Lattice Boltzmann method for phase-separating liquid-vapor systems

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Abstract. In this paper introduce a finite difference lattice Boltzmann model capable to describe the hydrodynamics and the phase separation of a two-dimensional liquid-vapor system. A numerical scheme involving flux limiter techniques is proposed to minimize spurious numerical effects. We report some simulation results of the spinodal decomposition process and we find evidence of two different growth regimes depending on the system viscosity.

INTRODUCTION

Lattice Boltzmann (LB) models approach physical phenomena in fluid systems using a phase-space discretized form of the Boltzmann equation [1, 2, 3, 4, 5]. LB models are widely used to investigate the complex behavior of single- or multi-component/phase fluid systems [3, 5]. A force term [6, 7, 8] is added to the discretized Boltzmann equations in order to model the liquid-vapor system.

The velocity-space discretized Boltzmann equations may be solved using a finite difference (FD) numerical scheme. In this contribution, we introduce two numerical schemes, namely, the first order upwind finite difference scheme and a higher order one which uses flux limiters [9, 10]. We refer here to a FDLB model in the two-dimensional space. The starting point is provided by the set of partial derivatives equations resulting from the discretization of the Boltzmann equation on a square lattice \( \mathcal{L} \) when the collision term is linearized using the BGK approximation [11]. In non-dimensional form, this set reads

\[
\partial_t f_i + e_{\beta} \partial_{\beta} f_i = \frac{1}{\chi c_R^2} f_i^{eq}(e_{\beta} - u_{\beta}) F_\beta - \frac{1}{\tau} (f_i - f_i^{eq}), \quad i = 0, 1, \ldots, \mathcal{N}
\]  

We use the following reference quantities for particle number density, temperature and speed to get the non-dimensional form (1) of the discretized Boltzmann equations for a van der Waals fluid: \( n_R = N_A/V_{mc} \), \( T_R = T_c \), \( c_R = \sqrt{k_B T_c/m} \). Here \( N_A \) is Avogadro’s number, \( V_{mc} \) is the molar volume at the critical point and \( T_c \) is the critical temperature. The dimensionless speed [12] is \( c = c_i/c_R = \sqrt{\theta/\chi} \), where \( \theta = T/T_R \) is the dimensionless temperature (the constant \( \chi \) equals 1/3 for the square lattice). If we take the system size \( L \) as the reference length, the non-dimensionalized lattice spacing is related to the number of lattice nodes \( \mathcal{N} \) through \( \delta s = L/N \).

The particle distribution functions \( f_i \equiv f_i(\mathbf{x}, t) \) are defined in the nodes \( \mathcal{L} \) of the square lattice \( \mathcal{L} \). In the D2Q9 model we use in this paper, \( \mathcal{N} = 8 \) and the velocities \( \mathbf{e}_i \) are [2, 3, 4, 5]

\[
\mathbf{e}_0 = 0, \quad \mathbf{e}_i = \left[ \cos \frac{\pi(i-1)}{2}, \sin \frac{\pi(i-1)}{2} \right] e, \quad (i = 1, \ldots, 4)
\]  

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The equilibrium distribution functions \( f^{eq}_i = f^{eq}_i(x,t) \) are expressed as series expansions of the Maxwellian distribution function, up to second order with respect to the local velocity \( u = u(x,t) \), whose Cartesian components are \( u_\beta \) [13]:

\[
f^{eq}_i = w_i n \left[ 1 + \frac{\mathbf{e}_i \cdot u}{\chi c^2} + \frac{(\mathbf{e}_i \cdot u)^2}{2 \chi c^4} - \frac{(u)^2}{2 \chi c^2} \right]
\]

where \( w_i \) are appropriate weight coefficients [13]. The local density \( n \), as well as the components of the local velocity which enter Eq. (3), are calculated from the distribution functions as follows:

\[
n = \sum_i f_i = \sum_i f^{eq}_i
\]

\[
u = \frac{1}{n} \sum_i f_i \nu_\beta = \frac{1}{n} \sum_i f^{eq}_i \nu_\beta
\]

The force term in Eqs. (1) is given by [14, 15]

\[
F_\beta = \frac{1}{n} \partial_\beta (p^i - p'^n) + \kappa \partial_\beta (\nabla^2 n)
\]

where

\[
p^i = \theta n
\]

and

\[
p'^n = \frac{3 \theta n}{3 - n} - \frac{9 n^2}{8}
\]

are the non-dimensionalized pressures of the ideal and the van der Waals fluid, respectively [12]. With the equation of state in the form (8), the critical point is located at \( \theta = 1 \) and \( n = 1 \). The parameter \( \kappa \) controls the surface tension [8].

