Lattice Boltzmann model for binary mixtures

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An *a priori* derivation of the lattice Boltzmann equations for binary mixtures is provided by discretizing the Boltzmann equations that govern the evolution of binary mixtures. The present model leads to a set of two-fluid hydrodynamic equations for the mixture. In existing models, employing the single-relaxation-time approximation, the viscosity and diffusion coefficients are coupled through the relaxation parameter τ, thus limited to unity Prandtl number and Schmidt number. In the present model the viscosity and diffusion coefficient are independently controlled by two relaxation parameters, thus enabling the modeling of mixtures with an arbitrary Schmidt number. The theoretical framework developed here can be readily applied to multiple-species mixing.

The lattice Boltzmann equation (LBE) [1–6] is emerging as an effective computational method based on fundamental physics for simulating complex flows such as multiphase [7–10] and multiple-component flows [11–13], flows through porous media (cf. Ref. [5]), and particulate suspensions in fluid flows (e.g., Ref. [14]). Recently, important strides have been made on the theoretical front, establishing, from fundamental principles, the physical legitimacy and mathematical rigor of the LBE method. Most importantly, it has been proved that the lattice Boltzmann equation can be derived from the Boltzmann equation *a priori* [2–4,9,10]. It should be pointed out that the Boltzmann equation bridges the gap between the microscopic dynamics and the macroscopic hydrodynamics. Indeed the Navier-Stokes equations can be rigorously derived from the Boltzmann equation via the Chapman-Enskog analysis. The second important theoretical result is the demonstration that the lattice Boltzmann equation is indeed equivalent to an explicit finite difference scheme of the Navier-Stokes equations [15]. These theoretical developments have completely and comprehensively resolved all doubts surrounding the early lattice-gas automata [16] and lattice Boltzmann models. The present day lattice Boltzmann equation is a viable alternative to the continuum methods for simulating fluid flows. Much of the rigorous work with lattice Boltzmann methods so far has been restricted to simple single-phase single-component fluids. Recently, the LBE model for single-component multiphase fluids has been derived from the Enskog equation [9,10]. A rigorous mathematical development of the lattice Boltzmann method for multicomponent fluids is still in its infancy and such is the object of the present work.

In many practical flows involving pollutant dispersion, chemical processing, and combustor mixing and reaction, mass and momentum transport in multispecies fluids plays an important role. For these applications, the continuum-based models can be difficult to compute due to various reasons such as complexity of flow geometry and phase change.

Moreover, it is difficult to construct the continuum-based models from first principles. Therefore for these flows, there is a growing interest in using the lattice Boltzmann equation [11–13]. In this paper, we develop a unified approach for developing the lattice Boltzmann models for multicomponent fluids within the framework of kinetic theory. This work is a part of our continuing effort to set the lattice Boltzmann equation on a rigorous foundation [2,3,9,10]. Specifically, we will derive a conservative discretized version of the continuum Boltzmann equation for fluid mixtures. We shall present a model that is capable of simulating either a miscible or immiscible binary mixture. The lattice Boltzmann equation considered here can be extended to a mixture of three or more species.

The kinetic theory of gas mixtures encompasses a significant amount of literature (e.g., Refs. [17–27]). In a manner similar to the derivation of the Boltzmann equation for a pure system of single species, one can derive N simultaneous equations for a system of N species by reducing the appropriate Liouville equation. For the sake of simplicity without loss of generality, we shall only discuss the Boltzmann equations for a binary system,

$$\partial_t f^A + \boldsymbol{\xi} \cdot \boldsymbol{\nabla} f^A + \boldsymbol{a}_A \cdot \boldsymbol{\nabla}_\xi f^A = Q^{AA} + Q^{AB}, \tag{1}$$

where $Q^{AB} = Q^{BA}$ is the collision term due to the interaction between two different species A and B. Obviously, for an N-component system, there will be N such equations, each containing N collision terms on the right-hand side. In general, the collision term is [23–25]

$$Q^{AB} = \int d\boldsymbol{\xi}_B d\Omega \, \sigma_{AB} \|\boldsymbol{\xi}_B - \boldsymbol{\xi}_A\| [f'^A f'^B - f^A f^B]. \tag{2}$$

Obviously, the equations for a system of multiple species are much more formidable to analyze than the comparable equation for a pure system of single species. The first modeling objective is to find a suitable approximation for the collision terms of Eq. (2). Many of the kinetic models for gas mixtures are based upon the linearized Boltzmann equation [25,28]. The simplest model for a binary mixture is that in Ref. [18],

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which is an extension of the single-relaxation-time model for a pure system (the Bhatnagar-Gross-Krook or BGK model) [28].

