An assessment of lattice Boltzmann methods for coating flow simulation

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The lattice Boltzmann method (LBM) is a rapidly developing alternative fluid flow simulation method based on kinetic theory rather than continuum mechanics. This paper presents a brief summary of a simple LBM model, which is subsequently used to simulate the spreading of a droplet on a surface with a wettability gradient. Results are compared with a corresponding simulation based on lubrication theory, and show that the LBM can capture the main physical effect (i.e. spreading) but its quantitative predictions of droplet shape are rather different from those found from the continuum-based simulation. The reasons for this difference are discussed, along with potential means of improving agreement.

1 Introduction

In recent years, a fluid flow simulation technique known as the lattice Boltzmann method (LBM) [1] has often been cited as a promising alternative to methods based on direct solution of the Navier-Stokes equations. The LBM is a non-continuum-based model developed from statistical mechanics, and simulates the dynamics of 'pseudo-particles' constrained to move and collide with each other on a structured lattice. As explained in more detail below, local values of macroscopic quantities such as velocity, density and pressure can be calculated from probability distribution functions, and it can be shown that the LBM reproduces the incompressible Navier-Stokes equations for small Knudsen and Mach numbers[†] [2]. Historically the LBM developed from lattice gas automata [3], driven by the desire to remove the statistical noise associated with such Boolean methods. Later the LBM was shown to be a special discretization of Boltzmann's equation [4,5].

From a mathematical point of view, the fact that the LBM's pseudo-particles only interact in a localized fashion means that there is no large matrix to invert, unlike in methods based on discretizations of global partial differential equations. This in turn means that the LBM is particularly good for simulating flows involving highly complex, multiply-connected domains – for example the flow through porous media like paper, or the flow over rough surfaces. Such simulations avoid the need for empirically-derived approximations such as Darcy's law. The numerical algorithm encapsulating the LBM is very simple, and this feature – combined with its localized nature – makes the LBM ideal for parallel programming to exploit modern large-scale computers. Indeed the method has been shown to yield very good scalability [6].

Advantages of the LBM from a physical point of view include the availability of several multiphase models, which can model interface break-up and coalescence and also interactions with chemically non-uniform surfaces. The method is also valid at smaller length scales than the Navier-Stokes equations, which makes it good for microfluidic simulations [7].

The LBM is a very rapidly growing field of research, with about 3 or 4 papers appearing in the literature every week. Much of the development of the method occurs in the physics community, and in many cases the driving motivation is to capture increasingly complex phenomena without necessarily testing the *quantitative* performance of the method. Simulations of coating flows are however of most use if they are able to make quantitatively accurate predictions of important flow features – for example the line speed at which a particular instability begins. The purpose of this paper is to examine the performance of a multiphase lattice Boltzmann method in the context of a problem of interest to the coating community,

[†] The Knudsen number, *Kn*, is the ratio of the molecular mean free path to the flow's characteristic length scale, while the Mach number, *Ma*, is the ratio of the flow's characteristic velocity scale to the speed of sound in the fluid.

namely the spreading of a droplet. Results are compared against corresponding simulations made using lubrication theory.

A very brief sketch of the LBM is given in the next section; the presentation is intended mainly to demonstrate how different the method is from conventional CFD techniques – more detailed and rigorous treatments are available in the cited literature. Section 3 then outlines the problem to be considered, with results and discussion following in section 4 and conclusions in section 5.