The mass and momentum equations are recovered from Eqs. (1) after using the standard Chapman - Enskog procedure up to second order with respect to Knudsen number \( Kn = c \tau \). These equations read [4, 5, 8]

\[
\partial_t n + \partial_\beta (nu_\beta) = 0
\]

\[
\partial_t (nu_\alpha) + \partial_\beta (nu_\alpha u_\beta) = -\partial_\alpha p'^n + \kappa n \partial_\alpha (\nabla^2 n) + \nu \partial_\beta \left[ n \left( \partial_\alpha u_\beta + \partial_\beta u_\alpha \right) \right]
\]

where

\[
\nu = \chi c^2 \tau
\]

is the physical value of the kinematic viscosity [16]. The force term (6) allows to recover the Navier - Stokes equation (10) where the pressure \( p'^n \) appearing on the r.h.s. is subjected to the van der Waals equation of state (8).

**FINITE DIFFERENCE SCHEMES**

The set of phase space discretized equations (1) may be solved numerically by using an appropriate finite difference scheme defined on the lattice \( \mathcal{L} \). The first-order upwind scheme, is a good candidate because of its stability. When associated to the forward time stepping rule, this scheme gives the following updating rule for the distribution functions defined in node \( x \in \mathcal{L} \) [16]

\[
f_i(x,t + \delta t) = f_i(x,t) - \frac{c \delta t}{\delta x} \left[ f_i(x,t) - f_i(x - \delta x e_i/c, t) \right] + \delta t Q_i(x,t)
\]

where

\[
Q_i = Q_i(x,t) = \frac{1}{\theta} \left\{ \frac{1}{n(x,t)} \partial_\beta \left[ p^i(x,t) - p'^n(x,t) \right] + \kappa \partial_\beta \left[ \nabla^2 n(x,t) \right] \right\} f^{eq}_i(x,t) \left[ \nu_\beta - u_\beta (x,t) \right]
\]

\[
- \frac{1}{\tau} \left[ f_i(x,t) - f^{eq}_i(x,t) \right], \quad i = 0, 1, \ldots \mathcal{N}
\]

627
As discussed in Ref. [16], finite difference schemes introduce spurious numerical terms in the conservation equations. This happens because the real evolution equations recovered (up to second order in space and time) from the updating rules (12) are

$$\partial_t f_i + \phi \partial_x^2 f_i + e_{\beta} \partial_x e_{\beta} f_i - \psi \partial_x e_{\beta} e_{\gamma} f_i = Q_i, \quad i = 0, 1, \ldots, N$$  \hspace{1cm} (14)

where $\phi = \delta t / 2$ and $\psi = \delta s / 2c$. We get the following form of the conservation equations up to second order in the Knudsen number:

$$\partial_t n + \partial_x (n u_\beta) = (\psi - \phi) \partial_x \left[ \chi c^2 n \partial_x^2 + n u_\beta u_\beta \right]$$  \hspace{1cm} (15)

$$\partial_t (n u_\alpha) + \partial_x (n u_\beta u_\gamma) = -\partial_x p^w + \kappa n_\alpha (n u_\beta) + \nu_u \partial_x \left[ n (\partial_x u_\beta + \partial_x u_\alpha) \right] + \chi c^2 (\psi - \phi) \partial_x \left[ \partial_x (n u_\beta) + u_\beta \partial_x n + u_\alpha \partial_x n \right]$$  \hspace{1cm} (16)

Thus, the finite difference scheme introduces spurious terms, depending on the quantity $(\psi - \phi)$, in both the conservation equations (compare with Eqs. (9)-(10)), while the physical value (11) of the kinematic viscosity is replaced by the apparent value [16]

$$\nu_{ap} = \chi c^2 (\tau + \psi)$$  \hspace{1cm} (17)

Small values of the relaxation parameter $(\tau < 10^{-3})$ are characteristic for large systems where the Knudsen number is negligible. It is well known that the continuum hypothesis and the Navier - Stokes equation are valid only for small values of the Knudsen number $Kn$ [17]. For these values of $\tau$, a huge computational effort is required to achieve a negligible numerical viscosity term $(\chi c^2 \psi)$ in Eq. (17) when doing 2D or 3D simulations using the first order upwind FDLB model. Higher order flux limiter schemes provide a possibility to overcome this problem.

