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PHASE SEPARATION OF BINARY FLUIDS WITH DYNAMICAL TEMPERATURE

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ABSTRACT

Thermal phase separation process, which follows a sudden quench in the coexistence region, is considered for binary fluid mixtures. It is studied applying a new version of an hybrid lattice Boltzmann model, where non-ideal terms of the pressure tensor, which takes into account thermal and concentration gradient contribution, are included as a body force in the LBM equations, used to solve the Navier-Stokes equations. The equations for concentration and temperature are solved using a finite difference scheme. Domains are observed to order preferably with interfaces parallel to the cold walls and this behavior is more pronounced at higher viscosities. An intermediate regime with two characteristics scales for the domain size, one close to the walls and the other in the middle of the system, has been also observed.

Key Words: *Lattice Boltzmann Methods; Phase Separation*

1. INTRODUCTION

The process of phase separation for binary fluids aroused a lot of interest in the scientific community [1] and many important results have been obtained in numerical simulations for different physical system and by different methods [2]. The importance of these processes is due to the innumerable physical phenomena in which they appear and to the theoretical frameworks necessary to explain them. On the other hand, the most part of the studies was made for systems whose temperature is assumed constant. In recent years there was an increasing interest to the study of the processes with a non constant temperature, due to the great variety of phenomena in which a heat flow occurs, like boiling processes and droplet motion under thermal gradient [3] or phase separation [4,5]. Experimental studies of phase separation of polymer mixtures [6] suggest the possibility of controlling the morphology of the coarsening domains by exploiting thermal diffusion properties and proper heat sources.

In this paper we present some results about the phase separation of a binary fluid mixture with non homogeneous temperature and the system quenched by contact with cold walls. We use a theoretical

framework presented in [7], in which the set of thermo-hydrodynamic equations is written for fluids with entropy and internal energy functionals including gradient terms. A generalized Gibbs-Duhem relation, that takes into account the presence of interface, allows to obtain a pressure tensor with explicit contributions from thermal gradient. We focused our studies on the morphology of the domains and on their growth dynamics, showing how ordered lamellar patterns result perpendicular to the direction of thermal gradients while interfaces assume more circular shapes when the viscosity is decreased. In an intermediate temporal regime a phase separation with two characteristic length, one close to the cold walls and the other in the central part of the system, has been observed.

The set of thermo-hydrodynamics equations, used to study these processes, are solved using a hybrid lattice Boltzmann (LB) approach, in which the continuity and the Navier-Stokes equations are simulated by using a LB algorithm (with a forcing term for non-ideal contributions), while the equations for concentration and energy are implemented by using a finite-difference scheme [8]. This is a convenient tool for studying thermal phase separation from a mesoscopic point of view. The paper is organized as follows. In the next section a brief description of the hybrid LB algorithm is presented and in the successive section some numerical results are shown. Some conclusions follow in the last section.

2. THE MODEL

For a binary mixture the conservation equations of mass, momentum and energy, expressed in terms of the total density n and concentration difference φ, take the form

(1) $$\partial_t n + \partial_\alpha (n u_\alpha) = 0,$$

(2) $$\partial_t (n u_\beta) + \partial_\alpha (n u_\alpha u_\beta) = -\partial_\alpha (P_{\alpha\beta} - \sigma_{\alpha\beta}),$$

(3) $$\partial_t \varphi = -\partial_\alpha (\varphi u_\alpha) - 2\partial_\alpha J^d_\alpha,$$

(4) $$\partial_t \hat{e} = \partial_\alpha (\hat{e} u_\alpha) - (P_{\alpha\beta} - \sigma_{\alpha\beta})\partial_\alpha u_\beta - \partial_\alpha J^q_\alpha,$$

where u is the velocity field, $\hat{e}$ is the internal energy of the mixture, $P_{\alpha\beta}$ is the reversible stress tensor, $\sigma_{\alpha\beta} = \eta(\partial_\alpha u_\beta + \partial_\beta u_\alpha) + (\xi - 2\eta/d)\delta_{\alpha\beta}\partial_\gamma u_\gamma$ is the dissipative stress tensor. J^d and J^q are the diffusive and heat currents, respectively given by $J^d = -L_{11}\overrightarrow{\nabla}(\frac{\mu}{T}) + L_{12}\overrightarrow{\nabla}(\frac{1}{T})$ and $J^q = -L_{21}\overrightarrow{\nabla}(\frac{\mu}{T}) + L_{22}\overrightarrow{\nabla}(\frac{1}{T})$, where μ is the chemical potential and T is the temperature. The quantities L_{ij} are phenomenological coefficients given by $L_{11} = Tl_{11}$ and $L_{22} = Tl_{22}$, with l_{11} and l_{22} diffusion and heat conductivity constants, respectively. Moreover η is the shear viscosity, ξ is the bulk viscosity and the Greek indexes are related to the cartesian axes.