2 Outline of the lattice Boltzmann method

2.1 Foundation and discretization

The LBM stems from the Boltzmann equation,

$$\frac{\partial f}{\partial t} + \underline{\xi} \cdot \nabla f + \underline{F} \cdot \left[\frac{\partial f}{\partial \underline{\xi}} \right] = \Omega(f) \tag{1}$$

where $f(\underline{x},\underline{\xi},t)$ is the single-particle probability distribution function (PDF), representing the probability of finding a particle at position $\underline{x}$ with velocity $\underline{\xi}$ at time t, $\underline{F}(\underline{x},t)$ is an external force, and $\Omega(f)$ is the integral collision operator. The latter entity is often simplified using the Bhatnagar-Gross-Krook (BGK) single-relaxation-time model [8],

$$\Omega(f) = -\frac{1}{\lambda}\left(f - f^{eq}\right) \tag{2}$$

where λ is the relaxation time, and

$$f^{eq} = \frac{\rho}{2\pi RT} \exp\left[-(\underline{\xi} - \underline{u}) \cdot (\underline{\xi} - \underline{u}) / 2RT\right] \tag{3}$$

is a local Maxwellian distribution dependent on the specific gas constant, R, the temperature, T, and the local value of the macroscopic density, ρ, and velocity, $\underline{u}$. These macroscopic quantities are determined from moments of the PDF with respect to the molecular velocity, i.e.

$$\begin{aligned} \rho(\underline{x},t) &= \int f(\underline{x},\underline{\xi},t) d\underline{\xi} \\ \rho(\underline{x},t)u(\underline{x},t) &= \int \underline{\xi} f(\underline{x},\underline{\xi},t) d\underline{\xi} \end{aligned} \tag{4}$$

etc. Thus one can see that the seemingly very simple collision operator (2) is in fact highly non-linear due to the inter-dependence of ρ, $\underline{u}$, f, and f^{eq} in equations (3) and (4).

Temporal discretization of equation (1) is achieved by integrating over one time step, δt, and expanding as a Taylor series, keeping terms of $O(\delta t)$. Momentum space is discretized by determining the integrals in (4) through quadrature, [4,5] i.e.

$$\int \psi(\underline{\xi}) f(\underline{x},\underline{\xi},t) d\underline{\xi} = \sum_i W_i \psi(\underline{\xi}_i) f(\underline{x},\underline{\xi}_i,t) \tag{5}$$

where $\underline{\xi}_i$ is a set of discrete molecular velocities, and W_i corresponding weights. In the standard LBM, the $\underline{\xi}_i$ are coupled to the spatial domain by means of a structured lattice, with a lattice spacing, $\delta x = c\delta t$ (c being the lattice speed). Figure 1 shows a popular two-dimensional lattice structure, consisting of 3 speeds (0, c and $c\sqrt{2}$) and 9 discrete molecular velocities (the eight pictured plus a zero velocity).

Thus the spatial discretization is linked to that of the momentum space. The discretization process also introduces discrete distribution functions, f_i, each associated with a different lattice link. The f_i are the 'pseudo-particles' mentioned in the introduction. The evolution of f_i involves two stages:

- *Streaming:* each f_i is simultaneously propagated to the next lattice node in the direction of the velocity vector with which it is associated.
- *Colliding:* the f_i interact with each other at lattice nodes via the collision model (2).