We consider the characteristic line on the square lattice involving the distribution functions $f_j(x, t)$ [18, 19]. For convenience, we denote $f_{i,j}^k$ the value of $f_j$ in node $j$ at time $t = k \delta t$ and rewrite the updating rule (12) in a conservative form using two fluxes [9, 20, 18, 19]:

$$f_{i,j}^{k+1} = f_{i,j}^k - \frac{c \delta t}{\delta s} \left[ F_{i,j+1/2}^k - F_{i,j-1/2}^k \right] + \delta t Q_{i,j}^k$$  \hspace{1cm} (18)

where

$$F_{i,j+1/2}^k = f_{i,j}^k + \frac{1}{2} \left[ 1 - \frac{c \delta t}{\delta s} \right] \left[ f_{i,j+1}^k - f_{i,j}^k \right] \Psi(\Theta_{i,j}^n)$$  \hspace{1cm} (19)

and

$$F_{i,j-1/2}^n = F_{i,j(n-1)+1/2}^n$$  \hspace{1cm} (20)

The flux limiter $\Psi(\Theta_{i,j}^n)$ introduced in (19) is expressed using the smoothness

$$\Theta_{i,j}^n = \frac{f_{i,j}^n - f_{i,j-1}^n}{f_{i,j+1}^n - f_{i,j}^n}$$  \hspace{1cm} (21)

In particular, the first order upwind scheme is recovered when $\Psi(\Theta_{i,j}^n) = 0$. LB simulations reported in this paper were done using the Monitorized Central Difference (MCD) limiter [9]

$$\Psi(\Theta_{i,j}^n) = \begin{cases} 0 & , \Theta_{i,j}^n \leq 0 \\ 2 \Theta_{i,j}^n & , 0 \leq \Theta_{i,j}^n \leq \frac{1}{3} \\ \frac{1 + \Theta_{i,j}^n}{2} & , \frac{1}{3} \leq \Theta_{i,j}^n \leq 3 \\ 2 & , 3 \leq \Theta_{i,j}^n \end{cases}$$  \hspace{1cm} (22)

LB simulations of diffusion phenomena done using flux limiter schemes suggest a second order dependence of the value $\psi$ on the lattice spacing $\delta s$ [21].

628
FIGURE 1. Evolution of the three measures of domains size $R_1$, $R_2$, $R_3$, recovered for $\tau = 10^{-4}$, $\beta = 0.5$ with: (a) – the flux limiter scheme; (b) – the upwind scheme. The $R$’s are measured in lattice spacings and $R_1$ has been multiplied by 4,000,000 to be shown in the same plot. Straight lines have the slope 2/3 (a) and 1/2 (b).

SIMULATION RESULTS

We used a square lattice with $N \times N$ nodes ($N = 1024$), $\delta s = 1/256$, and $\delta t = 10^{-5}$. The fluid system was quenched to the temperature $\theta = 0.79$ where the coexisting densities are $n_{\text{liquid}} = 1.956$ and $n_{\text{vapor}} = 0.226$. Each simulation was started with small fluctuations (0.1%) in the density about a mean value $\hat{n}$ that was either symmetric ($\hat{n} = 1.09$, liquid fraction $\hat{\beta} = 0.5$) or slightly off-symmetric ($\hat{n} = 1.0$, $\hat{\beta} = 0.45$). The parameter $\kappa$ controlling the surface tension was set to $5 \times 10^{-6}$ to have an interface thickness of $\sim 6$ lattice spacings. The viscosity was varied by changing $\tau$.

We implemented the upwind and the flux limiter schemes and compared results when $\tau = 10^{-4}$. For this value of $\tau$, the spurious numerical contribution of the upwind scheme is larger than the physical one. The numerical viscosity is significantly reduced when the flux limiter scheme is considered instead. We used also the value $\tau = 10^{-5}$ with the flux limiter scheme to access a higher viscosity regime.

