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Viscous coupling based lattice Boltzmann model for binary mixtures

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Abstract

A new lattice Boltzmann model for binary mixtures, which can naturally include both the two-fluid approach and the single-fluid approach, is developed. The model is derived from the continuous kinetic model proposed by Hamel, which independently takes into account self collisions and cross collisions. The original kinetic model is discussed in order to appreciate that cross collisions realize an internal coupling force, proportional to the diffusion velocity, and an additional coupling effect in the effective stress tensor, proportional to the deformation of the barycentric velocity field. For this reason, Hamel's model is the natural forerunner of all linearized models based on the two-fluid approach and allows us to describe binary mixtures at different limiting regimes consistently. A discrete lattice Boltzmann model, which recovers the original Hamel's model with second order accuracy in both time and space, is proposed. This discrete model can analyze ordinary diffusion, pressure diffusion and forced diffusion.

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I. INTRODUCTION

There are numerous flow systems in both natural and industrial processes that involve mass and momentum transport within different miscible species. In order to describe these phenomena, many theoretical models have been developed by means of the continuum approach. However some applications exist where a more fundamental point of view can be fruitfully applied. In particular the reactive mixtures, commonly considered in chemical applications, involve catalytic porous media where the critical size can be reduced so much to become comparable with the mean free path of the fluid particles. It seems natural to adopt in these cases a kinetic approach in order to try to overcome the difficulties due to the macroscopic approach.

Unfortunately, only in a few simple cases it is possible to formulate an accurate kinetic model, for example the model based on the Boltzmann equation for an ideal gas, and even in fewer cases it is possible to numerically solve it [1]. This consideration explains the interest in using simplified kinetic models [2–4] for reducing the computational efforts. These models involve some microscopic parameters which are usually adjusted in order to recover the macroscopic transport coefficients. In this way, the intrinsic nature of the microscopic approach is violated and finally a mesoscopic tool is obtained, which somehow produces a simplified microscopic picture of the phenomenon on the basis of the macroscopic information. On the other hand, as long as the macroscopic equations are recovered, the mesoscopic models allow us to widen the set of possible applications beyond the constraints due to the derivation process of the accurate kinetic models.

The lattice Boltzmann method (LBM) in the past few years has become a very popular discretization technique used to solve simplified kinetic models [5–7, 9–11]. When complex geometries are involved and the inter-particle interactions must be taken into account, the discretized models derived by means of the lattice Boltzmann method offer some computational advantages over continuum based models, particularly for large parellel computing. A more complete and recent coverage of various previous contributions on LBM is beyond the purposes of the present work, but can be found in [12].

A promising application for lattice Boltzmann models is the analysis of reactive mixtures in porous catalysts [13, 14]. For this reason, a lot of work has been performed in recent years to produce accurate lattice Boltzmann models for multi-component fluids and, in particular, for mixtures composed of miscible species [15–28]. The problem is to find a proper way, within the framework of a simplified kinetic model, for describing the interactions among particles of different types. Once this is defined, the extension of the model to reactive flows is straightforward [29, 30] and it will essentially involve additional source terms in the species equations according to the reaction rate. Unfortunately, most existing lattice Boltzmann models for mixtures are based on pseudo-potential interactions [16–18], heuristic free energies [15, 19–22] or linearized cross collisional operators [24–28]. Actually, the lattice model in [28] involves a nonlinear cross collisional operator, which has been heuristically conjectured without any theoretical foundation. However, the Chapman-Enskog asymptotic analysis, reported in the original paper, was performed only for a linearized approximation and for this reason the nonlinear features of this model are not completely clear.

The older models [15–23] were based on the *single-fluid approach*. Essentially, the averaged effect due to both self-collisions and cross-collisions is described by means of a total BGK-like collisional operator. Considering some of the special mixture properties in the Maxwellian distribution function of the BGK-like collisional operator, each species will be forced to evolve towards the mixture equilibrium conditions. For almost a decade now, diffusions driven by concentration, pressure, temperature and external forces have been studied by these models for an arbitrary number of components with non-ideal interactions. However, this approach is characterized by the following drawbacks [4, 31, 32].

1. It is not completely correct to jointly model self collisions, which involve particles of the same type, and cross collisions, which involve particles of different types, by using a unique simplified collisional operator. The obtained mesoscopic framework is very far from actual microscopic dynamics and from the macroscopic point of view the kinematic viscosity of each species, the mutual diffusivity and the mixture kinematic viscosity are coupled to each other. For this reason, the single momentum equation for each species cannot be properly solved.

2. The mixture momentum conservation is locally ensured by using a Maxwellian distribution function for these models that involve a mixture equilibrium velocity, which is essentially a properly conjectured linear combination of species velocities involving the relaxation time constants. Although the mixture equilibrium velocity allows us to ensure the desired momentum conservation, the fact that it is not completely defined

only in terms of the macroscopic parameters, because of the intrinsic dependence on the relaxation time constants, is not very convenient.

3. Modeling the interactions among particles of different types and macroscopically recovering the desired diffusion equations requires these models to adopt an interaction pseudo-potential or a long-range coupling force, allowing for the introduction of an additional momentum exchange among particles. In this way, a proper fitting of the consequent macroscopic equations allows one to mimic all the diffusion driving mechanisms of the continuous kinetic theory. Although this approach has been sufficiently justified by careful theoretical analysis [33], a model, which includes these diffusion driving mechanisms, without any kind of *ad hoc* cumbersome fitting would be obviously much more preferable.

The conclusion is that, although the single-fluid approach proved to be an accurate numerical tool for solving some macroscopic equations for a large number of applications, it provides a limited mesoscopic picture of the phenomena.

On the other hand, some models [25–28] based on the *two-fluid approach* have been proposed. According to this approach, each species relaxes towards its equilibrium configuration according to its specific relaxation time and some coupling must be considered in order to describe the collisions among different species. Some models [25–27] adopt a *force coupling* in the momentum equations, which is derived from a linearized kinetic term. This technique allows us to describe the effects of collisions among particles of different species by means of an approximated forcing term. Recently, another model has been proposed, which tries to overcome this approximation [28]. In this case any approximation is avoided in the formal formulation of the model, but there is no discussion about the effects of this improvement in the hydrodynamic equations. The original [28] reports a Chapman-Enskog asymptotic analysis of a linearized version of the proposed model based on a simple *force coupling* in the momentum equations, which produces results similar to those of previous models.

The best way to understand the limits of *force coupling* and the possible ways to overcome them is to consider, once again, the kinetic theory. It is well known that the lattice Boltzmann models can be directly derived from the kinetic models using some standard discretization procedures and proper approximations [7, 8, 34]. The paper [35] reportes a more complete theoretical framework concerning how to systematically construct LBM models from the continuous kinetic theory. There is a significant amount of literature concerning gas mixtures within the kinetic theory framework [31, 32]. In his doctoral thesis, Kolodner [36], following Grad's moment method, investigated what variables, in addition to the classical fundamental variables, must be considered in order to properly describe the phenomena occurring in binary mixtures. The classical work of Chapman and Cowling [31] was concerned with the determination of the transport coefficients for binary mixtures by means of the full Boltzmann equations. Among the simplified kinetic models, the first single-fluid model for binary mixtures is due to Gross and Krook [37, 38], which is based on a BGK-like collisional operator. Sirovich [39, 40] proposed the linearization of the previous model's equation. Actually the Sirovich's model is based on nonlinear equations because the linearization was done around a local Maxwellian [41]. This model historically started the two-fluid approach. Trying to generalize Sirovich's results, Hamel [42–44] proposed a simplified kinetic model which was able to include both the single-fluid and two-fluid approaches, by considering multiple equilibrium distribution functions involving the respective species velocities and the mixture velocity. Unfortunately in the original paper [43] no Chapman-Enskog asymptotic analysis of the model was reported and the transport properties were discussed by using the coefficients appearing in Sirovich's equation. Reducing the computational efforts, the linearized kinetic models became very popular and they were mathematically formalized [45]. More recently it has been pointed out that none of the previous models reduce to a BGK-like equation when mechanically identical components are considered, despite the fact that all of them are based on a BGK-like equation for each species [46]. This means that none of the previous models satisfies the indifferentiability principle, i.e. the fact that when all the species are identical one recovers the equation for a single component gas, which is correctly satisfied by a single-fluid model, recently proposed [47].

Following the style of Hamel's work, a two-fluid simplified kinetic model is proposed here and only small changes are introduced in order to satisfy the indifferentiability principle when cross collisions prevail. The model is formulated in such a way as to recover the conventional BGK equations for the limiting case of non-interacting particles and the consistent single-fluid approach for ideally coupled particles. The hydrodynamic equations are fully derived by means of the Chapman-Enskog asymptotic analysis, allowing us to point out that the model is characterized by an additional coupling among the species, called *viscous coupling* to distinguish it from the *force coupling* previously considered. A strategy for tuning the mesoscopic parameters of the model to recover the desired transport coefficients is proposed. Finally, a lattice (discretized) version of the previous model and a strategy for tuning the lattice mesoscopic parameters are also discussed. In the present paper, only isothermal conditions and nearly incompressible flows are considered, because they are enough to analyze the effects of *viscous coupling*. The properties of the Maxwell molecule are assumed in deriving the model and only the problem of binary mixtures is considered. The full generalization of the method to gas mixtures is quite straightforward, as only few changes are required.