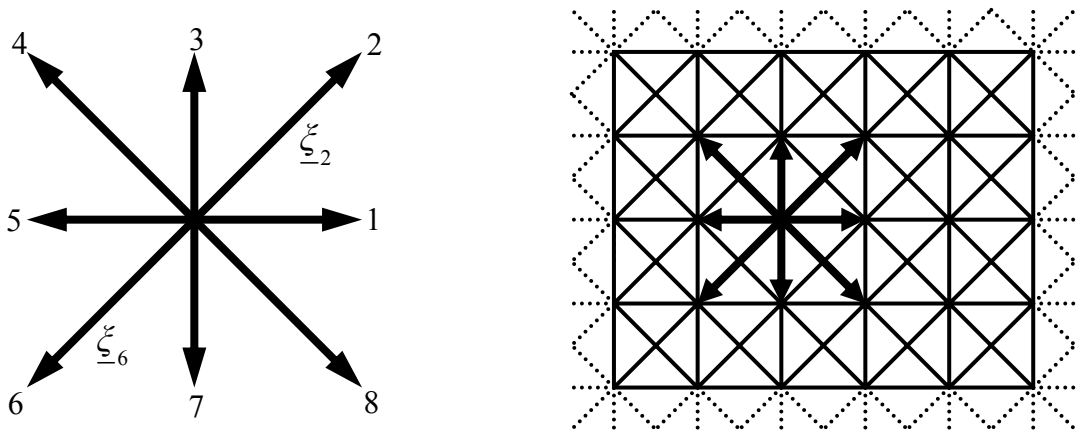

Figure 1: (*a*) discrete molecular velocities associated with each lattice node in the 'D2Q9' model; (*b*) how the discrete velocities form the lattice.

2.2 Multiphase model

There are essentially four multiphase models that incorporate different degrees of physics into the LBM [9-12]. The model used in this study is that of He *et al*. [13], which is an extension of the earlier work [12]. Using a mean-field approximation for intermolecular attractions [14] and including an exclusion-volume effect [15], He *et al.* [12,13] showed how the force in equation (1) can be reworked into a surface tension force, and how a stable numerical discretization of the system can be achieved by introducing an auxiliary distribution function associated with the pressure in the fluid. The lattice Boltzmann algorithm that arises from the above discretization and the introduction of the second distribution function is simply

$$f_i(\underline{x}+\underline{e}_i\delta t, t+\delta t) = f_i(\underline{x},t) - \frac{1}{\tau}\left[f_i(\underline{x},t) - f_i^{eq}(\underline{x},t)\right] - 2\widetilde{F}_i(\underline{u},\underline{e}_i,\phi) \tag{6}$$

$$g_i(\underline{x}+\underline{e}_i\delta t, t+\delta t) = g_i(\underline{x},t) - \frac{1}{\tau}\left[g_i(\underline{x},t) - g_i^{eq}(\underline{x},t)\right] - 2\widetilde{G}_i(\underline{u},\underline{e}_i,\rho) \tag{7}$$

where the f_i are PDFs associated with the 'index function', ϕ, which tracks the interface between the two phases, the g_i are PDFs associated with the fluid pressure and density, τ is the dimensionless relaxation time, and $\widetilde{F}_i$ and $\widetilde{G}_i$ are force terms resulting from the force term in equation (1). Notice that the right-hand sides of equations (6) and (7) form the collision stage of the algorithm, where the PDFs interact with each other to model molecular collisions, while the left-hand sides encapsulate the streaming stage, where the PDFs propagate along their links.

The moment equations (4) become:

$$\phi = \sum_{i=0}^{8} f_i \tag{8}$$

$$p = \sum_{i=0}^{8} g_i - \frac{1}{2}\underline{u}\cdot\nabla\psi(\rho)\delta t \tag{9}$$

$$\rho RT\underline{u} = \sum_{i=0}^{8}\underline{e}_i g_i + \frac{1}{2}RT(\underline{F}_s + \underline{G})\delta t \tag{10}$$

where ψ is a function dependent on the equation of state (i.e. the van der Waals' equation [14]), $\underline{G}$ is the body force due to gravity, and $\underline{F}_s$ is the surface tension force, which is dependent on the density gradient through the (diffuse) interface.

2.3 Boundary conditions

Obviously no fluid domain is infinite, so inevitably the lattice of figure 1(*b*) must terminate at some form of boundary. The presence of a boundary complicates the algorithm slightly, because at the streaming stage nodes adjacent to the boundary will be expecting information (i.e. values of f_i) from outside the fluid domain (e.g. from inside a solid wall). Hence one must obtain suitable values for this missing information. In the flow considered below, the two fluids are contained by four stationary smooth rigid walls aligned along the horizontal and vertical lattice directions. Three of these walls are in contact with only one fluid, and for these walls the simplest LBM boundary condition, the 'bounce-back', can be used effectively. This condition is implemented at wall nodes simply by reflecting each f_i and g_i propagated towards the wall back along the link upon which they arrived. More sophisticated boundary conditions are required to deal with moving walls (e.g. [16]) or with boundaries touching fluid-fluid interfaces, see the next section.

