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Simulation of Ternary Fluid Mixtures Separation by Phase-Field Free Energy LBM

Abstract. This article reviews the mathematical and computer modeling of the process of ternary fluid mixture separation by free energy based phase field Lattice Boltzmann equations method. The process under study is considered in a limited area having the shape of a rectangle. Three different sets of fluid components with different structures are specified. The mathematical model constructed to describe this process is based on the Navier-Stokes equation for an incompressible fluid and the Cahn-Hilliard equation. The numerical model is built on the basis of LBM using the D2Q9 model. Numerical experiments were performed for two scenarios: (1) – investigate the model without gravity, in order to determine the patterns of the surface tension effect and (2) – investigate the model with gravity force. Numerical results showed a spinodal separation depending on the initial fractions of fluid concentrations. The results obtained determine the adequacy of the constructed model for a three-component fluid.

Key words. Three-component fluid, fluid mixtures separation, Cahn-Hilliard equation, free energy, lattice Boltzmann method.

Introduction

The study of multiphase and multicomponent flows dynamics is primarily necessary because they are often found in nature, and also take place in industrial and production processes, which requires a detailed study of a number of engineering problems. As an application example of numerical simulation of multiphase and multicomponent fluid flows, one can note the oil and gas production, the chemical processing of raw materials, as well as the steam-water mixture flows in boilers and condensers.

Various models can be used to model multiphase and multicomponent fluid flows [1-4]. Depending on the thickness of the transition layer between the phases, two main approaches can be distinguished: sharp interface models (transition layer between phases has zero thickness) and diffuse interface models (transition layer between the phases has a finite thickness). In our paper, we use the second approach. Van der Waals was the first to consider the transition layer between phases as a layer of finite thickness [5]. Currently, the Cahn-Hilliard approach [6] is widely used to describe the diffuse interface models.

This paper presents a mathematical model of incompressible three-component fluid flow using the phase field method based on the solution of the complete Navier-Stokes equation and the Cahn-Hilliard convective equation. The numerical model is based on free energy LBM using the D2Q9 scheme. The accuracy and efficiency of the existing method have been tested on the basis of solving a number of problems. The results obtained determine the correctness of the constructed model for a three-component fluid.

Problem statement

The process under study is considered in a limited area having the shape of a rectangle with dimensions $[0, L] \times [0, L]$ (Figure1). In this area there are three fluid components with density ρ_1, ρ_2 and ρ_3, the ratio of which is: $\rho_1 > \rho_2 > \rho_3$. A less dense fluid is indicated in blue, a medium density fluid in green, and a denser fluid in red.

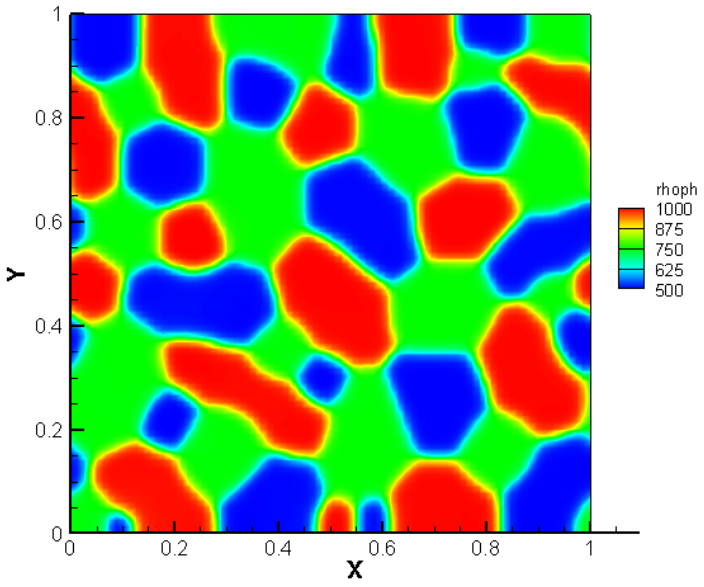


Figure 1 – Scheme of the computational domain

The mathematical model of the process includes the continuity equation, the momentum equation for the mixture and the Cahn-Hilliard convective equation:

$$\begin{gathered} \nabla \cdot \vec{u} = 0, \\ \frac{\partial(\rho\vec{u})}{\partial t} + \nabla(\rho\vec{u}\vec{u}) = \\ = -\nabla p + \nabla[\eta(\nabla\vec{u} + \nabla\vec{u}^T)] + F_s + \vec{F}_b, \\ \frac{\partial(c_i)}{\partial t} + \nabla(c_i\vec{u}) = \nabla(M_i\nabla\mu_i), i = 1,2,3 \end{gathered} \quad (1)$$

where $\vec{u}$ are the velocity components, p is the pressure, ρ is the density, η is the dynamic viscosity, c_i is the phase field for the fluid components: $c_1 + c_2 + c_3 = 1$, $\vec{g}$ is the gravitational acceleration, M_i is the mobility coefficient, μ_i is the chemical potential, $\vec{F} = F_s + \vec{F}_b = \sum_{i=1}^{3} \mu_i \nabla c_i + \rho\vec{g}$ is the total force of surface tension and gravity.

For a system of a multicomponent medium, the Landau free energy functional F can be determined based on the concentrations of fluids as follows [7]:

$$F(c, \nabla c) = \int \left[F_0(c) + \sum_{i,j=1}^{3} \frac{\lambda_{ij}}{2} \nabla c_i c_j \right] d\Omega$$

where $F_0(\mathrm{c}) = \sum_{i,j=1}^{3} \beta_{ij}[g(c_i) - g(c_j) - g(c_i + c_j)]$ is the bulk free energy, c – (c_1, c_2, c_3) is the phase variable of fluid components, $g(c) = c^2(1-c)^2$, $\beta_{ij} = \frac{3}{D}\sigma_{ij}$ and $\lambda_{ij} = -\frac{3D}{4}\sigma_{ij}$ are the constants, where σ_{ij} is the surface tension between the fluids and D is the thickness of the transition layer between fluids.

The variation of the free energy function F with respect to the concentration fractions c of fluids yields the chemical potential μ_i for component i as

$$\mu_1 = 2\beta_{11}(-28c_1^3 + 18c_1^2 - 2c_1) + 2\beta_{12}(-4c_2^3 - 12c_1^2c_2 - 12c_1c_2^2 + 6c_2^2 + 12c_1c_2 - 2c_2) + 2\beta_{13}(-4c_3^3 - 12c_1^2c_3 - 12c_1c_3^2 + 6c_3^2 + 12c_1c_3 - 2c_3) - (\lambda_{11}\nabla^2c_1 + \lambda_{12}\nabla^2c_2 + \lambda_{13}\nabla^2c_3),$$

$$\mu_2 = 2\beta_{21}(-4c_1^3 - 12c_1^2c_2 - 12c_1c_2^2 + 6c_1^2 + 12c_1c_2 - 2c_1) + 2\beta_{22}(-28c_2^3 + 18c_2^2 - 2c_2) + 2\beta_{23}(-4c_3^3 - 12c_2^2c_3 - 12c_2c_3^2 + 6c_3^2 + 12c_1c_3 - 2c_3) - (\lambda_{21}\nabla^2c_1 + \lambda_{22}\nabla^2c_2 + \lambda_{23}\nabla^2c_3),$$

$$\mu_3 = 2\beta_{31}(-4c_1^3 - 12c_1^2c_3 - 12c_1c_3^2 + 6c_1^2 + 12c_1c_3 - 2c_1) + 2\beta_{22}(-4c_2^3 - 12c_2^2c_3 - 12c_1c_3^2 + 6c_2^2 + 12c_2c_3 - 2c_1) + 2\beta_{33}(-28c_3^3 + 18c_3^2 - 2c_3) - (\lambda_{31}\nabla^2c_1 + \lambda_{32}\nabla^2c_2 + \lambda_{33}\nabla^2c_3).$$