This paper is organized as follows. Section II provides a brief review for some of the existing mixture kinetic models. In particular, Section II A discusses Hamel's model, which forms the theoretical basis of the present paper and Section II C discusses some linearized models, which include the well-known Sirovich's model. The proposed lattice Boltzmann model is designed and the hydrodynamic equations are derived in Section III. Lastly in Section IV some numerical results are reported for the proposed discrete lattice model and the conclusions of the paper are discussed. The Chapman-Enskog asymptotic analysis of the continuous Hamel's model and of the lattice Boltzmann model for binary mixtures are reported respectively in Appendix A and in Appendix B.

II. KINETIC THEORY OF BINARY MIXTURES

A. Adopted kinetic model

Let us consider a mixture simply composed of two types of particles, labeled a and b. Following the derivation of the Boltzmann equation for a pure system of single species, the kinetic equations for a mixture can be derived in a very similar way [4, 31, 32]:

$$\frac{\partial f_a}{\partial t} + \mathbf{v} \cdot \nabla f_a + \mathbf{g}_a \cdot \nabla_{\mathbf{v}} f_a = Q_{a\,a} + Q_{a\,b}, \tag{1}$$

$$\frac{\partial f_b}{\partial t} + \mathbf{v} \cdot \nabla f_b + \mathbf{g}_b \cdot \nabla_{\mathbf{v}} f_b = Q_{b\,b} + Q_{b\,a}, \tag{2}$$

where $Q_{a\,a}$ and $Q_{b\,b}$ are the collisional terms which describe the collisions among particles of the same type (self collisions), while $Q_{a\,b}$ and $Q_{b\,a}$ are the collisional terms due to the interactions among different species (cross collisions). Each collisional term has a well-known structure similar to the collisional operator involved in the Boltzmann equation for the single fluid. The time evolution of the distribution function for each species is affected both by collisions with particles of the same type and with particles of different type. These two phenomena are the kinetic driving forces of the equilibration process for the whole mixture. A simplified kinetic model which allows us to separately describe both driving forces, as it happens for the original Boltzmann equations, would be desirable. Essentially, the key idea is to substitute the previous collisional terms with simplified ones $Q(f,f) \rightarrow J(f)$, which are selected with a BGK-like structure. In the following only the equation for a generic species $\sigma = a, b$ will be considered. The simplified kinetic equation has the general form:

$$\frac{\partial f_\sigma}{\partial t} + \mathbf{v} \cdot \nabla f_\sigma + \mathbf{g}_\sigma \cdot \nabla_{\mathbf{v}} f_\sigma = -\frac{1}{\tau_\sigma}\left[f_\sigma - f_\sigma^e\right] - \frac{1}{\tau_{m\,\sigma}}\left[f_\sigma - f_{\sigma(m)}^e\right], \tag{3}$$

where τ_σ is the relaxation time constant for self collisions, $\tau_{m\,\sigma}$ is the relaxation time constant for the cross collisions, f_σ^e is a Maxwellian distribution function of the specific velocity, while $f_{\sigma(m)}^e$ is a Maxwellian distribution function of a mixture characteristic velocity. The explicit expressions of the previous Maxwellians are:

$$f_\sigma^e = \frac{\rho_\sigma}{m_\sigma \left(2\pi e_\sigma\right)^{D/2}} \exp\left[-\frac{\left(\mathbf{v} - \mathbf{u}_\sigma\right)^2}{2\, e_\sigma}\right], \tag{4}$$

$$f_{\sigma(m)}^e = \frac{\rho_\sigma}{m_\sigma \left(2\pi e_\sigma\right)^{D/2}} \exp\left[-\frac{\left(\mathbf{v} - \mathbf{u}_x\right)^2}{2\, e_\sigma}\right], \tag{5}$$

where ρ_σ is the density, m_σ is the particle mass and $\mathbf{u}_\sigma$ is the macroscopic velocity, while e_σ and $\mathbf{u}_x$ are tunable parameters of the model. The parameters $\tau_{m\,\sigma}$ and $\mathbf{u}_x$ are not independently tunable parameters. In order to satisfy the local momentum conservation for the whole mixture, the following condition must hold:

$$\sum_\sigma \int m_\sigma\, \mathbf{v}\left[f_\sigma - f_{\sigma(m)}^e\right]/\tau_{m\,\sigma}\, d\mathbf{v} = \sum_\sigma \rho_\sigma\left(\mathbf{u}_\sigma - \mathbf{u}_x\right)/\tau_{m\,\sigma} = 0. \tag{6}$$

The tunable parameters of the previous model may be easily obtained by demanding that the moments of the model equations yield, in addition to the conservation equations, the correct ratio for the times characterizing the relaxation of the velocity and temperature differences [41]. In this way the results of Hamel [42] are recovered without any approximation and the mixture characteristic velocity can be identified with the mass averaged velocity $\mathbf{u}_x = \mathbf{u}_m$, where

$$\mathbf{u}_m = \frac{\sum_\sigma m_\sigma \mathbf{u}_\sigma}{\sum_\sigma m_\sigma}. \tag{7}$$

The local momentum conservation given by Eq. (6) implies that the quantity $\rho_\sigma/(m_\sigma \tau_{m\,\sigma})$ must be a constant and so the cross-collision relaxation time constants differ from one another. It is easy to check that Hamel's model does not satisfy the indifferentiability principle. In the following, a strategy for setting the tunable parameters will be proposed, which essentially allows a smooth transition from the two-fluid approach to the single-fluid approach. The characteristic velocity will be set in such a way as to guarantee the indifferentiability principle at least for the fully-coupled configuration, when the mixture evolves as a single fluid. The characteristic velocity of the mixture can be identified with the barycentric velocity

$$\mathbf{u}_x = \mathbf{u} = \sum_\sigma x_\sigma \mathbf{u}_\sigma \tag{8}$$

where $x_\sigma = \rho_\sigma / \sum_\sigma \rho_\sigma$ is the mass concentration of the generic species. In this case, the local momentum conservation given by Eq. (6) implies $\tau_{m\,a} = \tau_{m\,b} = \tau_m$. It easy to check that if cross collisions prevail, the summation of the BKG-like kinetic equations for each species allows us to recover a BGK-like kinetic equation for the mixture.

The Chapman-Enskog asymptotic analysis of the previous kinetic model yields (see Appendix A):

$$\frac{\partial \rho_\sigma}{\partial t} + \nabla \cdot (\rho_\sigma \mathbf{u}_\sigma) = 0, \tag{9}$$

$$\begin{aligned}
\frac{\partial \left(\rho_\sigma \mathbf{u}_\sigma\right)}{\partial t} &+ \nabla \cdot \left[\left(1-\alpha_\sigma\right) \rho_\sigma \mathbf{u}_\sigma \otimes \mathbf{u}_\sigma + \alpha_\sigma \, \rho_\sigma \mathbf{u} \otimes \mathbf{u} \right. \\
&+ \left. \alpha_\sigma \rho_\sigma \mathbf{u}_{\alpha(\sigma)} \otimes \mathbf{w}_\sigma + \alpha_\sigma \rho_\sigma \mathbf{w}_\sigma \otimes \mathbf{u}_{\alpha(\sigma)} \right] = \\
&- \nabla \left(\rho_\sigma e_\sigma\right) + \rho_\sigma \mathbf{g}_\sigma - \frac{1}{\tau_m} \rho_\sigma \mathbf{w}_\sigma \\
&+ \nabla \cdot \left\{ \alpha_\sigma \rho_\sigma e_\sigma \tau_m \left[\nabla \mathbf{u}_{\alpha(\sigma)} + \nabla \mathbf{u}_{\alpha(\sigma)}^T \right] \right\},
\end{aligned} \tag{10}$$

where $\alpha_\sigma = \tau_\sigma / \left(\tau_\sigma + \tau_m\right)$ is a bounded function of the relaxation time constants such that $0 \leq \alpha_\sigma \leq 1$, $\mathbf{w}_\sigma = \mathbf{u}_\sigma - \mathbf{u}$ is the diffusion velocity with regard to the barycentric velocity and $\mathbf{u}_{\alpha(\sigma)} = \left(1-\alpha_\sigma\right) \mathbf{u}_\sigma + \alpha_\sigma \mathbf{u}$ is a linear combination between the specific velocity and the barycentric velocity. Unlike what happens at macroscopic level when the usual BGK equation is considered, in the previous Eq. (10) the relaxation time constants effect the advection term, the viscous term and an internal forcing term, which directly allows us to exchange momentum among the species. In a mesoscopic framework, a strategy for setting the relaxation time constants of the model is needed.