2.4 Wetting model

Cahn's theory of wetting [17] provides a one-dimensional relationship between the gradient of density at a solid surface and the equilibrium contact angle on that surface. This theory has been used as the basis of a successful wetting boundary condition [18] suitable for the 'free energy' multiphase LBM model [11]. However, this condition is difficult to implement in the multi-phase model used here because it results in an implicit equation for the index function, ϕ, which would require a complicated iterative procedure to be embedded within the otherwise simple LBM algorithm. A simpler and more convenient alternative is to adopt the 'surface affinity' idea of Iwahara *et al.* [19]. If ϕ_L and ϕ_G are the values of ϕ corresponding respectively to the liquid and gas phases of the van der Waals fluid, then the surface affinity is defined as

$$\chi = \frac{\phi - \overline{\phi}}{\phi_L - \overline{\phi}} \quad \text{where} \quad \overline{\phi} = \frac{1}{2}(\phi_L + \phi_G). \tag{11}$$

The interface has the profile [14]

$$\phi(z) = \overline{\phi} - \frac{1}{2}(\phi_L - \phi_G)\tanh\left(\frac{z - z_0}{\delta}\right) \tag{12}$$

where z is a coordinate normal to the interface, z_0 is the location of the 'middle' of the interface (i.e. where $\phi(z_0) = \overline{\phi}$), and δ is a parameter governing the thickness of the interface. Using this, the liquid-vapour surface tension can be calculated in terms of ϕ_L, ϕ_G and a free parameter, κ:

$$\sigma_{LG} = \kappa\int_{-\infty}^{\infty}\left(\frac{\partial\phi}{\partial z}\right)^2 dz = \frac{\kappa(\phi_L - \phi_G)^2}{4\delta}\int_{-1}^{1}(1-\chi^2)d\chi = \frac{\kappa(\phi_L - \phi_G)^2}{3\delta}. \tag{13}$$

Similar expressions can be found for the solid-liquid and solid-vapour interfacial tensions, and Young's equation then gives

$$\cos\theta_e = \chi(3 - \chi^2)/2 \tag{14}$$

for the equilibrium contact angle, θ_e. Thus variations in contact angle along the solid surface can readily be included via the surface affinity, which determines ϕ at the boundary via equation (11) with no need for messy iterative procedures.

3 Test problem

The problem to be tackled by both the above LBM and lubrication theory is simply the spreading of a two-dimensional 'droplet' – in other words a filament – along a solid surface with a constant gradient in wettability, i.e. a linear variation in contact angle. The initial cross-section of the filament is a parabolic profile,

$$y = H_0[1 - (x - x_0)^2 / R_0^2] \tag{15}$$

where H_0, and R_0 are the initial height and in-plane radius, and x_0 is the initial horizontal position of the filament's centre. The lubrication model used to simulate this flow is a simple adaptation of that presented by Gaskell *et al.* [20], in which the variable contact angle is incorporated through a disjoining pressure.

3.1 Physical parameters

The fluid properties used in the simulations are those of water, namely a density of 1000 kg m^{-3}, a kinematic viscosity of 10^{-6} m^2 s^{-1}, and a surface tension of 70 mN m^{-1}. The initial filament geometry was given by $R_0 = 10^{-3}\,\mathrm{m}$ and $H_0 = 6.4\times10^{-4}\,\mathrm{m}$, with the contact angle at x_0 being 30° and the gradient in θ_e being 2.1°/mm. All these parameters are very easy to include in the lubrication model.