With the BGK approximation [18,28], the collision integral $Q^{\sigma\varsigma}[\sigma,\varsigma \in (A, B)]$ becomes

$$J^{\sigma\sigma}=-\frac{1}{\lambda_\sigma}[f^\sigma-f^{\sigma(0)}], \quad J^{\sigma\varsigma}=-\frac{1}{\lambda_{\sigma\varsigma}}[f^\sigma-f^{\sigma\varsigma(0)}], \quad (3)$$

where $f^{\sigma(0)}$ and $f^{\sigma\varsigma(0)}$ are Maxwellians,

$$f^{\sigma(0)}=\frac{\rho_\sigma}{(2\pi R_\sigma T_\sigma)^{D/2}}\exp\left[-\frac{(\boldsymbol{\xi}-\boldsymbol{u}_\sigma)^2}{2R_\sigma T_\sigma}\right], \quad (4)$$

$$f^{\sigma\varsigma(0)}=\frac{\rho_\sigma}{(2\pi R_\sigma T_{\sigma\varsigma})^{D/2}}\exp\left[-\frac{(\boldsymbol{\xi}-\boldsymbol{u}_{\sigma\varsigma})^2}{2R_\sigma T_{\sigma\varsigma}}\right], \quad (5)$$

where D is the spatial dimension, $R_\sigma=k_B/m_\sigma$ is the gas constant of the σ species, k_B is the Boltzmann constant, and m_σ is the molecular mass of the σ species. There are three adjustable relaxation parameters in the collision terms: λ_σ, λ_ς, and $\lambda_{\sigma\varsigma}=(\rho_\varsigma/\rho_\sigma)\lambda_{\varsigma\sigma}$. The first Maxwellian $f^{\sigma(0)}$ is characterized by the conserved variables of each individual species: the mass density ρ_σ (or the number density $n_\sigma=\rho_\sigma/m_\sigma$), the mass velocity $\boldsymbol{u}_\sigma$, and temperature T_σ; while the second Maxwellian $f^{\sigma\varsigma(0)}$, and $f^{\varsigma\sigma(0)}$, is characterized by four adjustable parameters: $\boldsymbol{u}_{\sigma\varsigma}$, $\boldsymbol{u}_{\varsigma\sigma}$, $T_{\sigma\varsigma}$, and $T_{\varsigma\sigma}$. There are several considerations in determining these arbitrary parameters: simplicity of the resulting theory, accuracy of approximation, and ease of computation [24]. One salient difference between using $\boldsymbol{u}$ and T of the mixture in the Maxwellian $f^{\sigma\varsigma(0)}$ as opposed to using $\boldsymbol{u}_\sigma$ and T_σ for the species is that the former choice leads to a single-fluid theory while the latter leads to a two-fluid theory [20,21]. Obviously, when the properties of the two species are vastly different, the two-fluid theory is preferred [24].

The cross-collision term $J^{\sigma\varsigma}$ can be better approximated by expanding f^σ around the Maxwellian [20],

$$J^{\sigma\varsigma}=-\frac{f^{\sigma(0)}}{\rho_\sigma k_B T_\sigma}\left[\mu_D\boldsymbol{c}_\sigma\cdot(\boldsymbol{u}_\sigma-\boldsymbol{u}_\varsigma)+\mu_T\frac{3}{2}\left(\frac{\boldsymbol{c}_\sigma^2}{2R_\sigma T_\sigma}-1\right)\right.$$

$$\left.\times(T_\sigma-T_\varsigma)-M_{\sigma\varsigma}\left(\frac{\boldsymbol{c}_\sigma^2}{2R_\sigma T_\sigma}-1\right)(\boldsymbol{u}_\sigma-\boldsymbol{u}_\varsigma)^2\right], \quad (6)$$

where $\boldsymbol{c}_\sigma=(\boldsymbol{\xi}-\boldsymbol{u}_\sigma)$ is the peculiar (or thermal) velocity of the σ species, and $M_{\sigma\varsigma}$ is a function of the density and temperature, and other parameters [20,27].