We substitute the above chemical potential μ_i for component i into the equation (1), as a result, the system will be complete. The system of equations (1) has the following initial conditions:

$$u = v = 0,$$

$$c_1(\vec{x}, 0) = \bar{c}_1 + \alpha \times rand(\vec{x})$$

$$c_2(\vec{x}, 0) = \bar{c}_2 + \alpha \times rand(\vec{x})$$

$$c_3(\vec{x}, 0) = 1 - c_1(\vec{x}, 0) - c_2(\vec{x}, 0)$$

Boundary conditions:

On the bottom wall at $y = 0$:

$$u = v = 0, \quad \frac{\partial c_1}{\partial y} = \frac{\partial c_2}{\partial y} = \frac{\partial c_3}{\partial y} = 0.$$

On the side walls at $x = 0, L$:

for u, v, c_1, c_2, c_3 – periodic boundary conditions.

On the bottom wall at $y = L$:

$$u = v = 0, \quad \frac{\partial c_1}{\partial y} = \frac{\partial c_2}{\partial y} = \frac{\partial c_3}{\partial y} = 0.$$

Numerical method

The numerical solution of this model is based on the D2Q9 scheme of the lattice Boltzmann equations method. The lattice Boltzmann equation in the Batnagar-Gross-Krook (BGK) approximation is written as follows:

$$f_i(\vec{x} + \vec{e_i}\Delta t, t + \Delta t) - f_i(\vec{x}, t) = \Delta t\left[-\frac{f_i(\vec{x}, t) - f_i^{eq}(\vec{x}, t)}{\tau_f} + F_i\right]$$

$$g_i^m(\vec{x} + \vec{e_i}\Delta t, t + \Delta t) - g_i^m(\vec{x}, t) = \frac{\Delta t}{\tau_m}[g_i^m(\vec{x}, t) - g_i^{m,eq}(\vec{x}, t)]$$

where $m = 1,2,3$ – fluid components, f_i, g_i^m – velocity and phase field distribution functions, e_i – discrete lattice velocity, $\tau_f = \frac{1}{2} + c_1\left(\tau_1 - \frac{1}{2}\right) + c_2\left(\tau_2 - \frac{1}{2}\right) + (1 - c_1 - c_2)\left(\tau_3 - \frac{1}{2}\right), \tau_m = 0.8$ – relaxation times, F_i – force component, Δt – lattice time step, $f_i^{eq}, g_i^{m,eq}$ – equilibrium distribution functions for velocity field and phase field, respectively.

The equilibrium distribution functions are determined by the following formulas [8]

$$f_i^{eq} = \begin{cases} \rho - \sum_{i \neq 0} f_i^{eq}, i = 0 \\ \omega_i \rho\left(1 + \sum_{m=1}^{3} \frac{c_m \mu_m}{\rho c_s^2} + \frac{e_{i\alpha}u_\alpha}{c_s^2} + \frac{u_\alpha u_\beta\left(e_{i\alpha}e_{i\beta} - c_s^2\sigma_{\alpha\beta}\right)}{2c_s^2}\right), i \neq 0 \end{cases}$$

$$g_i^{m,eq} = \begin{cases} c_m - \sum_{i \neq 0} g_i^{m,eq}, i = 0 \\ \omega_i\left(\frac{\Gamma_m \mu_m}{c_s^2} + \frac{c_m e_{i\alpha}u_\alpha}{c_s^2} + \frac{c_m u_\alpha u_\beta\left(e_{i\alpha}e_{i\beta} - c_s^2\sigma_{\alpha\beta}\right)}{2c_s^2}\right), i \neq 0 \end{cases}$$

where $c_s = c/\sqrt{3}$ is the lattice speed of sound, $c = \Delta x / \Delta t$, Δx and Δt are the lattice space and time steps, which are equal to unity.