The system of macroscopic equations derived by the usual BGK equation for non-interacting species can be easily recovered by considering $\alpha_\sigma \to 0$. Two cases are possible: $1/\tau_\sigma \to \infty$ and $1/\tau_m \to 0$, but only the second one is allowed because it produces a non-zero viscosity $\alpha_\sigma \tau_m \to \tau_\sigma$. In a similar way, the system of macroscopic equations derived by the single-fluid BGK-like equation for ideally miscible components can be easily recovered by considering $\alpha_\sigma \to 1$. Two cases are possible: $1/\tau_m \to \infty$ and $1/\tau_\sigma \to 0$, but only the second one is allowed because it produces a non-zero viscosity $\alpha_\sigma \tau_m \to \tau_m$. The previous discussion allows us to prove that all the relaxation frequencies of the model must be bounded from above. Let us define $1/\tau_\sigma^0$ and $1/\tau_m^0$ the maximum value for the specific relaxation frequency and for the single-fluid relaxation frequency, respectively. Let us introduce two additional tunable parameters which are defined in the following way:

$$\chi = \frac{1/\tau_\sigma}{1/\tau_\sigma^0}, \tag{11}$$

$$\epsilon = \frac{1/\tau_m}{1/\tau_m^0}. \tag{12}$$

For simplicity, a unique value of the parameter χ for all the species will be considered. In this way, the whole set of relaxation frequencies is uniquely identified by a point $P(\epsilon, \chi)$ on the plane $[0,1] \times [0,1] \subset \mathbb{R}^2$, which will be called Hamel's plane. For example, the point $P(0,1)$ on Hamel's plane identifies mixtures of non-interacting species and implies the following macroscopic momentum equation:

$$\frac{\partial \left(\rho_\sigma \mathbf{u}_\sigma\right)}{\partial t} + \nabla \cdot \left[\rho_\sigma \mathbf{u}_\sigma \otimes \mathbf{u}_\sigma\right] = -\nabla \left(\rho_\sigma e_\sigma\right) + \rho_\sigma \mathbf{g}_\sigma + \nabla \cdot \left[\rho_\sigma e_\sigma \tau_\sigma^0 \left(\nabla \mathbf{u}_\sigma + \nabla \mathbf{u}_\sigma^T\right)\right]. \tag{13}$$

By assuming $\rho_\sigma e_\sigma = p_\sigma$ and $e_\sigma \tau_\sigma^0 = \nu_\sigma$, where p_σ is the partial pressure and ν_σ is the kinematic viscosity for the generic species, the Navier-Stokes equation is recovered. This allows us to identify the value of the internal energy $e_\sigma = p_\sigma/\rho_\sigma$ and the minimum value of the relaxation time $\tau_\sigma^0 = \nu_\sigma/e_\sigma$.

The point $P(1,0)$ on Hamel's plane identifies the mixtures which can be described by the single-fluid approach. In this case, the momentum equation reads:

$$\begin{aligned} &\frac{\partial \left(\rho_\sigma \mathbf{u}_\sigma\right)}{\partial t} + \nabla \cdot \left[\rho_\sigma \mathbf{u} \otimes \mathbf{u} + \rho_\sigma \mathbf{u} \otimes \mathbf{w}_\sigma + \rho_\sigma \mathbf{w}_\sigma \otimes \mathbf{u}\right] = \\ &-\nabla \left(\rho_\sigma e_\sigma\right) + \rho_\sigma \mathbf{g}_\sigma - \frac{1}{\tau_m^0} \rho_\sigma \mathbf{w}_\sigma + \nabla \cdot \left[\rho_\sigma e_\sigma \tau_m^0 \left(\nabla \mathbf{u} + \nabla \mathbf{u}^T\right)\right]. \end{aligned} \tag{14}$$

The identification process is not obvious, because the relaxation time constant τ_m^0 is involved in two different terms: the internal forcing term and the viscous term. Hamel's model has the advantage of highlighting the effects of cross collisions on both diffusion process and effective mixture viscosity. These phenomena can induce coupling among the species, i.e. they force the species velocities to be similar to the barycentric velocity. For this reason, the *force coupling* and the *viscous coupling* can be distinguished at the macroscopic level by considering the respective terms in the momentum equations, but they have the same microscopic origin, i.e. cross collisions.

1. Tuning strategy based on diffusion

Since the internal forcing term is the leading term of the diffusion process, a popular practice, derived from the single-fluid approach [23], consists of relating the relaxation time constant τ_m^0 with the diffusion coefficient. For this reason, it is worthy to analyze the different mechanisms driving the diffusion processes in Hamel's model. Neglecting the inertial effects, the difference between the two Navier-Stokes equations for each species (a and b) leads to the following equation:

$$\frac{1}{\tau_m^0}\,(\mathbf{u}_a-\mathbf{u}_b)=-\frac{\rho^2\,e}{\rho_a\,\rho_b}\,\mathbf{d}+\tau_m^0\,(e_a-e_b)\,\nabla^2\mathbf{u}, \tag{15}$$

where the driving force is:

$$\mathbf{d}=\frac{\rho_a\,\rho_b}{\rho^2\,e}\left[\frac{1}{\rho_a}\,\nabla\,(\rho_a\,e_a)-\frac{1}{\rho_b}\,\nabla\,(\rho_b\,e_b)-(\mathbf{g}_a-\mathbf{g}_b)\right], \tag{16}$$

and $e=\sum_\sigma x_\sigma\,e_\sigma$. The viscous effects in Eq. (15) are usually negligible because it can be assumed that the derivatives are slowly varying with respect to the diffusion process [25, 26]. Making explicit the role of mass concentrations yields

$$\mathbf{d}=\frac{\rho_a\,\rho_b}{\rho^2\,e}\left[\frac{e_a\,\rho}{\rho_a}\,\nabla x_a-\frac{e_b\,\rho}{\rho_b}\,\nabla x_b+(e_a-e_b)\,\frac{1}{\rho}\,\nabla\rho-(\mathbf{g}_a-\mathbf{g}_b)\right]. \tag{17}$$

We can attribute the discrepancy in the species velocities to three different driving mechanisms: the concentration gradients, the pressure gradient (which is proportional to the whole mixture density for ideal gases) by means of different particle masses ($e_a\neq e_b$) and the inequality of the external forces acting on different components. The diffusions driven by these driving mechanisms are called the *ordinary diffusion*, *pressure diffusion* and the *forced diffusion* respectively. Since for binary mixtures $\nabla x_b=-\nabla x_a$, the final expression of the diffusive flux can be recovered

$$\begin{aligned}\rho_a\,\mathbf{w}_a = \;& -\tau_m^0\,(\rho_b\,e_a + \rho_a\,e_b)\,\nabla x_a \\ & +\tau_m^0\,\frac{\rho_a\,\rho_b}{\rho}\,(e_a - e_b)\left(\tau_m^0\,\nabla^2\mathbf{u} - \frac{1}{\rho}\,\nabla\rho\right) + \tau_m^0\,\frac{\rho_a\,\rho_b}{\rho}\,(\mathbf{g}_a - \mathbf{g}_b).\end{aligned} \tag{18}$$

If the cross collision relaxation time constant is set such as $\tau_m^0 = D/(x_b\,e_a + x_a\,e_b)$, then the correct expression of the Fick's first law of diffusion can be recovered, where D is the mutual diffusivity.

Let us consider a fluid flow characterized by a low Reynolds number. In this case, the viscous effects are relevant because it cannot be assumed that the derivatives are slowly varying with regard to the diffusion process. According to Eq. (18) the viscous term leads to an additional diffusive mechanism. However, if we want to model only the ordinary diffusion driven by concentration gradients, then it is possible to consider a different linear combination between the two Navier-Stokes equations for each species (a and b) and to consequently obtain a more convenient expression of the diffusion flux. In particular, the diffusive flux becomes

$$\rho_a\,\mathbf{w}_a = -\tau_m^0\,\frac{e_a\,e_b}{e}\,\rho\,\nabla x_a + \tau_m^0\,\frac{\rho_a\,\rho_b}{\rho}\,\left(\frac{e_b}{e}\,\mathbf{g}_a - \frac{e_a}{e}\,\mathbf{g}_b\right). \tag{19}$$

Obviously the expression given by Eq. (19) is equivalent to that given by Eq. (18). If the external forcing terms are tuned such as $e_b\,\mathbf{g}_a = e_a\,\mathbf{g}_b$, then only ordinary diffusion survives. If the cross collision relaxation time constant is set such as $\tau_m^0 = e\,D^*/(e_a\,e_b)$, then the correct expression of the Fick's first law of diffusion can be also recovered, but a modified mutual diffusivity D^* has been considered.

Once the cross collision relaxation time constant is set in order to reproduce the desired diffusive process, then the mixture viscosity is consequently defined. For high diffusive processes, then $1/\tau_m^0 \to \infty$ and the internal forcing term yields high coupling among the species, i.e. $\mathbf{u}_\sigma \approx \mathbf{u}$. This means also $\tau_m^0 \to 0$, and so only systems characterized by negligible viscous effects can be simulated [32].

In order to fix this problem, the two-fluid approach, based on linearized collisional operator that takes into account cross collisions [25, 26], allows one to tune the internal forcing term independently of the mixture viscosity. However in this case, the mixture viscosity becomes a linear combination of the component viscosities. Unfortunately the mixture viscosity can be a very complex function of the component viscosities [48] due to cross collisions and the linear approximation may be valid only in the simplest cases.

2. *Tuning strategy based on mixture viscosity*

The following tuning strategy for the relaxation time constant τ_m^0 is proposed. By summing the momentum equations for the species (14) and recalling the definition of barycentric velocity, the momentum equation for the mixture is recovered:

$$\frac{\partial (\rho \, \mathbf{u})}{\partial t} + \nabla \cdot (\rho \, \mathbf{u} \otimes \mathbf{u}) = -\nabla (\rho \, e) + \rho \, \mathbf{g} + \nabla \cdot \left[\rho \, e \, \tau_m^0 \left(\nabla \mathbf{u} + \nabla \mathbf{u}^T \right) \right], \tag{20}$$

where $\rho = \sum_\sigma \rho_\sigma$ is the mixture density, $e = \sum_\sigma x_\sigma \, e_\sigma = p/\rho$ is the mixture internal energy, $p = \sum_\sigma p_\sigma$ is the mixture pressure and $\mathbf{g} = \sum_\sigma x_\sigma \, \mathbf{g}_\sigma$ is the mass averaged effect of the external field. Recovering the Navier-Stokes momentum equation for the mixture requires the minimum value of the cross-collision relaxation time constants to be $\tau_m^0 = \nu_m/e$, where ν_m is the mixture kinematic viscosity for fully coupled configurations, i.e. when cross collisions prevail. According to the mesoscopic framework, this strategy allows us to recover any experimental mixture viscosity instead of conjecturing a simplified value based on the component viscosities.