We now consider the following model equations for a binary mixture due to Sirovich [20]:

$$\partial_t f^\sigma+\boldsymbol{\xi}\cdot\boldsymbol{\nabla}f^\sigma+\boldsymbol{a}_\sigma\cdot\boldsymbol{\nabla}_\xi f^\sigma=J^{\sigma\sigma}+J^{\sigma\varsigma}, \quad (7)$$

where the self-collision term $J^{\sigma\sigma}$ is approximated by the BGK model of Eq. (3), and the cross-collision term $J^{\sigma\varsigma}$ is given by Eq. (6). Solving Eqs. (7) by means of iteration (cf. Refs. [20,27]), one first obtains

$$\boldsymbol{u}_\sigma=\boldsymbol{u}_\varsigma=\boldsymbol{u}, \quad T_\sigma=T_\varsigma=T, \quad (8)$$

$$f^{\sigma(0)}=f^{\sigma\varsigma(0)}=\frac{\rho_\sigma}{(2\pi R_\sigma T)^{D/2}}\exp\left[-\frac{(\boldsymbol{\xi}-\boldsymbol{u})^2}{2R_\sigma T}\right]. \quad (9)$$

Two salient features of the model equations (7) should be addressed. First, the cross-collision term $J^{\sigma\varsigma}$ of Eq. (6) is exact for the Maxwell molecules, whose interaction potential obeys the inverse fifth-power law. Equations (7), therefore, can be considered to be a model for the Maxwell gas [20]. One immediate consequence of this approximation is that the diffusion force does not contain a thermal diffusion term, as it should. Second, the BGK approximation of the self-collision term $J^{\sigma\sigma}$ of Eq. (3) imposes the limitation of a fixed Prandtl number (of unity). However, both these limitations of the model can be overcome by using the linearized Boltzmann equation with multiple relaxation times [29–31] and a nonlinear approximation of the collision terms [20,21,32].

We construct a lattice Boltzmann model for a binary mixture based on the model equations (7). In the present work we will only consider the isothermal case such that $T_\sigma=T_\varsigma=T_{\sigma\varsigma}=T=$ const. Consequently, we can also ignore the terms related to thermal effects in $J^{\sigma\varsigma}$ of Eq. (6) by setting $\mu_T=M_{\sigma\varsigma}=0$, i.e.,

$$J^{\sigma\varsigma}=-\frac{1}{\tau_D}\frac{\rho_\varsigma}{\rho}\frac{f^{\sigma(0)}}{k_B T}(\boldsymbol{\xi}-\boldsymbol{u})\cdot(\boldsymbol{u}_\sigma-\boldsymbol{u}_\varsigma), \quad (10)$$

where the equilibrium function $f^{\sigma(0)}$ for the σ species is chosen to be the Maxwellian equilibrium distribution depending on the mass velocity of the σ species $\boldsymbol{u}_\sigma$ as

$$f^{\sigma(0)}=\frac{\rho_\sigma}{(2\pi R_\sigma T)^{D/2}}\exp\left[-\frac{(\boldsymbol{\xi}-\boldsymbol{u}_\sigma)^2}{2R_\sigma T}\right]. \quad (11)$$

We can derive the lattice Boltzmann equation by discretizing the model equations (7) as in Refs. [2,3,9,10]:

$$f_\alpha^\sigma(\boldsymbol{x}_i+\boldsymbol{e}_\alpha\delta_t,t+\delta_t)-f_\alpha^\sigma(\boldsymbol{x}_i,t)=\Omega_\alpha^\sigma, \quad (12)$$

where the collision term $\Omega_\alpha^\sigma=J_\alpha^{\sigma\sigma}+J_\alpha^{\sigma\varsigma}-F_\alpha^\sigma\delta_t$, and

$$J_\alpha^{\sigma\sigma}=-(1/\tau_\sigma)[f_\alpha^\sigma-f_\alpha^{\sigma(0)}], \quad (13a)$$

$$J_\alpha^{\sigma\varsigma}=-\frac{1}{\tau_D}\frac{\rho_\varsigma}{\rho}\frac{f_\alpha^{\sigma(\mathrm{eq})}}{c_s^2}(\boldsymbol{e}_\alpha-\boldsymbol{u})\cdot(\boldsymbol{u}_\sigma-\boldsymbol{u}_\varsigma), \quad (13b)$$