3.2 Lattice Boltzmann parameters

Determination of the correct parameters for the LBM simulation is not as straightforward as for the lubrication model. For single-phase flows the LBM parameters can be adjusted fairly easily to match physical flow conditions such as the Reynolds number. However, in multiphase models there is a diffuse interface with a characteristic thickness to consider. If this typically very small length scale is to be captured properly (say by having at least four lattice lengths across the interface), the lattice will have to be very fine, which will substantially restrict the spatial and temporal extent of problems that can feasibly be tackled.

For example, suppose the interface thickness is $h = 10^{-8}\,\mathrm{m}$. If $\delta = 1.5$, to give 4 or 5 lattice lengths across the interface, the lattice spacing should be of order $h/\delta = 7\times10^{-9}\,\mathrm{m}$. This means that a 1mm drop (which is admittedly rather large for a drop) would require a lattice comprising over 20 billion nodes! Therefore on must compromise either on the size of the system to be modelled or the accuracy with which the interface thickness is captured. When one notes that the fluid viscosity, surface tension and body force are also linked to the lattice size and the choice of relaxation time, it becomes clear that correctly specifying the LBM parameters to match the 'real world' is no trivial undertaking.

4 Results And Discussion

In obtaining the results presented here, the interface thickness was made artificially large ($10^{-6}\,\mathrm{m}$) in order to model a 1mm filament using a feasible lattice size. Figure 2 shows the filament profiles predicted by lubrication theory. Note that in all the graphs shown here, the vertical scale is exaggerated by a factor of two. The lubrication results are as expected: the parabolic initial profile quickly spreads to form an approximately semi-circular shape, before the whole drop translates due to the variation in contact angle along the substrate.

The corresponding LBM results are shown in figure 3, which reveals that the LBM captures the same spreading and translating effects, but the shape and speed of the filament are somewhat different from those from lubrication theory. Figures 4 and 5 show direct comparisons of the two methods' predictions for the maximum height of the filament and the position of this maximum as a function of time. Again the same qualitative behaviour is seen, but there is a quantitative discrepancy in the spreading rates. Figures 4 and 5 also include data calculated by halving the interface thickness in the LBM. In practice this involves doubling the lattice and filament sizes in both dimensions, while keeping δ fixed. The results show that the choice of interface thickness/lattice size does influence the quantitative performance of the method.

One should not set too much store by the lubrication predictions of spreading rates, however, as these rates are known to be over-predictions due to the presence of a precursor film in the model. Of greater concern is the difference in shape of the filament – the LBM profiles are rather less squat than the corresponding lubrication profiles (this remains the case for thinner

