.275$; $\kappa = 1.00$, $\omega_{\text{max}} - \omega_{\text{min}} = 0.045$). The parameters $\omega_{\text{max}}$ and $\omega_{\text{min}}$ represent the extremum values when $\theta_s \simeq 0^\circ$ and $\theta_s \simeq 180^\circ$, respectively.

difficult property to be determined [36]. Such difficulty results from the dependence of the contact angle on the surface roughness and chemical heterogeneity. Such non-unicity on the static contact angle is referred to frequently as contact angle hysteresis [36].

For the purpose of illustrating the influence of solid roughness on the measured static contact angle, a rough surface is sketched by triangle-like patterns. For the drop case, the
Contact angle hysteresis can be easily observed when very different initial conditions are imposed. The simulation is initiated by considering three different liquid drops in contact with a rough surface, as shown in figure 6.

In the simulation, the mechanical equilibrium condition was supposed to be reached after around $3 \times 10^4$ time steps. In figure 7(a) is shown the final configuration of three liquid drops arranged according to the initial conditions in figure 6. As can be seen, from a macroscopic point of view, the static contact angles seem to be very distinct from each other, even though the entire solid surface exhibits the same fluid–solid interaction, with a corresponding static contact angle of around $69^\circ$. By tracing a straight line parallel to the solid surface, one can determine the following apparent contact angles (from left to right): $\theta_{ap}^{(1)} \simeq 66^\circ$, $\theta_{ap}^{(2)} \simeq 105^\circ$ and $\theta_{ap}^{(3)} \simeq 119^\circ$. It is important to notice that, if the surface were ideally smooth, the three drops would show exactly the same static contact angle, that is, $\theta_{ap}^{(1)} = \theta_{ap}^{(2)} = \theta_{ap}^{(3)} = \theta_s \simeq 69^\circ$, without any influence of the initial conditions. These obtained results show that the measured static contact angle is strongly influenced by the roughness of the surface as a result of the changes in the curvature of the liquid–vapor interface. Though those dissimilar initial conditions were generated artificially, they might occur in laboratory conditions. Thus, a given measured contact angle is dependent on its own history.

doi:10.1088/1742-5468/2009/06/P06008
A sinusoidal-shaped solid surface was studied theoretically by Johnson and Dettre [37]. Focusing the analysis on the drop size scale, those authors were able to show that more than one static contact angle can be obtained and that all observable contact angles are in a bounded range, similar to the results shown in this work. Notice that in the simulation shown in figure 7(a) the effect of roughness is overestimated, since that roughness pattern has the order of magnitude of the drop size. Nevertheless, when the characteristic drop size is increased, the same effect is observed. Figure 7(b) illustrates such a result, in which the doubled size drops were simulated on the same rough surface of figure 7(a). Evidently, the effect of roughness on the apparent contact angle decreases when this roughness is reduced.

Such interesting results demonstrate an intrinsic property of the liquid–vapor–solid system: the locality of the contact angle. As was mentioned above, the measured static contact angle is modified by the roughness, requiring the definition of an apparent contact angle.

3.4. Spreading dynamics

The relaxation of a liquid drop to the equilibrium state is analyzed by considering the time evolution of its apparent base radius, \( R(t) \), outlined in figure 3. It is well known (see, e.g., [38, 28, 29]) that, under complete wetting conditions (\( \theta_s \to 0^\circ \)), a liquid droplet spreads on a solid surface exhibiting a universal behavior, independent of the material properties of the involved substrate. Such behavior is commonly referred to as Tanner’s law [38] and is represented by a power law for droplet base radius, \( R(t) \), given by

\[
R(t) \propto t^N,
\]

(17)

where \( N \) is the spreading exponent, being \( N = 1/7 \) for two-dimensional drops and \( N = 1/10 \) for three-dimensional ones. In most of the experimental results reported in the literature in three-dimensional systems, the exponent \( N \) was found in the range between 0.1 and 0.145, although values of 0.033 and 0.3135 have already been reported [28]. Tanner’s law is only valid in the macroscopic scale, since microscopic droplets follow a different spreading dynamics dominated by van der Waals forces [39].