$$F_\alpha^\sigma=-w_\alpha\rho_\sigma[(\boldsymbol{e}_\alpha\cdot\boldsymbol{a}_\sigma)/c_s^2], \quad (13c)$$

where ρ_σ and ρ_ς, and $\boldsymbol{u}_\sigma$ and $\boldsymbol{u}_\varsigma$ are the mass densities and flow velocities for species σ and ς, they are the moments of the distribution functions

$$\rho_\sigma=\sum_\alpha f_\alpha^\sigma=\sum_\alpha f_\alpha^{\sigma(0)}, \quad (14a)$$

$$\rho_\sigma\boldsymbol{u}_\sigma=\sum_\alpha f_\alpha^\sigma\boldsymbol{e}_\alpha=\sum_\alpha f_\alpha^{\sigma(0)}\boldsymbol{e}_\alpha, \quad (14b)$$

and ρ and $\boldsymbol{u}$ are, respectively, the mass density and the barycentric velocity of the mixture: $\rho = \rho_\sigma + \rho_s$, $\rho \boldsymbol{u} = \rho_\sigma \boldsymbol{u}_\sigma + \rho_s \boldsymbol{u}_s$. The equilibrium $f_\alpha^{\sigma(0)}$ is a second-order Taylor expansion of $f^{\sigma(0)}$ of Eq. (11) [27], and has the following form, in general [27]:

$$f_\alpha^{\sigma(0)} = f_\alpha^{\sigma(\text{eq})} \left[1 + \frac{1}{c_s^2} (\boldsymbol{e}_\alpha - \boldsymbol{u}) \cdot (\boldsymbol{u}_\sigma - \boldsymbol{u}) \right], \quad (15a)$$

$$f_\alpha^{\sigma(\text{eq})} = w_\alpha \rho_\sigma \left[1 + \frac{(\boldsymbol{e}_\alpha \cdot \boldsymbol{u})}{c_s^2} + \frac{(\boldsymbol{e}_\alpha \cdot \boldsymbol{u})^2}{2c_s^4} - \frac{u^2}{c_s^2} \right], \quad (15b)$$

where coefficients $\{w_\alpha\}$ depend on the discrete velocity set $\{\boldsymbol{e}_\alpha\}$. For the sake of concreteness and simplicity without losing generality, we shall restrict ourselves to a nine-velocity model on a two-dimensional square lattice (D2Q9 model). In this case,

$$w_\alpha = \begin{cases} 4/9, & \alpha = 0 \\ 1/9, & \alpha = 1\text{--}4 \\ 1/36, & \alpha = 5\text{--}8. \end{cases} \quad (16)$$

The sound speed of the model $c_s = c/\sqrt{3}$, $c = \delta_x/\delta_t$, where δ_x is the lattice constant of the underlying square lattice. The forcing term F_α^σ is derived in Refs. [9,10]. The collision terms $J_\alpha^{\sigma\sigma}$ and $J_\alpha^{\sigma s}$ are so constructed to preserve the local mass and momentum conservation laws. The zeroth- and first-order moments of these terms are

$$\sum_\alpha J_\alpha^{\sigma\sigma} = \sum_\alpha J_\alpha^{s\sigma} = \sum_\alpha F_\alpha^\sigma = 0, \quad (17a)$$

$$\sum_\alpha J_\alpha^{\sigma\sigma} \boldsymbol{e}_\alpha = 0, \quad (17b)$$

$$\sum_\alpha J_\alpha^{s\sigma} \boldsymbol{e}_\alpha = -(1/\tau_D)(\rho_\sigma \rho_s/\rho)(\boldsymbol{u}_\sigma - \boldsymbol{u}_s), \quad (17c)$$

$$\sum_\alpha F_\alpha^\sigma \boldsymbol{e}_\alpha = -\rho_\sigma \boldsymbol{a}_\sigma. \quad (17d)$$