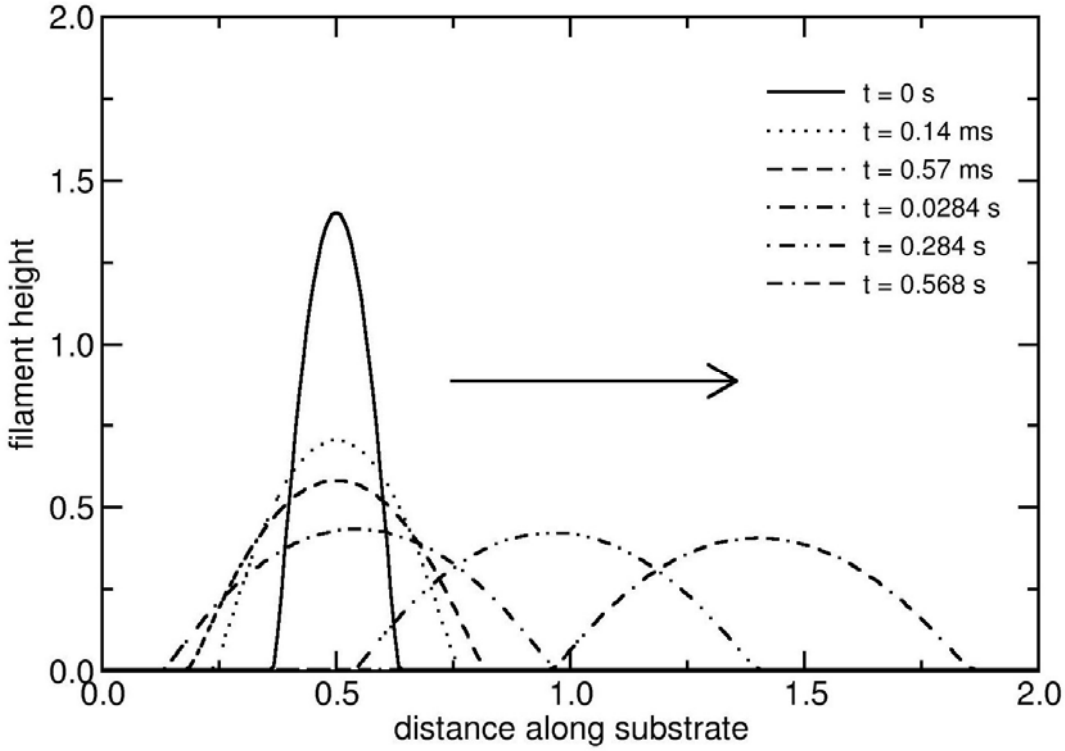

Figure 2: Filament profiles calculated using the lubrication model.

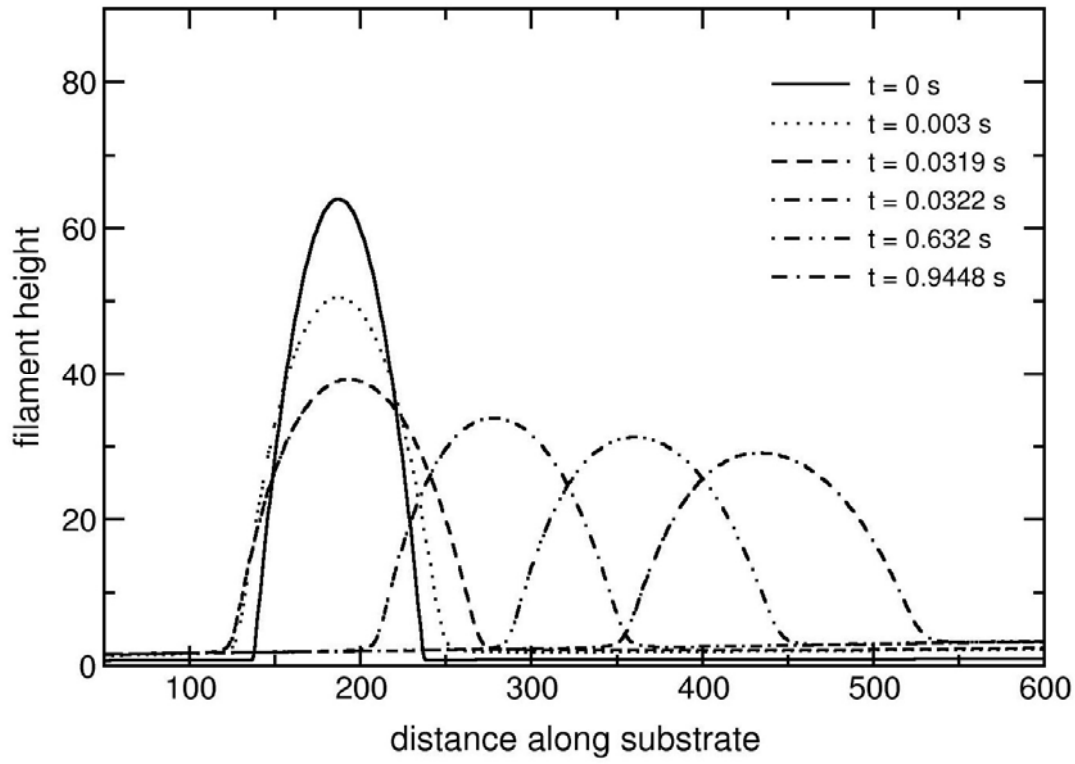

Figure 3: Filament profiles calculated using the LBM.