These equations describe the dynamics of the system with non homogeneous temperature. The thermodynamics of the mixture is fixed by the free energy

(5)
$$F = \int d\mathbf{r} \left(AT + \frac{n\lambda}{4}(1 - \frac{\varphi^2}{n^2}) - k_B T[n \ln n - \frac{n+\varphi}{2}\ln(\frac{n+\varphi}{2}) - \frac{n-\varphi}{2}\ln(\frac{n-\varphi}{2})] + \frac{1}{2}\kappa|\nabla\varphi|^2 \right),$$

where A is a constant, k_B is the Boltzmann constant and κ weights the interface contribution. From the above expression the chemical potential and the pressure tensor can be derived [11]. Moreover the constant λ is given by $\lambda = T_c/2$, where T_c is the critical temperature of the mixture.

To solve numerically the Eqs. (1)-(4), we use an hybrid approach in which a LB scheme solves Eqs. (1) and (2) and a finite difference scheme solves the Eqs. (3) and (4). The LB scheme is built starting from a two dimensional square lattice where each site $\mathbf{r}$ is connected to the first and second neighbors. Horizontal and vertical links have length Δx, diagonal links $\sqrt{2}\Delta x$ and Δt is the simulation time step. On each site nine velocity vectors $\mathbf{e}_i$ are defined, according to the D_2Q_9 scheme. A set of distribution function $\{f_i(\mathbf{r}, t)\}$, whose evolution equation is

(6) $$f_i(\mathbf{r} + \mathbf{e}_i\Delta t, t + \Delta t) - f_i(\mathbf{r}, t) = -\frac{\Delta t}{\tau}[f_i(\mathbf{r}, t) - f_i^{eq}(\mathbf{r}, t)] + \Delta t F_i,$$

is defined in each site. f_i^{eq} is the equilibrium distribution function, τ is a relaxation parameter related to the viscosity and F_i is a forcing term depending on the concentration through the chemical potential

and giving in the continuum by $\mathbf{F} = -\varphi\nabla\mu$. The mass and momentum are defined by

$$n = \sum_i f_i, \qquad n\mathbf{u} = \sum_i f_i \mathbf{e}_i + \frac{1}{2}\mathbf{F}\Delta t. \tag{7}$$

By using a Chapman-Enskog expansion and conservation laws for mass and momentum, the continuum equations can be recovered. The finite difference scheme consists of an explicit Euler algorithm that solves the Eqs. (3) and (4) implementing in two successive time steps the convective and the diffusive term, respectively. In the convective part the velocity comes from the solution of the LB equation and the gradient of the concentration is calculated by using a backward or a forward scheme if the velocity is positive or negative, respectively. Moreover for both the equations (3) and (4), the space and time steps are the same of the ones used in the LB scheme. This approach allows to have a better numerical stability.

3. RESULTS AND CONCLUSIONS

In this section we present some results obtained for a phase separation process in absence of hydrodynamic with symmetric composition. The system, initially in a disordered state above T_c, is quenched by contact with cold walls. The effects of the temperature evolution on the morphology of phase separation are shown on the left in Figure 1, for a system at very high viscosity. We used a lattice size $L_x \times L_y = 512 \times 512$ with $\Delta x = \Delta t = 1$ and $\Omega = 3/5$, where $\Omega = T/T_c$. In Figure 1 the cold walls are placed at the bottom and top sides at the same temperature. Moreover we set $\gamma = l_{22}/l_{11} = 1/10$ and $l_{12} = l_{21} = 0$. The bulk properties are controlled via the coefficients $a = k_B T/n - \lambda/2n$ and $b = k_B T/3n^3$, obtained by a φ^4 approximation of (5). At time $t = 250$ of the simulation, domains with well-defined interface are observed close to the walls while in the central region the system is still in the disordered phase. From time $t = 1000$ a stack of domains perpendicular to the temperature gradient can be observed in all the system, except close to the walls, where we imposed the neutral wetting condition, which means that $\overrightarrow{a} \cdot \overrightarrow{\nabla}\varphi|_{y=0,L_y} = 0$. The quantity $\overrightarrow{a}$ is the inward normal unit vector at the boundaries. The corresponding evolution of the temperature across the system is shown on the