In the D2Q9 model the discrete velocities are calculated using the formulas

$$e_{ix} = (0,1,1,0,-1,-1,-1,0,1)c$$

$$e_{iy} = (0,0,1,1,1,0,-1,-1,-1)c$$

The values of weight coefficients are as follows

$$\omega_i = \begin{cases} \frac{4}{9}, i = 0, \\ \frac{1}{9}, i = 1-4, \\ \frac{1}{36}, i = 5-8 \end{cases}$$

In this paper, to add the force term $\vec{F} = F_s + \vec{F}_b = \sum_{i=1}^{3} \mu_i \nabla c_i + \rho \vec{g}$ to LBM we apply the scheme suggested by Guo et al. [9]

$$F_i = \omega_i \left(1 - \frac{\Delta t}{2\tau_f}\right)\left[\frac{\vec{e}_i - \vec{u}}{c_s^2} + \frac{\vec{e}_i(\vec{e}_i \cdot \vec{u})}{c_s^4}\right] \cdot \vec{F}$$

Equations for the distribution functions can be divided into two steps, collision and streaming:

$$f_i^*(\vec{x}, t) = f_i(\vec{x}, t) +$$

$$+\Delta t\left(-\frac{f_i(\vec{x}, t) - f_i^{eq}(\vec{x}, t)}{\tau_f} + F_i\right)$$

$$g_i{}^{m,*}(\vec{x}, t) = g_i{}^m(\vec{x}, t) +$$

$$+\Delta t\left(-\frac{g_i{}^m(\vec{x}, t) - g_i{}^{m,eq}(\vec{x}, t)}{\tau_c}\right)$$

$$f_i(\vec{x} + \vec{e}_i \Delta t, t + \Delta t) = f_i^*(\vec{x}, t)$$

$$g_i^m(\vec{x} + \vec{e}_i \Delta t, t + \Delta t) = g_i^{m,*}(\vec{x}, t)$$

After the second step, it is necessary to calculate the macroscopic variables for density, velocity and phase field:

$$\rho = \sum_{i=0}^{8} f_i, \rho\vec{u} = \sum_{i=0}^{8} f_i \vec{e_i} + \frac{\Delta t}{2}\vec{F}, c_m = \sum_{i=0}^{8} g_i^m$$

Derivatives of macroscopic c_i are calculated using the following second-order isotropic differences [7]:

$$\nabla^2 c_m(\vec{x}, t) = \sum_{i=1}^{8} \frac{2\omega_i[c_m(\vec{x}+\vec{e}_i\Delta t, t) - c_m(\vec{x}, t)]}{c_s^2 \Delta t^2}.$$

For the velocity field, as the no-slip boundary condition in fixed walls ($\vec{x}_w$) the bounce back scheme is used [10]:

$$f_i(\vec{x}_w, t + \Delta t) = f_{-i}(\vec{x}_w, t + \Delta t), \qquad \vec{e}_i \cdot \vec{n} > 0,$$

where the phase is constant and the boundary conditions for the concentration distribution functions are chosen as follows:

$$g_i^m(\vec{x}_w, t + \Delta t) =$$

$$= g_{-i}^m(\vec{x}_w, t + \Delta t) + 2\omega_i c_w, \ \vec{e}_i \cdot \vec{n} > 0,$$

where c_w – near-wall phase.

The Neumann condition for the phase on all other walls:

$$g_i^m(\vec{x}_w, t + \Delta t) = g_{-i}^m(\vec{x}_w, t + \Delta t), \qquad \vec{e}_i \cdot \vec{n} > 0.$$

Algorithm for applying the lattice Boltzmann equations method [11]:

1) Discretization of the physical domain and non-dimensionalization of the related parameters
2) Choice of simulation parameters
3) Domain initialization
4) Executing the collision step
5) Application of the boundary conditions
6) Executing the streaming step
7) Calculation of the macroscopic parameters.

Simulation results

We consider the evolution of the ternary fluid mixture in a rectangular computational domain with dimensions: $N_x \times N_y$, $N_x = 80, N_y = 80$. The physical size of the length is $L = 0.01\ m$. The space and time steps are defined as $\Delta x = \frac{L}{N_x} = 0{,}000125, \Delta t = 0.000117188$.