In order to have different and independent tools to estimate the domains size we used the following quantities: $R_1(t)$, the inverse of the length of the interfaces of domains, measured by counting lattice points where the order parameter $\rho(x,t) = n(x,t) - \hat{n}$ is such that $\rho(x,t) \rho(x',t) < 0$; $R_2(t)$, the inverse of the first moment of the spherically averaged structure factor

$$R_2(t) = \pi \int \frac{\mathcal{C}(k,t)dk}{kC(k,t)dk},$$

where $k = |k|$ is the modulus of the wave vector in Fourier space, and

$$C(k,t) = \langle \hat{\rho}(k,t)\hat{\rho}(-k,t) \rangle$$

with $\hat{\rho}(k,t)$ the spatial Fourier transform of the order parameter $\rho(x,t)$. The angle brackets denote an average over a shell in $k$ space at fixed $k$. The last quantity $R_3(t)$ is defined as the inverse of the first moment of the spherically averaged structure factor of the fluid velocity

$$R_3(t) = \pi \int \frac{\mathcal{C}_u(k,t)dk}{k\mathcal{C}_u(k,t)dk},$$

with $\mathcal{C}_u(k,t) = \langle |\mathbf{u}(k)|^2 \rangle$. In all the figures $R_1$ was multiplied by a constant factor to be shown in the same plot with $R_2$ and $R_3$.

In Fig. 1a we present the three measures of domains size as function of time for the case $\tau = 10^{-4}$ with flux limiter scheme, and symmetric composition. It is interesting to note that $R_2$ and $R_3$ have the same trend, with a similar
prefactor. After a swift initial growth the evolution of all quantities suggests the existence of the growth exponent 2/3. This is in accordance with previous studies on symmetric liquid-vapor systems at low viscosity [22] when hydrodynamic flow is operating. In this regime hydrodynamics is the mechanism to get domains circular since the flow is driven by the difference in Laplace pressure between points of different curvature on the boundary of domains. This remark is confirmed when looking at configurations of the density \( n \). In Fig. 2 we show contour plots of a part of the whole system at consecutive times. The vapor bubble in the down left corner at \( t = 12 \), while evaporating, is rounded by the flow as it can be seen by comparing it with the shape at \( t = 15 \).

In the case with the upwind scheme the estimation of the growth exponent is more difficult since data are noisy (Fig. 1b). However, the late regime is characterized by an exponent consistent with the value 1/2. We suppose that this behavior is due to spurious terms in the macroscopic equations that are considerably larger when using the upwind scheme than in the case with flux limiter. This is confirmed by the analysis of the velocity fields in the two cases [23].

In Fig. 3 we plot the three measures of domains size as function of time for the case \( \tau = 10^{-3} \) with symmetric composition. These results were obtained using the flux limiter scheme. All quantities suggest the existence of the growth exponent 1/2. This is in accordance with previous studies on liquid-vapor systems at high viscosity [22] at symmetric composition when growth is expected to be described by the Allen-Cahn theory of interfaces dynamics which gives an exponent 1/2 [24] and hydrodynamics is not operating.
CONCLUSIONS

The correct choice of the numerical scheme is essential to recover the real physics of a fluid system subjected to LB simulations. In the case of a liquid-vapor system we have seen that simulation results exhibit significant changes when the numerical contribution of the finite difference scheme to the apparent value of the transport coefficients becomes comparable with the expected physical value. The numerical contribution of the first order upwind scheme is linearly dependent on the lattice spacing $\delta$ and switches to an higher order for the flux limiter scheme. Spurious velocities at interfaces can be considerably damped and very low viscosity systems can be simulated preserving numerical stability. Our code takes 6 hours to perform $10^5$ algorithm steps by using 32 Xeon 3.055 GHz processors on the IBM Linux Cluster 1350 at CINECA [25] with Myrinet IPC network and the Portable Extensible Toolkit for Scientific Computation (PETSc 2.1.6) developed at Argonne National Laboratory, Argonne, Illinois [26].

The model allowed to clarify the picture of phase separation in liquid-vapor system. We found that the growth exponent depends on the apparent fluid viscosity, which strongly depends on the numerical scheme used in the Lattice Boltzmann model. When liquid and vapor are present in the same amount, the growth exponent is $2/3$ and $1/2$ at low and high viscosity, respectively.

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