Once the cross-collision relaxation time constant is set in order to reproduce the desired mixture viscosity, then the diffusive process is consequently defined. It is possible to quantify this effect by means of the Schmidt number which is the ratio between kinematic viscosity and mutual diffusivity. The Schmidt number can be considered analogous to the Prandtl number for mass transfer and obviously depends on the adopted definition of mutual diffusivity. Assuming that $D = \tau_m^0 \, (x_b \, e_a + x_a \, e_b)$ because the viscous effects are negligible and spatial derivatives are slowly varying with regard to the diffusion process, the Schmidt number is $Sc_0 = e/(x_b \, e_a + x_a \, e_b)$ or, if $D^* = \tau_m^0 \, (e_a \, e_b)/e$ is assumed in the low Reynolds number limit, it becomes $Sc_0^* = D^*/\nu_m = e^2/(e_a \, e_b)$. Hamel's model in both cases implies a fixed value of the Schmidt number because both diffusion and mixture viscosity depend on the same cross-collision relaxation time constant τ_m. This feature will be better discussed later on, concerning neither fully-decoupled nor ideally coupled intermediate configurations and linearized models.

Unfortunately there is no proof that this strategy of setting the relaxation time constants ensures the ideal coupling among the species, i.e. $\mathbf{u}_\sigma \approx \mathbf{u}$, as it should be expected. Moreover, the behavior of the model for mixtures which cannot be considered neither fully-decoupled nor ideally coupled, implies for $0 < \alpha_\sigma < 1$.

Let us consider a given number of different mixtures in the same isothermal condition: each mixture is made by components which share the same characteristics in terms of mass concentrations, molecular weights, kinematic viscosities and only differ in terms of diffusivity, that is, coupling strength. Equivalently, let us analyze a given mixture in isothermal conditions but for different temperature values and suppose that temperature effects diffusivity more than what happens for other thermo-physical properties. In both cases, it is possible to exclusively vary the coupling strength among the species. On Hamel's plane, we can smoothly move from the fully-decoupled configuration $P(0,1)$ to the ideally diffusive configuration $P(1,0)$ and the set of the intermediate points $P_H(\chi_H,\epsilon)$ defines a curve. If we arbitrarily adopt the parameter ϵ as an index of the coupling strength, this means that some function $\chi_H(\epsilon)$ which smoothly describes the intermediate configurations, such that $\chi_H(0) = 1$ and $\chi_H(1) = 0$, will exist. The function $\chi_H(\epsilon)$ will be called Hamel's function and determines the behavior of the kinetic model by means of the function α_σ involved in Eq. (10). This function can be reformulated by means of the new variables:

$$\alpha_\sigma(\epsilon) = \frac{\epsilon\,\gamma_\sigma}{\chi_H + \epsilon\,\gamma_\sigma}, \tag{21}$$

where $\gamma_\sigma = (x_\sigma\,\nu_\sigma)/(y_\sigma\,\nu_m)$ and y_σ is the volume concentration for the generic species, defined as $y_\sigma = p_\sigma/\sum_\sigma p_\sigma$.

In this paper, the tuning strategy based on mixture viscosity will be assumed to better highlight the features of viscous coupling. Particularly in the next section, Hamel's function will be calculated for controlling the effective viscosity of the mixture at any intermediate coupling strength.

B. Infinitely long channel

For simplicity, let us consider an infinitely long channel in the x-direction (y identifies the transverse direction). When a single fluid realizes a laminar flow through it, the typical conditions of the Poiseuille flow are recovered. A binary mixture will be considered in the following. This test problem because of its simplicity allows us to find a general expression for Hamel's function: the effectiveness of this result will be verified by numerical simulations for two dimensional domains. In the low Mach number limit, the inertial effects described by the left hand side of Eq. (10) can be neglected. In the same limit, the velocity field is essentially solenoidal (divergence free) and the effects due to the pressure gradient are also negligible, when ideal gases are considered. Under these hypotheses Eq. (10) becomes:

$$\mathbf{V}\,\frac{\partial^2\,\mathbf{u}^x}{\partial\,y^2} = \epsilon\,\mathbf{F}\,\mathbf{u}^x - \mathbf{a}^x, \tag{22}$$

where $\mathbf{V}$ is the viscosity matrix, $\mathbf{F}$ is the matrix which describes the internal force coupling, $\mathbf{u}^x = [\,u_a^x, u_b^x\,]^T$ is a vector collecting the x components of the specific velocities and

$$\mathbf{a}^x = \begin{bmatrix} -e_a\,\partial\rho_a/\partial x + \rho_a\,g_a^x \\ -e_b\,\partial\rho_b/\partial x + \rho_b\,g_b^x \end{bmatrix}. \tag{23}$$

The elements of the viscosity matrix are:

$$V_{ij}(\epsilon) = \rho_i\,\nu_i\,\frac{\chi_H\,\delta_{ij} + \epsilon\,\gamma_i\,x_j}{[\,\chi_H + \epsilon\,\gamma_i\,]^2}, \tag{24}$$

where i and j stand for species labels, i.e. a or b. The matrix which describes the internal force coupling is:

$$\mathbf{F} = x_a\,x_b\,\frac{\rho\,e}{\nu_m}\begin{bmatrix} +1 & -1 \\ -1 & +1 \end{bmatrix}. \tag{25}$$

The *force coupling* can be defined internally because $det\,(\mathbf{F}) = 0$. Analyzing the solutions of the previous system, given by the Eq. (22), let us discuss the determinant of the matrix $\mathbf{V}$:

$$det\,(\mathbf{V}) = \frac{\rho_a\,\nu_a\,\rho_b\,\nu_b}{[\,\chi_H + \epsilon\,\gamma_a\,]^2\,[\,\chi_H + \epsilon\,\gamma_b\,]^2}\,\left[\chi_H^2 + 2\,\chi_H\,\epsilon\,(\,x_a\,\gamma_a + x_b\,\gamma_b\,)\right] \geq 0. \tag{26}$$

In particular for any $\epsilon \in [0, 1)$, the determinant is positive and the inverse matrix $\mathbf{V}^{-1}$ exists. For this reason, Eq. (22) can be rewritten in the following way:

$$\frac{\partial^2\,\mathbf{u}^x}{\partial\,y^2} = \epsilon\,\mathbf{V}^{-1}\,\mathbf{F}\,\mathbf{u}^x - \mathbf{V}^{-1}\,\mathbf{a}^x, \tag{27}$$

This system of equations can be diagonalized, resulting in:

$$\frac{\partial^2\,\hat{\mathbf{u}}^x}{\partial\,y^2} = \epsilon\,\mathbf{D}\,\hat{\mathbf{u}}^x - \mathbf{b}^x, \tag{28}$$

where $\mathbf{D} = \mathbf{E}^{-1}(\mathbf{V}^{-1}\,\mathbf{F})\,\mathbf{E}$ is the diagonal matrix formed by the eigenvalues of the matrix $(\mathbf{V}^{-1}\,\mathbf{F})$, $\mathbf{E}$ is the matrix formed by the columns of the right eigenvectors of the matrix $(\mathbf{V}^{-1}\,\mathbf{F})$ and $\mathbf{b}^x = \mathbf{E}^{-1}\mathbf{V}^{-1}\,\mathbf{a}^x$ is the modified forcing term. Independently from the mixture properties, the system of equations is characterized by a null eigenvalue and a positive eigenvalue: let us suppose $D_{11} = 0$ and $D_{22} \geq 0$. The equation which corresponds to the null eigenvalue is:

$$\frac{\partial^2 \hat{u}_1^x}{\partial y^2} = -\frac{a_a^x + a_b^x}{\rho\, \nu_c} < 0, \tag{29}$$

where $\hat{u}_1^x$ and ν_c are defined as

$$\hat{u}_1^x(\epsilon) = \frac{V_{11}(\epsilon) + V_{21}(\epsilon)}{\rho\, \nu_c(\epsilon)}\, u_a^x + \frac{V_{12}(\epsilon) + V_{22}(\epsilon)}{\rho\, \nu_c(\epsilon)}\, u_b^x, \tag{30}$$

$$\nu_c(\epsilon) = \frac{x_a\, \nu_a}{\chi_H + \epsilon\, \gamma_a} + \frac{x_b\, \nu_b}{\chi_H + \epsilon\, \gamma_b}. \tag{31}$$

The equation which corresponds to the positive eigenvalue is:

$$\frac{\partial^2 \hat{u}_2^x}{\partial y^2} = \epsilon\, x_a\, x_b\, \frac{\rho\, e}{\nu_m}\, \frac{\rho\, \nu_c}{det\,(\mathbf{V})}\, \hat{u}_2^x - \frac{V_{12}(\epsilon) + V_{22}(\epsilon)}{\rho\, \nu_c(\epsilon)}\, \frac{1}{det\,(\mathbf{V})} \left(\frac{\rho_b\, \nu_b\, a_a^x}{\chi_H + \epsilon\, \gamma_b} - \frac{\rho_a\, \nu_a\, a_b^x}{\chi_H + \epsilon\, \gamma_a} \right). \tag{32}$$

where $\hat{u}_2^x$ is defined as

$$\hat{u}_2^x(\epsilon) = \frac{V_{12}(\epsilon) + V_{22}(\epsilon)}{\rho\, \nu_c(\epsilon)}\, (\, u_a^x - u_b^x\,)\, . \tag{33}$$

Equation (29) admits parabolic solutions with negative curvature regardless of the mixtures properties. On the other hand, Eq. (32) admits parabolic solutions if and only if the components of the mixture do not interact with each other ($\epsilon = 0$). In this case, the physical situation is exactly the same as in Poiseuille flow: the viscous matrix $\mathbf{V}$ is diagonal and the solutions in terms of the original variables u_a^x and u_b^x will be also parabolic, because they come from linear combinations of the diagonalized variables $\hat{u}_1^x(0)$ and $\hat{u}_2^x(0)$. In particular, the diagonalized velocity $\hat{u}_1^x(0) = z_a\, u_a^x + z_b\, u_b^x$ reduces to the viscous velocity for the mixture $\mathbf{u}_\nu$:

$$\mathbf{u}_\nu = \sum_\sigma z_\sigma\, \mathbf{u}_\sigma, \tag{34}$$

where $z_\sigma = (x_\sigma\, \nu_\sigma) / \sum_\sigma (x_\sigma\, \nu_\sigma)$. In the general case $\epsilon \neq 0$, the coupling among species introduces exponential solutions because the coefficient multiplying $\hat{u}_2^x$ on the right hand side of Eq. (32) is strictly positive. Hence the internal *force coupling* changes the nature of the solutions.