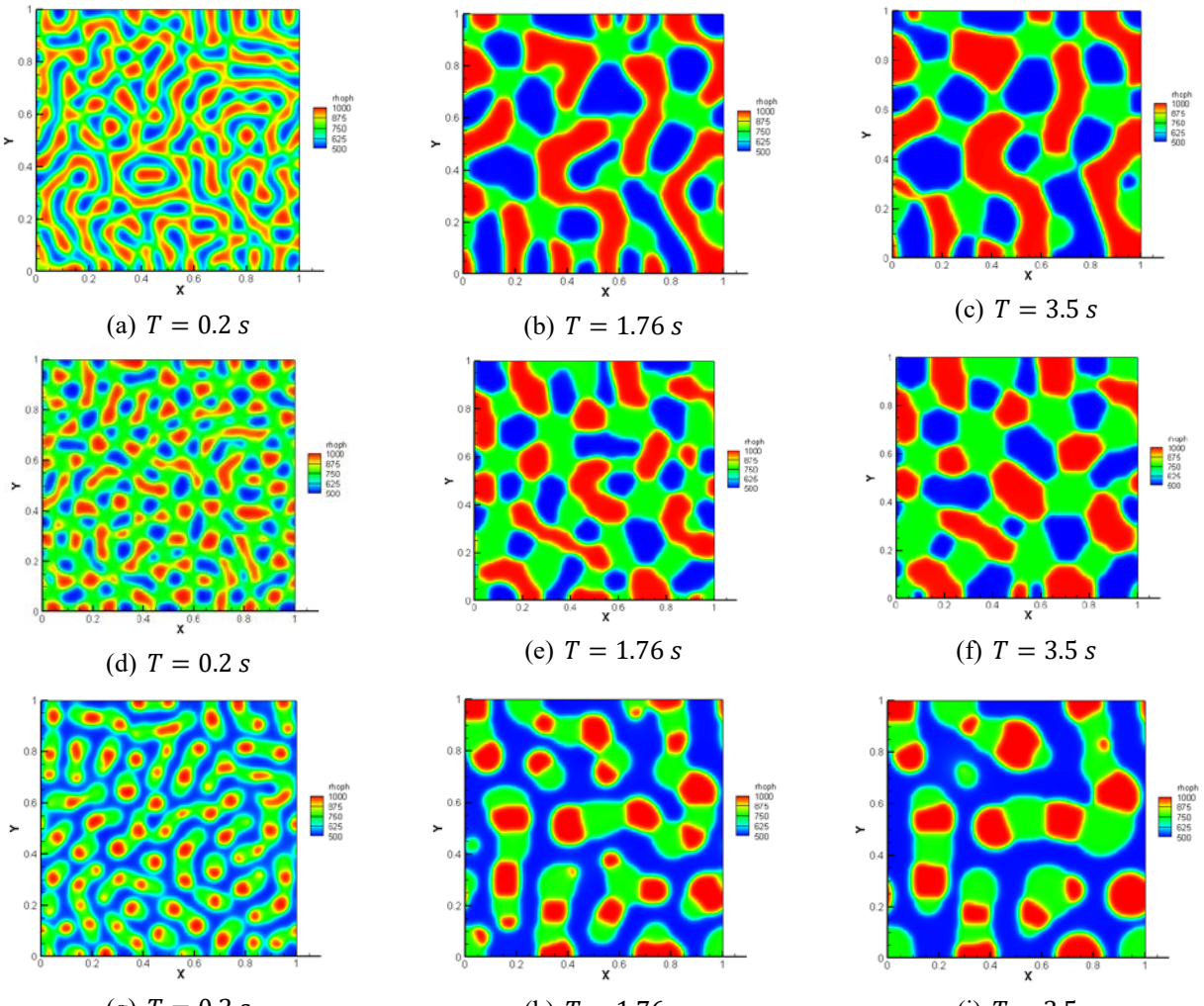
Physical quantities: the density – $\rho_1 = 1000\frac{kg}{m^3}$, $\rho_2 = 750\frac{kg}{m^3}$, $\rho_3 = 500\ \frac{kg}{m^3}$ and the viscosity – $\mu_1 = \mu_2 = \mu_3 = 0.01\ Pa * s$, the acceleration of gravity – $g = 9.8\frac{m}{s^2}$. Dimensionless quantities: Reynolds number – $Re = 234.787$, the capillarity number – $Ca = 0.000417399$ and Atwood number $A = 0.142857$.