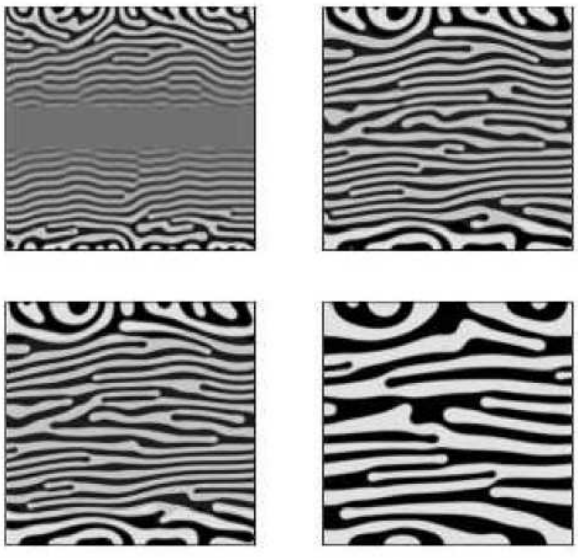

FIGURE 1. On the left a contour plot of φ at consecutive times (from the top left to the bottom right $t = 250, 1000, 1250, 5500$) during a phase separation in a system of size 512×512, with $\Omega = 3/5$. The mixture composition is symmetric

right in Figure 1. We also studied the phase separation with dynamical temperature at lower viscosity. In Figure 3 we show what happens at the value $\tau = 1$ in Eq. (6), on the same lattice and with $\Omega = 4/5$. During the phase separation one can observe two scales that characterize the growth of the domains in the bulk and close to the walls, where a more spherical shape can be observed. Further results have been obtained with asymmetric composition, in which the regular lamellar pattern obtained without hydrodynamics is substituted by circular domains that increase their size with different evolution and morphology depending on the value of viscosity.

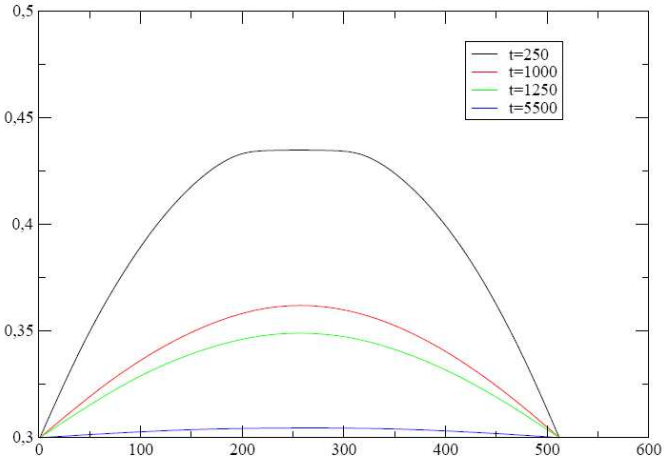

FIGURE 2. On the left the temperature profile across the system averaged along the x-direction, during a phase separation in a system of size 512×512, with $\Omega = 3/5$. The mixture composition is symmetric.

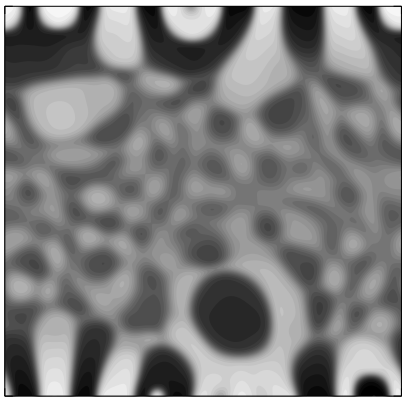

FIGURE 3. Contour plot of φ during a phase separation at $\tau = 1$ with $\Omega = 4/5$, in a system of size 512×512.

In conclusion we studied the phase separation process for a binary fluid where also the temperature is a dynamical quantity with a hybrid LB approach. We found that the morphology of the domains strongly depends on thermal diffusivity properties.

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