How the proposed model describes the diffusion process can be analyzed by considering the limiting case $\epsilon \to 1$. Substituting the definition given by Eq. (33) into Eq. (32) yields

$$\frac{\partial^2 \hat{u}_2^x}{\partial y^2} = \frac{V_{12}(\epsilon) + V_{22}(\epsilon)}{\rho\, \nu_c(\epsilon)}\, \frac{\rho_a\, \rho_b}{det\,(\mathbf{V})} \left[\epsilon\, \frac{\nu_c(\epsilon)\, e}{\nu_m}\, (\, u_a^x - u_b^x\,) - \left(\frac{\nu_b\, a_a^x / \rho_a}{\chi_H + \epsilon\, \gamma_b} - \frac{\nu_a\, a_b^x / \rho_b}{\chi_H + \epsilon\, \gamma_a} \right) \right]. \tag{35}$$

If $\epsilon \rightarrow 1$, then $det\,(\mathbf{V}) \rightarrow 0$ and this means that the right hand side of the previous equation tends to prevail over the second order spatial derivative. In this case, it is easy to derive an expression for the difference between the species velocities $u_a^x - u_b^x$, namely

$$u_a^x - u_b^x = \frac{\nu_m}{\epsilon\, e} \left[-\frac{e_a\, e_b}{e} \frac{\rho^2}{\rho_a\, \rho_b} \frac{\partial}{\partial x} \left(\frac{\rho_a}{\rho} \right) + \left(\frac{e_b}{e}\, g_a^x - \frac{e_a}{e}\, g_b^x \right) \right]. \tag{36}$$

The previous difference of specific velocities can be used to derive the diffusion equation for the concentration $x_a = \rho_a/\rho$. Recalling the continuity equation for the generic species given by Eq. (9), the following equation holds in the low Mach number limit

$$\frac{\partial x_a}{\partial t} + u^x \frac{\partial x_a}{\partial x} = -\frac{\partial}{\partial x} \left[\frac{\rho_a\, \rho_b}{\rho^2} \, (u_a^x - u_b^x) \right]. \tag{37}$$

Substituting the difference of the specific velocities given by Eq. (36) in the previous equation, the final expression of the diffusion equation is recovered, namely

$$\frac{\partial\, x_a}{\partial t} + u^x \, \frac{\partial\, x_a}{\partial x} = D_c^* \, \frac{\partial^2\, x_a}{\partial x^2}, \tag{38}$$

where $D_c^*(\epsilon) = \nu_m\, e_a\, e_b/(\epsilon\, e^2)$ is the modified mutual diffusivity. This equation will be used later on in order to numerically measure the diffusivity of the discrete lattice Boltzmann model.

Finally, the particular case of $\epsilon = 1$ must be discussed. The system of equations is singular and a solution may exist if and only if the forcing term satisfies a compatibility condition. Considering $\epsilon = 1$ in the system (22), neglecting the concentration gradients and applying the difference between the equations of the system, the compatibility condition is obtained:

$$x_a\, x_b\, \frac{e}{\nu_m} \, (\, u_a^x - u_b^x \,) = x_a\, y_b\, g_a^x - x_b\, y_a\, g_b^x. \tag{39}$$

The previous result is compatible with Eq. (36), if the species density is spatially homogeneous, i.e. $\partial \rho_\sigma / \partial x = 0$. If the forcing terms due to the external field are such as $x_a\, y_b\, g_a^x = x_b\, y_a\, g_b^x$, then the solution of the system of equations is unique, i.e. $u_a^x = u_b^x$. Let us model a mixture affected by a given forcing term $\rho\, g^x$. This forcing term acts as a source term in the mixture momentum equation (20). In the porous media simulations, it is quite usual to describe the effects of the pressure gradient as a forcing term. For the mixtures, where only the total value of the pressure gradient is known, the splitting of the forcing term among the momentum equations for the components can be made by means of the previous compatibility condition. If the source term for a generic species is called

$\rho_\sigma\, g_\sigma^x$, then the compatibility condition prescribes that $\rho_\sigma\, g_\sigma^x = y_\sigma \rho\, g^x$, i.e. the splitting of the forcing term must be made on the basis of the volume concentrations.

An important feature of the proposed kinetic model is that the model allows us to tune the determinant of the viscous matrix $\mathbf{V}$ by means of the coupling strength ϵ. For this reason, the mixtures characterized by ideally miscible components can be very easily described by setting $\epsilon = 1$, i.e. a finite value. In the usual two-fluid models, the ideally miscible configuration is an asymptotic limiting case, which only in principle can be recovered by increasing the coupling force [26]. For the lattice Boltzmann models, some stability constraints exist which do not allow us to consider forcing terms too large and this makes the usual way of recovering the ideally miscible configuration actually impracticable. Since the proposed model simulates the coupling among species by means of the viscous matrix $\mathbf{V}$ more than by means of the force matrix $\mathbf{F}$, it will be called a *viscous coupling* based model.

The previous discussion suggests a way to calculate Hamel's function. For intermediate coupling strengths, the diagonalized velocity $\hat{u}_1^x(\epsilon)$ can be expressed in the following way:

$$\hat{u}_1^x(\epsilon) = u_\nu^x + \left[\frac{V_{11}(\epsilon) + V_{21}(\epsilon)}{\rho\, \nu_c(\epsilon)} - z_a \right] (\, u_a^x - u_b^x \,)\,. \tag{40}$$

When the difference among the species velocities is large, the components of the mixture are characterized by weak interactions and the diagonalized velocity is equivalent to the mixture viscous velocity $\hat{u}_1^x(\epsilon) \approx \hat{u}_1^x(0) = \hat{u}_1^x(1) = u_\nu^x$, as previously discussed for the Poiseuille flow. For strong interactions among species, the components velocities essentially become the same and all the possible velocity averages produce the same result. In both cases, the second term involved in the right hand side of Eq. (40) is negligible for different reasons. This suggests considering the approximation $\hat{u}_1^x(\epsilon) \approx u_\nu^x$ acceptable for any coupling strength. In this way, Eq. (29) becomes

$$\frac{\partial^2\, u_\nu^x}{\partial\, y^2} = -\frac{a_a^x + a_b^x}{\rho\, \nu_c} < 0, \tag{41}$$

and this means that the mixture viscous velocity is substantially affected by the critical viscosity, given by Eq. (31). This equation will be used later on to numerically measure the mixture kinematic viscosity of the discrete lattice Boltzmann model. The proper value of the critical viscosity can be tuned by means of Hamel's function to reproduce the experimental data for the mixture's viscous velocity. It is reasonable to assume that, for an intermediate coupling strength, the critical viscosity belongs to the range defined by the mass averaged viscosity $\nu_c(0) = \sum_\sigma x_\sigma \nu_\sigma$ and by the mixture viscosity for the ideally miscible configuration

$\nu_c(1) = \nu_m$. Since Hamel's function is bounded $0 \leq \chi_H \leq 1$, some constraints exist for the way to connect the previous values of critical viscosity. In particular, for the linear strategy,

$$\nu_c(\epsilon) = \sum_\sigma \frac{x_\sigma\, \nu_\sigma}{\chi_H + \epsilon\, \gamma_\sigma} = (1-\epsilon) \sum_\sigma x_\sigma \nu_\sigma + \epsilon\, \nu_m, \tag{42}$$

is allowed for any configuration such as $\nu_m \geq 1/2 \sum_\sigma x_\sigma\, \nu_\sigma$. The previous constraint can be easily verified by the fact that the upper bound of the critical viscosity is $\nu_c^{max} = \nu_m/\epsilon$. This imposes a maximum rate of change for the critical viscosity, when ideally miscible components are considered. Anyway a connecting path always exists, but for $\nu_m \leq 1/2 \sum_\sigma x_\sigma\, \nu_\sigma$ it is no longer linear. The condition (42) allows us to calculate Hamel's function, which is the last parameter needed to define the kinetic model.

According to the previous tuning strategy and considering the low Reynolds number limit, the Schmidt number is

$$Sc^* = \frac{\nu_c(\epsilon)}{D_c^*(\epsilon)} = \frac{e^2}{e_a\, e_b} \left[\epsilon\, (1-\epsilon) \frac{\sum_\sigma x_\sigma\, \nu_\sigma}{\nu_m} + \epsilon^2 \right]. \tag{43}$$

It is easy to prove that the previous result is consistent with the ideally miscible configuration: in fact if $\epsilon = 1$, then $Sc^* = Sc_0^* = e^2/(e_a\, e_b)$. In the next section, this model will be compared with the linearized models.