The left-hand side of Eq. (12) can be expanded in a Taylor series in δ_t up to second order (cf. Refs. [3,27]), then

$$\delta_t D_\alpha f_\alpha^\sigma + \tfrac{1}{2} \delta_t^2 D_\alpha^2 f_\alpha^\sigma = J_\alpha^{\sigma\sigma} + J_\alpha^{\sigma s} - F_\alpha^\sigma \delta_t, \quad (18)$$

where $D_\alpha = (\partial_t + \boldsymbol{e}_\alpha \cdot \boldsymbol{\nabla})$. By means of the Chapman-Enskog analysis, we can derive the hydrodynamic equations for the mixture from Eq. (18) (see details in Ref. [27]). The mass conservation law for each individual species and the mixture can be derived immediately from Eq. (18):

$$\partial_t \rho_\sigma + \boldsymbol{\nabla} \cdot (\rho_\sigma \boldsymbol{u}_\sigma) = \frac{1}{2} \boldsymbol{\nabla} \cdot \left[\frac{\rho_\sigma \rho_s}{\tau_D \rho} (\boldsymbol{u}_\sigma - \boldsymbol{u}_s) \right], \quad (19)$$

$$\partial_t \rho + \boldsymbol{\nabla} \cdot (\rho \boldsymbol{u}) = 0. \quad (20)$$

The mass conservation of each individual species reflects the fact that all the particles, regardless of their identity, are conserved in a collision without chemical reactions.

The Navier-Stokes equation of the mixture is [27]

$$\rho_\sigma \partial_t \boldsymbol{u}_\sigma + \rho_\sigma \boldsymbol{u}_\sigma \cdot \boldsymbol{\nabla} \boldsymbol{u}_\sigma = -\boldsymbol{\nabla} p_\sigma + \rho_\sigma \nu_\sigma \nabla^2 \boldsymbol{u}_\sigma$$
$$- (\rho_\sigma \rho_s/\tau_D \rho)(\boldsymbol{u}_\sigma - \boldsymbol{u}_s) + \rho_\sigma \boldsymbol{a}_\sigma, \quad (21)$$

where $p_\sigma = n_\sigma k_B T = \rho_\sigma R_\sigma T$ is the partial pressure of the σ species, and the viscosity of the σ species is

$$\nu_\sigma = c_s^2 \delta_t (\tau_\sigma - 1/2). \quad (22)$$

Equation (21) is consistent with the results in Ref. [21].

The difference between the Navier-Stokes equation for individual species (σ and s) leads to

$$\frac{1}{\tau_D}(\boldsymbol{u}_\sigma - \boldsymbol{u}_s) = -\frac{\rho p}{\rho_\sigma \rho_s} \boldsymbol{d}_\sigma - \{[\nu_\sigma \nabla^2 \boldsymbol{u}_\sigma - \nu_s \nabla^2 \boldsymbol{u}_s]$$
$$+ [\partial_t \delta \boldsymbol{u} + \bar{\boldsymbol{u}} \cdot \boldsymbol{\nabla} \delta \boldsymbol{u} + \delta \boldsymbol{u} \cdot \boldsymbol{\nabla} \bar{\boldsymbol{u}}, \quad (23)$$

where $\delta \boldsymbol{u} = (\boldsymbol{u}_\sigma - \boldsymbol{u}_s)$, $\bar{\boldsymbol{u}} = \frac{1}{2}(\boldsymbol{u}_\sigma + \boldsymbol{u}_s)$, $p = n k_B T$ is the total pressure, and $n = n_\sigma + n_s$. The diffusion force

$$\boldsymbol{d}_\sigma = \boldsymbol{\nabla} \left(\frac{n_\sigma}{n} \right) + \frac{n_\sigma n_s}{n \rho} (m_s - m_\sigma) \boldsymbol{\nabla} \ln p + \frac{\rho_\sigma \rho_s}{\rho p}(\boldsymbol{a}_\sigma - \boldsymbol{a}_s) \quad (24)$$

includes the effects due to the molar concentration gradient $\boldsymbol{\nabla}(n_\sigma/n)$, the pressure gradient $\boldsymbol{\nabla} p$, and external forces ($\boldsymbol{a}_\sigma$ and $\boldsymbol{a}_s$).