Computer simulation by the lattice Boltzmann equations method is performed in lattice units, i.e. the physical parameters of the model are replaced by their lattice analogs using transformation coefficients $Cu = 1.06667$, $Cg=9102.22$. LBM parameters: the density – $\rho_1 = 1.33, \rho_2 = 1, \rho_3 = 0.67$, relaxation times – $\tau_1 = \tau_2 = \tau_3 = 0.8$, the surface tension – $\sigma_{12} = \sigma_{13} = \sigma_{23} = 0.01$, the surface thickness – $D = 2$, the acceleration of gravity – $g = 0.00107666$, and $U_{lbm} = 0.293484$.

The simulation results (Figure 2, 3) show the dynamic change of fluids – the mixture separation of immiscible fluids depending on the fractions of fluid concentrations. The average values of the concentration fractions are taken equal to $(\bar{c}_1, \bar{c}_2, \bar{c}_3) = (0.4,0.2,0.4)$, $\left(\frac{1}{3}, \frac{1}{3}, \frac{1}{3}\right)$, $(0.25, 0.25, 0.5)$. In the first scenario (Figure 2), which does not take into account the acceleration of gravity, one can see the spinodal decomposition of the mixture over time, resulting from the influence of surface tension between the fluids.

In the second scenario (Figure 3), which takes into account the acceleration of gravity, at an early stage ($T < 1\ s$) a less dense fluid ($\rho_3 = 500$) begins to rise, while a denser fluid ($\rho_1 = 1000$) begins its downward movement. Eventually, stable three layers of fluid components are formed: the denser fluid at the bottom and the less dense fluid at the top.

The results obtained determine the adequacy of the constructed model for a three-component fluid.