C. Linearized kinetic models

The main difficulty of Hamel's model is due to the fact that the zero-order approximation of the velocity distribution function is a linear combination of Maxwellian functions, which, in general, is not a Maxwellian itself (see Appendix A). This is a direct consequence of the fact that, in the simplified collisional operator, two different Maxwellian distribution functions are involved. If the species velocity $\mathbf{u}_\sigma$ does not differ too much from the barycentric velocity $\mathbf{u}$, this mathematical complication is not needed and it can be avoided by means of an asymptotic approximation. It is important to point out that, in principle, a linearized model can be considered valid only for configurations close to the constitutive hypotheses used to derive it: in the case, that the cross collisions are so relevant to force the species velocity to be close to the barycentric velocity.

Equation (3) can be recast in the following form:

$$\frac{\partial f_\sigma}{\partial t} + \mathbf{v} \cdot \nabla f_\sigma + \mathbf{g}_\sigma \cdot \nabla_{\mathbf{v}} f_\sigma = -\frac{1}{\alpha_\sigma\, \tau_m} \left[f_\sigma - f_\sigma^e \right] - \frac{1}{\tau_m} \left[f_\sigma^e - f_{\sigma(m)}^e \right]. \tag{44}$$

In order to simplify the last term on the right hand side of the previous equation, it is possible to expand f_σ^e around $f_{\sigma(m)}^e$ or, equivalently, to expand $f_{\sigma(m)}^e$ around f_σ^e, in the limiting case that the specific velocity and the barycentric velocity are sufficiently similar. The asymptotic formulas are:

$$f_\sigma^e = f_{\sigma(m)}^e + \frac{f_{\sigma(m)}^e}{e_\sigma} (\mathbf{v} - \mathbf{u}) \cdot (\mathbf{u}_\sigma - \mathbf{u}) + O\left(|\mathbf{u}|^3\right), \tag{45}$$

and

$$f_{\sigma(m)}^e = f_\sigma^e + \frac{f_\sigma^e}{e_\sigma} (\mathbf{v} - \mathbf{u}_\sigma) \cdot (\mathbf{u} - \mathbf{u}_\sigma) + O\left(|\mathbf{u}|^3\right). \tag{46}$$

Neglecting the higher order terms and considering a linear combination of the previous formulas by means of a dimensionless parameter $0 \leq \beta \leq 1$, a set of approximations for the difference between the Maxwellian distribution functions can be obtained:

$$f_\sigma^e - f_{\sigma(m)}^e \approx \left[\beta \, \frac{f_{\sigma(m)}}{e_\sigma} (\mathbf{v} - \mathbf{u}) + (1-\beta) \, \frac{f_\sigma^e}{e_\sigma} (\mathbf{v} - \mathbf{u}_\sigma)\right] \cdot (\mathbf{u}_\sigma - \mathbf{u}). \tag{47}$$

Substituting the previous approximation in Eq. (44) yields:

$$\begin{aligned} \frac{\partial f_\sigma}{\partial t} \; + \; \mathbf{v} \cdot \nabla f_\sigma + \mathbf{g}_\sigma \cdot \nabla_{\mathbf{v}} f_\sigma &= -\frac{1}{\alpha_\sigma \, \tau_m} \left[f_\sigma - f_\sigma^e\right] \\ - \; \frac{f_{\sigma(m)}^e}{e_\sigma} \, \frac{\beta}{\tau_m} \, (\mathbf{v} - \mathbf{u}) \cdot \mathbf{w}_\sigma &- \frac{f_\sigma^e}{e_\sigma} \, \frac{1-\beta}{\tau_m} \, (\mathbf{v} - \mathbf{u}_\sigma) \cdot \mathbf{w}_\sigma \end{aligned} \tag{48}$$

The kinetic model originally proposed by Sirovich can be recovered if $\beta = 0$ [39]. The additional terms in Eq. (48) do not effect the zero-order approximation of the distribution function involved in the asymptotic analysis. For this reason, although some coupling among species exists, the zero order approximation of the distribution function is still Maxwellian. Essentially, the additional terms are similar to the terms which appear in the Chapman-Enskog asymptotic analysis when the external force field is considered: see Eq. (A9) in Appendix A. It is well known that only the moments of the forcing term up to the second order are involved in the previous analysis. In particular, all the approximations (47) produce the same results for both zero and first-order moments, while they differ for the second-order moments. Let us consider, for example, the following second-order moment:

$$\begin{aligned} \int m_\sigma \, \mathbf{v} \otimes \mathbf{v} \, \left[f_\sigma^e - f_{\sigma(m)}^e\right] \, d\mathbf{v} \; \approx \; & \rho_\sigma \left[\, 2\,(1-\beta)\, \mathbf{u}_\sigma \otimes \mathbf{u}_\sigma - 2\beta \, \mathbf{u} \otimes \mathbf{u} \right. \\ & \left. + \, (2\beta - 1)\, (\mathbf{u}_\sigma \otimes \mathbf{u} + \mathbf{u} \otimes \mathbf{u}_\sigma)\right]. \end{aligned} \tag{49}$$

The result due to the original Maxwellian distribution functions can be recovered, if and only if the approximation characterized by $\beta = 1/2$ is considered. In fact, it is well known that the central difference approximation ($\beta = 1/2$) produces better results than one-side approximations ($\beta = 1$ or $\beta = 0$, where the last is considered by Sirovich's model). It is possible to conclude that Sirovich's model considers only one possible approximation, which is not the most accurate.

The Chapman-Enskog asymptotic analysis of the linearized models can be easily performed by analogy with the analysis of Hamel's model (see Appendix A). The continuity equation for each species is the same of Hamel's model and it is described by Eq. (9). The momentum equation is:

$$\begin{aligned}\frac{\partial \left(\rho_\sigma \mathbf{u}_\sigma\right)}{\partial t} &+ \nabla\cdot\left(\rho_\sigma \mathbf{u}_\sigma \otimes \mathbf{u}_\sigma + 2\beta\, \alpha_\sigma \rho_\sigma \mathbf{w}_\sigma \otimes \mathbf{w}_\sigma\right) = -\nabla\left(e_\sigma \rho_\sigma\right) + \rho_\sigma\, \mathbf{g}_\sigma \\ &- \frac{1}{\tau_m}\,\rho_\sigma \mathbf{w}_\sigma + \nabla\cdot\left[\alpha_\sigma\, \rho_\sigma\, e_\sigma\, \tau_m\, \left(\nabla \mathbf{u}_\sigma + \nabla \mathbf{u}_\sigma^T\right)\right]. \end{aligned} \tag{50}$$

All the linearized models produce a coupling force proportional to the diffusion velocity $\mathbf{w}_\sigma$. In some linearized models ($\beta \neq 0$), the diffusion velocity can effect the advection term but in none of them the diffusion velocity can effect the viscous term, which is usually the leading term in the low Mach number limit. If we consider once more the infinitely deep channel discussed in the previous section and in particular Eq. (22), all the linearized models are characterized by a diagonal viscous matrix $\mathbf{V}$. This means that the determinant of the viscous matrix is always strictly positive and *viscous coupling* is not possible.

Again, the problem is finding a method for correlating the microscopic relaxation time constants with the macroscopic transport coefficients. The most usual strategy will be discussed [25, 26]. First of all, if the specific relaxation time constant τ_σ is set in such a way that:

$$\tau_\sigma = \frac{\nu_\sigma\, \tau_m}{e_\sigma\, \tau_m - \nu_\sigma}, \tag{51}$$

then the component viscosity is decoupled by the diffusion process because $\alpha_\sigma\, e_\sigma\, \tau_m = \nu_\sigma$ and Eq. (50) exactly recovers the Navier-Stokes momentum equation. Negative values of the relaxation time constants τ_σ are possible because in the linearized kinetic equation (44) only the quantity $\alpha_\sigma\, \tau_m$ is involved. The cross-collision relaxation time constant τ_m can be tuned according to the mutual diffusion coefficient. The difference between the two Navier-Stokes equations for each species (a and b) leads to the following equation:

$$\frac{1}{\tau_m}\,\left(\mathbf{u}_a - \mathbf{u}_b\right) = -\frac{\rho^2\, e}{\rho_a\, \rho_b}\,\mathbf{d} + \left(\nu_a\, \nabla^2 \mathbf{u}_a - \nu_b\, \nabla^2 \mathbf{u}_b\right), \tag{52}$$

where the inertial effects have been neglected and the driving force $\mathbf{d}$ is given by Eq. (16). If the cross-collision relaxation time constant is set in such a way that

$$\tau_m = \frac{D}{x_b\, e_a + x_a\, e_b}, \tag{53}$$

where D is the mutual diffusion coefficient, then the expression for the velocity difference is recovered [4]:

$$\mathbf{u}_a - \mathbf{u}_b = -\frac{D\,\rho^2}{\rho_a\,\rho_b}\,\nabla x_a - \frac{D}{\rho_b\, e_a + \rho_a\, e_b}\,\left[(e_a - e_b)\,\nabla\rho + \rho\,(\mathbf{g}_a - \mathbf{g}_b) + \rho\,\left(\nu_a\,\nabla^2\mathbf{u}_a - \nu_b\,\nabla^2\mathbf{u}_b\right)\right]. \tag{54}$$

It is easy to verify that the linearized models are characterized by all the driving mechanisms, which have been previously discussed for Hamel's model: the concentration gradient, the pressure gradient by means of different particle masses ($e_a \neq e_b$), the inequality of the external forces acting on different components and, finally, the inequality of the viscous effects, if they are not negligible.