For studying the spreading dynamics resulting from the model described above, a two-dimensional system of \( L \times H \) sites was used with ‘mirror’ and periodic boundary conditions imposed on the top and left/right, respectively. The bottom line in the domain is filled with solid sites, representing a solid surface with a thickness of one lattice spacing and corresponding physical dimension, \( \delta_x \). In the considered cases, the interaction length of the long-range fluid–solid forces, \( \ell_{FS} \), is not negligible with respect to the system size (\( \ell_{FS} \approx 6\delta_x \) for \( \kappa = 0.5 \)). It is thus necessary to determine if the characteristic scale in the simulations is comparable to the laboratory conditions, for which Tanner’s law is recovered. For that, a liquid drop with radius \( R_0 \) was initialized in contact with the solid surface. The drop radius was varied from 30 to 150 lattice spacings, but keeping the values of the ratios \( L/2R_0 = C_1 \) and \( L/H = C_2 \) constant, where \( C_1 \approx 6.4 \) and \( C_2 \approx 3.6 \). In this manner, an increase in the droplet size led to a proportional increase in the domain size. The model parameters \( G \) and \( \omega \) were adjusted to \(-0.15\) and \(0.06\), respectively, resulting in a liquid drop with density \( n_L = 2.261 \) immersed in vapor with density \( n_V = 0.116 \), a surface tension \( \gamma = 0.086 \) and a static contact angle \( \theta_s \approx 180^\circ \), where all parameters are in lattice units. That configuration was kept constant for \( 3 \times 10^4 \) time steps, for which
the system came into the mechanical equilibrium state. After that, the strength of the fluid–solid interaction, $\omega$, was set to 0.335, for which $\theta_s \to 0^\circ$, simulating a liquid–vapor–solid system in which the liquid wets the solid surface completely. The obtained results are shown in figures 8 and 9.

In figure 8, the time evolution of the droplet at the first steps of spreading is seen, when the droplet assumes different shapes before attaining an almost circular shape in the liquid bulk. There is observed an initial formation of a film ahead of the droplet, which is faster than the main part. Figure 9 shows the apparent droplet base radius, $R^* = R/R_0$, as a function of the reduced time, $t^* = t\gamma/(\nu_L L R_0)$ [40], where $\nu_L$ is the kinematic viscosity given by $1/6$ in the D2Q9 lattice [8]. For analyzing the spreading behavior at long times, the obtained curves were fitted in the ranges $100 \leq t^* \leq 520$ and $150 \leq t^* \leq 520$ by supposing a power law like

$$R^* = C (t^* - t_0^*)^N,$$

where $N$, $C$ and $t_0^*$ are constants calculated by the fitting process.

Figure 9(a) presents the results for the dimensionless apparent droplet base radius versus reduced time for different droplet sizes. It is noticed that there is a superposition of curves for $R_0$ greater than about 100 lattice spacings, which means that larger droplets spread in a similar way, leading to a single ‘master’ curve. The analysis of the results presented in figure 9(b) indicates that the effect of the droplet size on the spreading dynamics is almost irrelevant in practical terms, when the analysis was performed by taking into account later periods of time, as was made for the reduced time in the range

$$100 \leq t^* \leq 520 \quad \text{and} \quad 150 \leq t^* \leq 520.$$
150 ≤ t∗ ≤ 520. As is observed in the range 100 ≤ t∗ ≤ 520, there is a size dependence on the spreading, related to the influence of the initial conditions on the spreading dynamics.

However, it is noticed that the mean spreading exponent simulated by the LB model, $\overline{N} \approx 0.2423 \pm 0.0005$ (with a mean coefficient of determination $R^2 = 0.9997$), exhibits a considerable deviation from the expected value of $1/7$, predicted by equation (18). Some

doi:10.1088/1742-5468/2009/06/P06008
LBM results under two-dimensional and complete wetting conditions were also provided by Iwahara et al [41], obtaining $N$ equal to 0.43 and attributing such a discrepancy to the imposed initial conditions. They also stated that the droplet size did not influence the spreading exponent, according to the results obtained here in the range $150 \leq t^* \leq 520$. A possible explanation for the discrepancy in these results can be related to the high resolution of these numerical experiments.