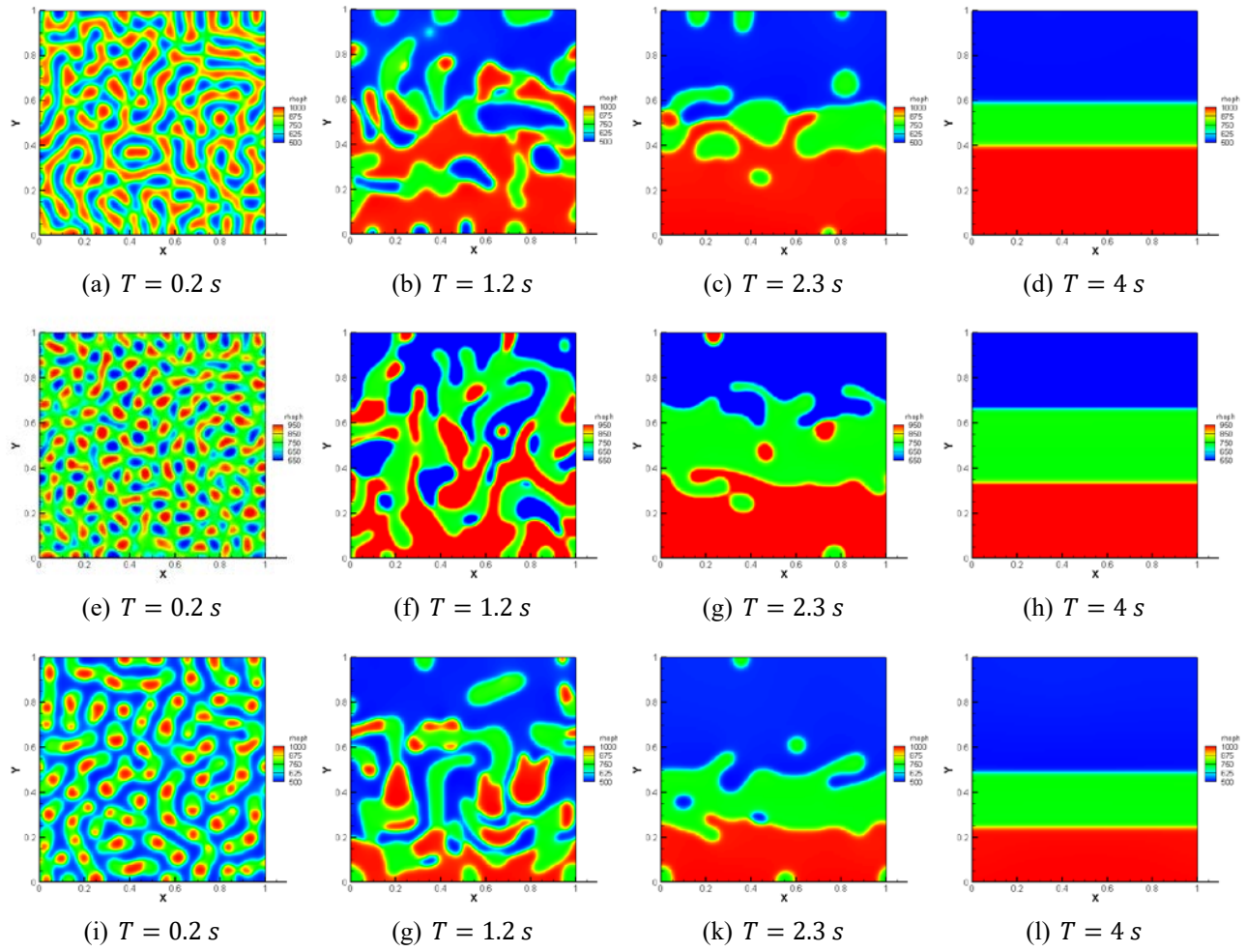


(a) $T = 0.2\ s$ (b) $T = 1.76\ s$ (c) $T = 3.5\ s$

(d) $T = 0.2\ s$ (e) $T = 1.76\ s$ (f) $T = 3.5\ s$

(g) $T = 0.2\ s$ (h) $T = 1.76\ s$ (i) $T = 3.5\ s$

Figure 2 – Ternary fluid separation depending on time for different fractions of fluid concentrations: (a)-(c) $(\bar{c}_1, \bar{c}_2, \bar{c}_3) = (0.4,0.2,0.4)$, (d)-(f) $(\bar{c}_1, \bar{c}_2, \bar{c}_3) = \left(\frac{1}{3}, \frac{1}{3}, \frac{1}{3}\right)$, (g)-(i) $(\bar{c}_1, \bar{c}_2, \bar{c}_3) = (0.25,0.25,0.5)$.



(a) $T = 0.2\ s$ (b) $T = 1.2\ s$ (c) $T = 2.3\ s$ (d) $T = 4\ s$

(e) $T = 0.2\ s$ (f) $T = 1.2\ s$ (g) $T = 2.3\ s$ (h) $T = 4\ s$

(i) $T = 0.2\ s$ (g) $T = 1.2\ s$ (k) $T = 2.3\ s$ (l) $T = 4\ s$

Figure 3 – Effect of body force on the time evolution of density contours of a ternary fluid mixture for different fluid concentration fractions: (a)-(d) $(\bar{c}_1, \bar{c}_2, \bar{c}_3) = (0.4, 0.2, 0.4)$, (e)-(h) $(\bar{c}_1, \bar{c}_2, \bar{c}_3) = \left(\frac{1}{3}, \frac{1}{3}, \frac{1}{3}\right)$, (i)-(l) $(\bar{c}_1, \bar{c}_2, \bar{c}_3) = (0.25, 0.25, 0.5)$.

Conclusion

The paper proposes a mathematical and numerical model for studying the separation process of a three-component fluid. To implement this model, a 2D numerical algorithm has been developed based on the D2Q9 scheme of lattice Boltzmann equations method in a limited cavity in the shape of a rectangle. Numerical simulation was carried out with and without taking into account gravity. The results of numerical simulation showed that, depending on the initial fractions of fluid concentrations, spinodal separation occurs in different ways. From a physical point of view, this is explained by the effect of surface tension between fluids. Due to the influence of gravity, over time, denser, medium-density and less dense fluids begin to arrange themselves in order, from bottom to top, respectively. The stability of the process sets in when the denser fluid component moves down completely. The results of this paper prove the applicability of the methods used in the paper for solving problems of this type.

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