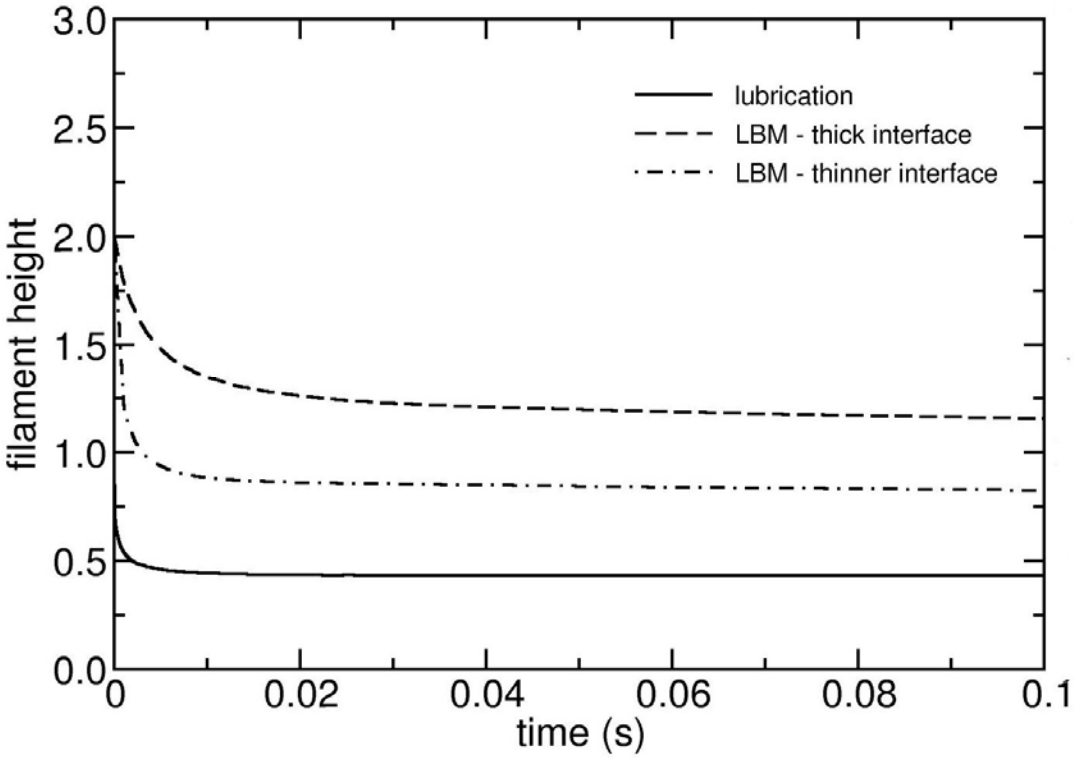

Figure 4: Comparison of the predictions of the maximum height of the filament as a function of time