In the low Reynolds number limit, it is more convenient to consider a different linear combination of the two Navier-Stokes equations for each species (a and b), in order to obtain the following equation:

$$\frac{1}{\tau_m}\,(\mathbf{u}_a - \mathbf{u}_b) = -\frac{e_a\,e_b}{e}\,\frac{\rho^2}{\rho_a\,\rho_b}\,\nabla x_a + \left(\frac{e_b}{e}\,\mathbf{g}_a - \frac{e_a}{e}\,\mathbf{g}_b\right). \tag{55}$$

If the external forcing terms are tuned in such a way that $e_b\,\mathbf{g}_a = e_a\,\mathbf{g}_b$ and cross-collision relaxation time constant is

$$\tau_m = \frac{D^*\,e}{e_a\,e_b}, \tag{56}$$

then the Fick's first law of diffusion can be recovered, but a modified mutual diffusivity D^* has been considered. Consequently, in the low Reynolds number limit, the Schmidt number is

$$Sc^* = \frac{e\,\sum_\sigma x_\sigma\,\nu_\sigma}{\tau_m\,e_a\,e_b}, \tag{57}$$

and it can be freely tuned by setting the cross-collision relaxation time constant. This is probably the best advantage of linearized models [25, 26].

However, the usual strategy for setting the relaxation time constants has a drawback. When very high coupling strengths are considered, it is possible to assume $\mathbf{u}_\sigma \approx \mathbf{u}$. In this case, summing the species momentum equations (50), it is possible to obtain the momentum equation for the barycentric velocity and it is easy to verify that the value of the mixture viscosity for ideally miscible components coincides with the mass averaged viscosity $\sum_\sigma x_\sigma \nu_\sigma$. From the experimental point of view, this formula is valid as a first-order approximation: actually the mixture viscosity can be a very complicated function of the mixture properties [48]. Also in this case, the problem can be solved by modifying the strategy for setting the relaxation time constants, as previously done for Hamel's model. In this way, the mixture viscosity becomes a tunable parameter, but this is not sufficient to describe the ideally miscible configuration because the viscous matrix, involved in Eq. (22), is always non-singular.

Since the viscous coupling for the linearized models is not possible, the ideally miscible configuration, i.e. when $\mathbf{u}_\sigma$ does not differ too much from the barycentric velocity $\mathbf{u}$, can be recovered if and only if the the coupling force is infinitely large, namely $1/\tau_m \to \infty$. From the practical point of view, the numerical stability constraints do not allow us to simulate an infinitely large coupling force and for this reason the ideally miscible configuration can not be practically recovered. On the other hand, in the complete Hamel's model it is sufficient to assume the maximum coupling strength, i.e. $\epsilon = 1$, to exactly ensure the ideally miscible configuration. This result proves that the *viscous coupling* is more effective than the *force coupling* in reproducing the ideally miscible configuration because a lower (finite) value of the coupling strength is enough to reproduce the single-fluid approach. In the next section, the lattice Boltzmann version of Hamel's model is constructed.

III. LATTICE BOLTZMANN MODEL FOR BINARY MIXTURES

In the following section, a lattice Boltzmann model for binary mixtures based on Hamel's model, defined by the Eqs. (3, 4, 5, 8), is constructed. The discrete ordinate method (DOM) is commonly utilized for solving the integro-differential equations involved for modeling the rarefied gas flows [1] and it will be adopted in this case to construct the lattice Boltzmann model [34]. According to this method, a set of discrete microscopic velocities $\mathbf{v}^\lambda$ must be defined and the velocity distribution function will be exclusively evaluated for the selected velocities. This set of microscopic velocities is called *lattice* and it is usually defined without fixing the magnitude of the microscopic velocities, which is a tunable parameter. In the proposed model, a square lattice (D2Q9) for a two-dimensional computational domain, which makes use of nine discrete velocities, is considered [6]. The lattice discrete velocities are defined in the following way:

$$\mathbf{v}^\lambda = \begin{cases} c\ [0,\, 0] & \lambda = 0 \\ c\ [\cos(\lambda\,\pi/2 - \pi/2),\, \sin(\lambda\,\pi/2 - \pi/2)] & \lambda = 1,2,3,4 \\ \sqrt{2}\,c\ [\cos(\lambda\,\pi/2 - 9\,\pi/4),\, \sin(\lambda\,\pi/2 - 9\,\pi/4)] & \lambda = 5,6,7,8 \end{cases}, \tag{58}$$

where c is a tunable parameter which is called lattice velocity. Since only the discrete velocities are allowed, the problem reduces to computing the generic discretized distribution function f_σ^λ, which is essentially the value of the velocity distribution function when the λ-th discrete velocity is considered $f_\sigma^\lambda(t,\mathbf{x}) = f_\sigma(t,\mathbf{x},\mathbf{v}^\lambda)$. In this way, the original kinetic equation, which is an integro-differential equation, reduces to a system of differential equations:

$$\frac{\partial f_\sigma^\lambda}{\partial t} + \mathbf{v}^\lambda \cdot \nabla f_\sigma^\lambda + \mathbf{g}_\sigma \cdot \nabla_{\mathbf{v}} f_\sigma^\lambda = -\frac{\chi_H}{\tau_\sigma^0}\left[f_\sigma^\lambda - f_\sigma^{e\,\lambda}\right] - \frac{\epsilon}{\tau_m^0}\left[f_\sigma^\lambda - f_{\sigma(m)}^{e\,\lambda}\right], \tag{59}$$

for any $0 \le \lambda \le 8$. The kinetic term which takes into account the effects of the external force field can be simplified. This practice is based on the fact that the non-equilibrium distribution function does not differ too much from the equilibrium distribution with regard to the microscopic velocity, in the fluid regime limit [33]. In this way, the following approximation can be adopted:

$$-\nabla_{\mathbf{v}} f_\sigma^\lambda \approx -\nabla_{\mathbf{v}} f_\sigma^{e\,\lambda} = (1-\alpha_\sigma)\,\frac{f_\sigma^{e\,\lambda}}{e_\sigma}\left(\mathbf{v}^\lambda - \mathbf{u}_\sigma\right) + \alpha_\sigma \frac{f_{\sigma(m)}^{e\,\lambda}}{e_\sigma}\left(\mathbf{v}^\lambda - \mathbf{u}\right). \tag{60}$$

Substituting the previous approximation in the equation for the discretized distribution function yields:

$$\begin{aligned} \frac{\partial f_\sigma^\lambda}{\partial t} \;&+\; \mathbf{v}^\lambda \cdot \nabla f_\sigma^\lambda = -\frac{\chi_H}{\tau_\sigma^0}\left[f_\sigma^\lambda - f_\sigma^{e\,\lambda}\right] - \frac{\epsilon}{\tau_m^0}\left[f_\sigma^\lambda - f_{\sigma(m)}^{e\,\lambda}\right] \\ &+\; (1-\alpha_\sigma)\,\frac{f_\sigma^{e\,\lambda}}{e_\sigma}\left(\mathbf{v}^\lambda - \mathbf{u}_\sigma\right)\cdot \mathbf{g}_\sigma + \alpha_\sigma \frac{f_{\sigma(m)}^{e\,\lambda}}{e_\sigma}\left(\mathbf{v}^\lambda - \mathbf{u}\right)\cdot \mathbf{g}_\sigma. \end{aligned} \tag{61}$$

Since only the discrete distribution functions for the lattice microscopic velocities are considered, an interpolation test function must be adopted to calculate the macroscopic moments. The key idea is to reduce the statistical moments of the continuous distribution function to weighted summations of the discretized distribution functions by means of proper quadrature formulas. The interpolation test function should be chosen in such a way as to include the equilibrium distribution function as a particular case, in order to allow us to recover the equilibrium conditions. The problem is that the equilibrium distribution function is an exponential function, while the moments are polynomial forms of the macroscopic quantities. This mismatch can be easily overcome by continuous integration but not by a quadrature formula, which cannot change the nature of the interpolation test function. For this reason the equilibrium distribution function must be also approximated with a polynomial form. If the low Mach number limit is considered, then the equilibrium distribution function can be linearized around the state at rest. For the Maxwellian distribution function centered on the specific velocity, this approximation yields:

$$f_\sigma^{e\,\lambda} \approx \frac{\rho_\sigma}{m_\sigma\,(2\pi e_\sigma)^{D/2}} \exp\left[-\frac{(\mathbf{v}^\lambda)^2}{2\,e_\sigma}\right] \left[1+\frac{\mathbf{v}^\lambda\cdot\mathbf{u}_\sigma}{e_\sigma}+\frac{\left(\mathbf{v}^\lambda\cdot\mathbf{u}_\sigma\right)^2}{2\,e_\sigma^2}-\frac{\mathbf{u}_\sigma^2}{2\,e_\sigma}\right], \tag{62}$$

and similarly for the Maxwellian distribution function centered on the barycentric velocity:

$$f_{\sigma(m)}^{e\,\lambda} \approx \frac{\rho_\sigma}{m_\sigma\,(2\pi e_\sigma)^{D/2}} \exp\left[-\frac{(\mathbf{v}^\lambda)^2}{2\,e_\sigma}\right] \left[1+\frac{\mathbf{v}^\lambda\cdot\mathbf{u}}{e_\sigma}+\frac{\left(\mathbf{v}^\lambda\cdot\mathbf{u}\right)^2}{2\,e_\sigma^2}-\frac{\mathbf{u}^2}{2\,e_\sigma}\right], \tag{63}$$