It has already been assumed in the two-fluid equations that derivatives are slowly varying on the time scale of Maxwellization [21]. Thus to the leading order, we have

$$(\boldsymbol{u}_\sigma - \boldsymbol{u}_s) = -(n^2/n_\sigma n_s) D_{\sigma s} \boldsymbol{d}_\sigma, \quad (25)$$

where the mutual diffusion coefficient in the mixture is

$$D_{\sigma s} = [(k_B T)/(n m_\sigma m_s)] \tau_D. \quad (26)$$

By definition [26] the mass flux of the σ species is

$$\boldsymbol{j}_\sigma = \rho_\sigma (\boldsymbol{u}_\sigma - \boldsymbol{u}) = -\tau_D p \boldsymbol{d}_\sigma.$$

The continuity equation (19) can be rewritten as

$$D_t \rho_\sigma + \rho_\sigma \boldsymbol{\nabla} \cdot \boldsymbol{u} + \boldsymbol{\nabla} \cdot \boldsymbol{j}_\sigma = 0, \quad (27)$$

where $D_t = (\partial_t + \boldsymbol{u} \cdot \boldsymbol{\nabla})$. By assuming that $\boldsymbol{\nabla} \cdot \boldsymbol{u} = 0$, we obtain the following advection-diffusion equation for an isothermal mixture:

$$\partial_t \rho_\sigma + \boldsymbol{u} \cdot \boldsymbol{\nabla} \rho_\sigma = \boldsymbol{\nabla} \cdot (\tau_D - \tfrac{1}{2}) p \boldsymbol{d}_\sigma. \quad (28)$$

Thus the mutual diffusion coefficient given by Eq. (26) must be modified for the LBE model,

$$D_{\sigma s}^* = [c_s^2/(n m_\sigma m_s)](\tau_D - \tfrac{1}{2}), \quad (29)$$

to include the second-order discrete effect [33].

Unlike the existing lattice Boltzmann models for binary mixtures [11–13], the diffusion coefficients of the present model are independent of the viscosity. The diffusion coefficients only depend on the parameter τ_D and other relevant physical properties of the mixture. Also, positive and negative ($\tau_D - 1/2$) correspond to the miscible and immiscible mixtures, respectively. We have verified, Eq. (29), and observed the phase separation when ($\tau_D - 1/2$)<0 in numerical simulations [34].

We have constructed a lattice Boltzmann model for binary mixtures with several important features. The lattice Boltzmann model is directly derived from the kinetic model equations using a formal discretization procedure. The lattice model thus inherits the sound physics and mathematical rigor incumbent in kinetic theory. This is in contrast to previous lattice Boltzmann models for mixtures [11–13], which are not directly based on the fundamental physics of kinetic equations. These models rely on fictitious "interactions" [11,12] or *ad hoc* "free energies" [13] to produce the requisite mixing. These nonphysical effects present a further problem since they are not easily amenable to mathematical analysis [9,10]. The heuristic elements of the previous lattice Boltzmann models [11–13] have been eliminated, resulting in a physically justifiable model that is simple to compute. Further, due to the direct connection to kinetic theory, the derivation of the hydrodynamic equations associated with the lattice Boltzmann model is significantly simplified and rendered mathematically more rigorous.

The second important feature of the present work is that the model is based upon a two-fluid theory of binary mixtures. The previous models [11–13,22], on the other hand, are derived from a simpler, but restrictive, one-fluid theory. In the single-fluid models one is constrained to use the *ad hoc* "equilibrium velocity" [11,12,22]

$$\boldsymbol{u}^{(\mathrm{eq})} = (\tau_s \rho_\sigma \boldsymbol{u}_\sigma + \tau_\sigma \rho_s \boldsymbol{u}_s)/(\tau_s \rho_\sigma + \tau_\sigma \rho_s)$$

in the equilibrium $f_\alpha^{\sigma(0)}$ in order to satisfy the local conservation laws. As a result, the viscosity and diffusion coefficient cannot be independently adjusted without exerting fictitious interactions or *ad hoc* free energies. The analysis of these models becomes unnecessarily tedious and cumbersome [11,12]. The models with free energies [13] do not yield correct hydrodynamic equations [9,10]. Furthermore, these models are single-fluid models which cannot be applied to mixtures of species with vastly different properties. In the present two-fluid model, the diffusion coefficient is independent of the viscosity, and is determined by the parameter τ_D and other relevant physical properties of the mixture. The model is capable of simulating either miscible or immiscible fluids by changing the sign of ($\tau_D - 1/2$) [34].

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