To clarify the reason for that difference between numerical and experimental results, the power law, equation (18), was determined for distinct liquid layers next to the solid wall, represented by the liquid layer–solid distance, $y - y_s$, where $y_s$ is the solid plate position on the $y$ axis. The results shown in figure 9 were obtained by measuring the exponent $N$ on the first liquid layer above the solid wall, i.e. where $y - y_s = 1$ lattice spacing.

For the determination of the spreading exponent as a function of the distance between each layer and the solid surface, two methods were used: (i) the apparent droplet base radius was measured in the same way as was done above by using the inflection point on the droplet profile, $h(x)$, and (ii) the actual droplet base radius was measured directly from the density field, $n(x, y)$, with the maximum gradient method. The obtained results are shown in figure 10.

It is noticed that the spreading exponent is dependent on the distance from the solid wall; the liquid layers spread following different power laws and those nearest the solid move faster than others. The measuring process based on the actual droplet base radius shows that the liquid layers relative to the precursor film spread at higher rates than the bulk liquid, which is not seen when the method based on the profile slope at inflection is applied.

Both methods provide the same dependence between $N$ and $y - y_s$ for distances greater than or equal to 8 lattice spacings, where the liquid layer spreads with the exponent in a power law similar to that observed experimentally. Such a distance between the liquid layer and the solid, or at least the region between 6 and 8 lattice spacings far from the solid wall, seems to represent very well the transition between macroscopic and microscopic regions. It should be noticed that most of the experimental results for droplet base radius are based on low-resolution optical measurements, so that only the macroscopic portion of the droplet is monitored, i.e. the inner region composed by the precursor film is invisible and does not influence the measurement of the power law exponent. In terms of the interaction length, $\ell_{FS}$, that liquid layer where the simulated power law agrees with the experimental results is found at $(y - y_s)/\ell_{FS} \approx 1.33$ and is represented by the dashed line in figure 10(b). As can be seen, the droplet profile above that delimiting transition region is weakly affected by the presence of the precursor film. Therefore, when the tangent line is extrapolated from the inflection point to the solid wall, one gets a spreading dynamics which appears to be non-representative of the experimental visualizations. This could explain the differences that were found between the numerical and experimentally determined exponents in Tanner’s law, equation (18).

### 3.5. Influence of the initial conditions

Tanner’s law [38] has been considered a universal law, under complete wetting conditions, since most of the experimental results has pointed out an exponent $N$ in the range between
Figure 10. (a) Spreading exponent as a function of the distance from the solid wall calculated by two methods mentioned in the text for $R_0 = 50$ lattice spacings. In (b) one has droplet profiles, $h(x)$, at different reduced times. Notice the dashed line at $(y - y_s)/\ell_{FS} \approx 1.33$, where the simulated $N$ is closer than that predicted by Tanner's law.
Modeling and simulation of the fluid–solid interaction in wetting

Figure 11. First stages in the droplet spreading for the initial condition defined by $\theta_0 = 90^\circ$.

0.1 and 0.145, although values of 0.033 and 0.3135 have been reported [28]. In general, Tanner’s law is observed after a short transient succeeding the liquid–solid contact (see figure 8) [29,26,42]. That short transient period is influenced by the imposed physical conditions when the droplet is put in contact with the solid surface, and should depend on the height, velocity and shape of the droplet, etc. In this way, strong deviations from the universal law are expected due to the large number of variables involved in that initial stage.

Here, the influence of the initial conditions on the spreading dynamics is analyzed by considering the same droplet volume and radius of $R_0 = 50$ lattice spacings under three different initial conditions defined by the initial static contact angle, $\theta_0$, equal to 90°, 135° and 180°. For each initial contact angle, the system was kept unchanged for $6 \times 10^4$ time steps. After that, the strength of the fluid–solid interaction, $\omega$, was set to 0.335, for which $\theta_s \to 0^\circ$. The first stages of the droplet spreading for the cases $\theta_0 = 90^\circ$ is shown in figure 11 and the reduced time evolution of the dimensionless apparent droplet base radius, $R^* = R/R_0$, is seen in figure 12. Notice that the exponent $N$ is calculated on the first liquid layer above the wall, as mentioned previously.