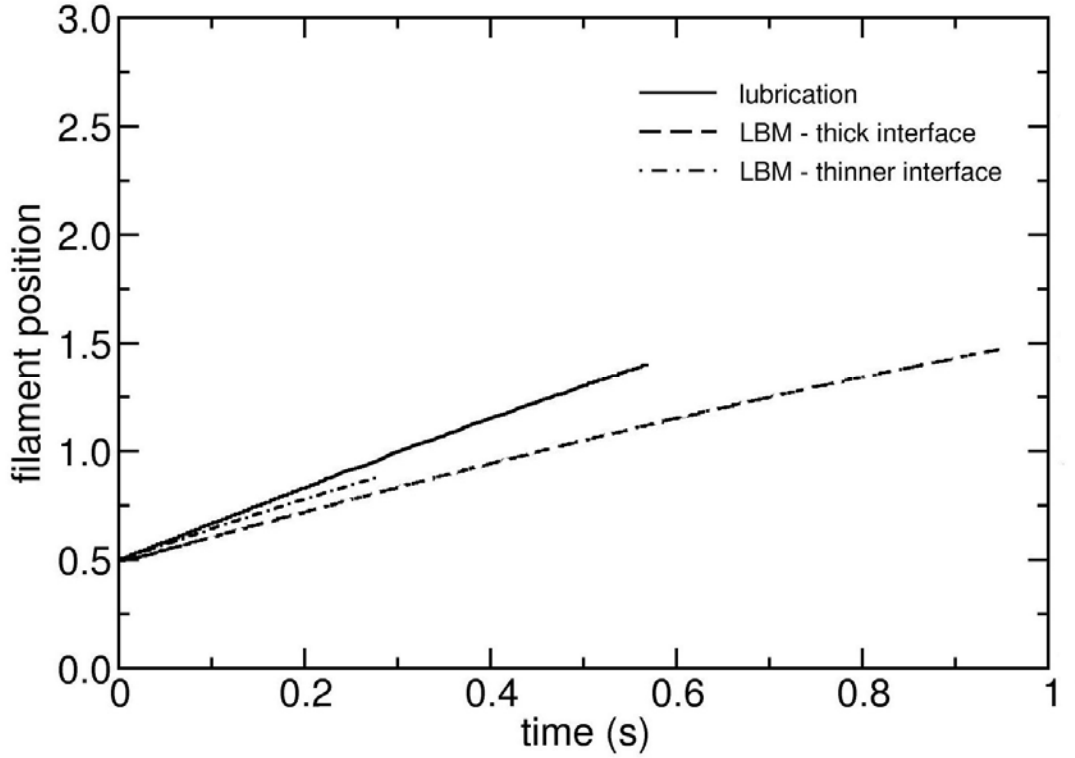

Figure 5: Comparison of the predictions of the position of the maximum height of the filament as a function of time

interfaces). This can be attributed to the fact that in the LBM the ambient fluid must be included in the simulation (in the lubrication simulations the ambient fluid is neglected). Moreover, the ratio of the fluid densities is rather limited, since instabilities can arise due to large density gradients across the interface. Using the present model, stable solutions could only be achieved with a density ratio of 20:1 or less. This is in fact the most serious shortcoming of the LBM as far as the simulation of coating flows is concerned, since a density ratio approaching 1000:1 is more appropriate to the liquid-air systems of interest.

Progress has been made in this area, however, with the development of a high density-ratio model by Inamuro *et al.* [21], but this has not yet been successfully exploited independently. Other shortcomings of the LBM include the limitations on the lattice size due to the requirement that the interface thickness should be properly represented. Again progress has been made through the development of local refinement techniques [22] and the use of non-uniform lattices [23]. Another major concern, the stability of the LBM, has likewise been the subject of some development, and it has been shown that adopting a multiple-relaxation-time collision model rather than the simple one in equation (2) can greatly improve this aspect of the method [24].

5 Conclusion

The lattice Boltzmann method is an alternative flow simulation method with the benefits of a simple algorithm, making parallel implementation attractive, and the ability to include smaller-scale physics than continuum-based simulation techniques. The method has been tested here on a simple droplet/filament spreading problem, and has been shown to capture the expected physical behaviour of the system. However, the predicted filament shapes are somewhat different from the corresponding predictions from lubrication theory. This is attributed to the necessary presence of an ambient fluid whose density cannot be reduced to a realistic value for a liquid-gas system due to stability restrictions. Achieving appropriate density ratios is obviously a major issue as far as coating flow simulation is concerned. While the LBM is perhaps not quite up to the challenge of accurate coating simulations just yet, it is a very rapidly developing technique that seems set to contribute to the whole spectrum of fluid mechanics.

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