Figure 12(a) exhibits the evolution of the droplet base radius for three different initial conditions defined by $\theta_0$. It is noticed that the spreading dynamics for each case is completely dependent on the imposed initial conditions, varying from $N \approx 0.60$ for $\theta_0 \simeq 180^\circ$, $N \approx 0.50$ for $\theta_0 \simeq 135^\circ$ and $N \approx 0.37$ for $\theta_0 \simeq 90^\circ$, when the measurement was performed during this initial stage, corresponding to $t^* < 25$ in figure 12(a). Notice that the method based on the actual droplet base radius was applied for the first steps, since the inflection point was defined here after the formation of the precursor film. After that short-time transient regime, one verifies that in all cases the droplet spreads in a quite similar way and with very small effects coming from the initial conditions.

doi:10.1088/1742-5468/2009/06/P06008
Figure 12. Log–log plot of the dimensionless actual (for $t^* < 25$) and apparent (for $t^* > 25$) droplet base radius, $R^* = R/R_0$, as a function of the reduced time, $t^*$, for three different initial conditions. The continuous line represents the fitting curve in the range $100 \leq t^* \leq 520$.

Tanner’s law [38] is well established in the complete wetting regime and any comparison between numerical and experimental results must be done in this regime, after the short initial period. Three-dimensional results reported by Raiskinmaki et al [22] and Dupuis and Yeomans [43] were carried out under partial wetting conditions. Although the power law, equation (17), was observed, it is related to the transitional regime succeeding the contact between the droplet and surface. Thus, the values of $N$ found by the authors were higher than expected, and about $N \approx 0.33$ by Raiskinmaki et al [22] and $N = 0.28$ by Dupuis and Yeomans [43] for the case of $\theta_0 \approx 180^\circ$. Furthermore, Raiskinmaki et al [22] reported a strong influence of the initial conditions, showing that, if $\theta_0 \to 90^\circ$, the simulated exponents tend towards the experimental results, for which $N = 1/10$. Despite the results presented in the present work being two-dimensional, it can be seen in figure 12 that, when $\theta_0 \to 90^\circ$, the slope of the curve $R^*$ versus $t^*$ at short times in the log–log plot tends to be similar to that slope at long times, and this was possibly the reason for that conclusion. Iwahara et al [41] provided two-dimensional results for $\theta_0 \approx 180^\circ$ under the complete wetting regime and obtained $N \approx 0.43$ and attributed such a discrepancy to the initial boundary conditions.

The results obtained in this section show that the initial conditions have a strong influence on the first steps of the droplet spreading, but such influence disappears after long times. This is consistent with the experimentally available results.

4. Conclusions

A lattice-Boltzmann method based on field mediators is proposed for modeling the fluid–solid interaction. The obtained results were shown to be consistent with results found
Modeling and simulation of the fluid–solid interaction in wetting

in the literature and provided additional information about the macroscopic behavior resulting from microscopic interactions around the contact line. It was observed that the static contact angle is strongly dependent on the interaction potential defined by the field strength and interaction length, and that a precursor film can appear under certain conditions. The contact angle hysteresis and its dependence on the initial conditions was simulated for simple rough surfaces. That pointed out the locality of the contact angle, which is an intrinsic property of the liquid–vapor–solid system. The spreading dynamics on solid surfaces was studied, showing a power law time behavior but with a remarkable difference in the exponent given by Tanner’s law. Finally, the influence of the initial conditions was shown to be important only in the first steps of the droplet spreading, becoming negligible after long times according to the experimental results.

Acknowledgments

Financial support was given by the National Petroleum Agency—PRH09-ANP/MME/MCT, CENPES/PETROBRAS, Financial Supporter of Studies and Projects—FINEP and the National Council for Scientific and Technological Development—CNPq—Brazil.

References

Modeling and simulation of the fluid–solid interaction in wetting

[27] Hardy W, The spreading of fluids on glass, 1919 Phil. Mag. 38 49
[39] Pérez E, Schäffer E and Steiner U, Spreading dynamics of polydimethylsiloxane drops: crossover from laplace to van der waals spreading, 2001 J. Colloid Interface Sci. 234 178

doi:10.1088/1742-5468/2009/06/P06008