where in both cases only the terms up to the second-order in the macroscopic velocities have been considered. Equation (61) can be formulated by introducing some auxiliary variables:

$$\begin{aligned}\frac{\partial\varphi_\sigma^\lambda}{\partial t} \;&+\; \mathbf{v}^\lambda\cdot\nabla\varphi_\sigma^\lambda = -\frac{\chi_H}{\tau_\sigma^0}\left[\varphi_\sigma^\lambda-\varphi_\sigma^{e\,\lambda}\right]-\frac{\epsilon}{\tau_m^0}\left[\varphi_\sigma^\lambda-\varphi_{\sigma(m)}^{e\,\lambda}\right]\\ &+\;(1-\alpha_\sigma)\,\frac{\varphi_\sigma^{e\,\lambda}}{e_\sigma}\left(\mathbf{v}^\lambda-\mathbf{u}_\sigma\right)\cdot\mathbf{g}_\sigma+\alpha_\sigma\frac{\varphi_{\sigma(m)}^{e\,\lambda}}{e_\sigma}\left(\mathbf{v}^\lambda-\mathbf{u}\right)\cdot\mathbf{g}_\sigma,\end{aligned} \tag{64}$$

where $\varphi_\sigma^\lambda = f_\sigma^\lambda/Q_\sigma^\lambda$, $\varphi_\sigma^{e\,\lambda} = f_\sigma^{e\,\lambda}/Q_\sigma^\lambda$, $\varphi_{\sigma(m)}^{e\,\lambda} = f_{\sigma(m)}^{e\,\lambda}/Q_\sigma^\lambda$ and

$$Q_\sigma^\lambda = \frac{1}{m_\sigma\,(2\pi\,e_\sigma)^{D/2}}\exp\left[-\frac{(\mathbf{v}^\lambda)^2}{2\,e_\sigma}\right]. \tag{65}$$

The deviation of the distribution function from the one at rest is also small in the fluid regime limit. It can be assumed that the function φ_σ^λ belongs to the same class of functions which includes the equilibrium functions $\varphi_\sigma^{e\,\lambda}$ and $\varphi_{\sigma(m)}^{e\,\lambda}$, i.e. the class of the D-dimensional second-order polynomial forms. The unknown parameters involved into the interpolation test function can be determined by using the calculated values of the distribution function for the lattice microscopic velocities. Once the interpolation test function is well defined [34], the quadrature formulas for the calculation of the macroscopic moments can be obtained.

In this case they are:

$$\rho_\sigma = \sum_{\lambda=0}^{8} \varsigma^\lambda \, \varphi_\sigma^\lambda \,, \tag{66}$$

$$\rho_\sigma \, \mathbf{u}_\sigma = \sum_{\lambda=0}^{8} \varsigma^\lambda \, \mathbf{v}^\lambda \, \varphi_\sigma^\lambda \,, \tag{67}$$

where ς^λ are the weighting factors

$$\varsigma^\lambda = \begin{cases} 4/9 & \lambda = 0 \\ 1/9 & \lambda = 1, 2, 3, 4 \\ 1/36 & \lambda = 5, 6, 7, 8 \end{cases} . \tag{68}$$

The terms up to the second-order in the macroscopic quantities have been considered in the approximations (62, 63), so the forcing terms in Eq. (64) can be simplified by neglecting higher order terms. It is well known that considering different-order approximations can lead to numerical inaccuracies. Since the acceleration due to the external force field can be considered of the first-order, the terms multiplying the acceleration must be of the first-order with regard to the macroscopic velocities [49]. For this reason, the equations for the discretized distribution functions become:

$$\frac{\partial \varphi_\sigma^\lambda}{\partial t} + \mathbf{v}^\lambda \cdot \nabla \varphi_\sigma^\lambda = -\frac{\chi_H}{\tau_\sigma^0} \left[\varphi_\sigma^\lambda - \varphi_\sigma^{e\,\lambda}\right] - \frac{\epsilon}{\tau_m^0} \left[\varphi_\sigma^\lambda - \varphi_{\sigma(m)}^{e\,\lambda}\right] + \frac{1}{\sqrt{e_\sigma}} \mathbf{k}_{\alpha(\sigma)}^\lambda \cdot \mathbf{g}_\sigma, \tag{69}$$

where

$$\mathbf{k}_{\alpha(\sigma)}^\lambda = \rho_\sigma \left[\frac{\mathbf{v}^\lambda - \mathbf{u}_{\alpha(\sigma)}}{\sqrt{e_\sigma}} + \frac{\mathbf{v}^\lambda \cdot \mathbf{u}_{\alpha(\sigma)}}{\sqrt{e_\sigma^3}} \, \mathbf{v}^\lambda \right] . \tag{70}$$

For recovering Eq. (69), the property that the vector $\mathbf{k}_{\alpha(\sigma)}^\lambda$ is linear with regard to the macroscopic velocities has been used.

The left hand side of Eq. (69) is essentially a substantial derivative and it involves a known microscopic velocity of the lattice, defined by Eq. (58). The ordinary derivatives can be numerically estimated by considering the rate of change for a finite time step δt smaller than the characteristic time scales of the phenomena. The spurious terms, which derive from the previous approximation at the hydrodynamic level, are called discrete lattice effects. In order to cancel the discrete lattice effects, some corrections are need. Let us introduce the following corrected velocities [49, 50]:

$$\rho_\sigma \, \mathbf{u}_\sigma^\star = \sum_{\lambda=0}^{8} \varsigma^\lambda \, \mathbf{v}^\lambda \, \varphi_\sigma^\lambda - \rho_\sigma \left(\mathbf{w}_\sigma^\star / \tau_m - \mathbf{g}_\sigma\right) \delta t/2. \tag{71}$$

The corrected barycentric velocity $\mathbf{u}^\star = \sum_\sigma x_\sigma \mathbf{u}_\sigma^\star$ is consequently defined. Similarly the corrected equilibrium distribution function $\varphi_\sigma^{e\,\lambda\,\star}$ centered on the specific velocity $\mathbf{u}_\sigma^\star$ and the corrected equilibrium distribution function $\varphi_{\sigma(m)}^{e\,\lambda\,\star}$ centered on the barycentric velocity $\mathbf{u}^\star$ can be obtained. Thanks to these quantities, the final lattice Boltzmann method can be formulated:

$$\begin{aligned}\varphi_\sigma^\lambda\left(t+\delta t, \mathbf{x}+\mathbf{v}^\lambda \delta t\right) - \varphi_\sigma^\lambda \;=\; & -\chi_H \frac{\delta t}{\tau_\sigma^0}\left[\varphi_\sigma^\lambda - \varphi_\sigma^{e\,\lambda\,\star}\right] - \epsilon\,\frac{\delta t}{\tau_m^0}\left[\varphi_\sigma^\lambda - \varphi_{\sigma(m)}^{e\,\lambda\,\star}\right] \\ & + \frac{\delta t}{\sqrt{e_\sigma}}\,\mathbf{k}_{\alpha(\sigma)}^{\lambda\,\star} \cdot \left[\, d_\sigma\, \mathbf{g}_\sigma + (1-d_\sigma)\; \mathbf{w}_\sigma^\star/\tau_m \,\right],\end{aligned} \tag{72}$$

where d_σ is defined as

$$d_\sigma = 1 - \frac{1}{2}\,\frac{\delta t}{\alpha_\sigma\,\tau_m} = 1 - \frac{\delta t}{2}\left(\frac{\chi_H}{\tau_\sigma^0} + \frac{\epsilon}{\tau_m^0}\right), \tag{73}$$

and it takes into account the discrete lattice effects, while $\mathbf{k}_{\alpha(\sigma)}^{\lambda\,\star}$ is the quantity defined by Eq. (70) when the corrected velocities are considered. It is easy to check that, in the continuous limit $\delta t \rightarrow 0$, the corrected equations coincide with the previous Eqs. (69). The corrected specific velocity involves the corrected diffusion velocity and for this reason Eq. (71) realizes an implicit formulation. This feature can be made evident by considering the definition of diffusion velocity:

$$\rho_\sigma \sum_k \left[(1+\omega_m/2)\,\delta_{k\,\sigma} - x_{\,k}\,\omega_m/2\,\right]\mathbf{u}_k^\star = \sum_{\lambda=0}^{8} \varsigma^\lambda\,\mathbf{v}^\lambda\,\varphi_\sigma^\lambda + \rho_\sigma\,\mathbf{g}_\sigma\,\delta t/2, \tag{74}$$

where $\omega_m = \delta t/\tau_m$ is the dimensionless frequency for the cross collisions. It is possible to derive an explicit formulation for the corrected velocities from the previous equation:

$$\rho_\sigma\,\mathbf{u}_\sigma^\star = \sum_k \left[\left(\frac{2}{2+\omega_m}\,\delta_{k\,\sigma} + \frac{\omega_m}{2+\omega_m}\,x_{\,k}\right)\left(\sum_{\lambda=0}^{8} \varsigma^\lambda\,\mathbf{v}^\lambda\,\varphi_k^\lambda + \rho_\sigma\,\mathbf{g}_{\,k}\,\delta t/2\right)\right]. \tag{75}$$

When the cross collisions are negligible $\omega_m = 0$, the previous correction reduces to the usual definition for the corrected velocity, which has been modified in order to take into account the effects of the external field [24]. The final lattice Boltzmann method exactly recovers the following equations (see Appendix B):

$$\frac{\partial \rho_\sigma}{\partial t} + \nabla\cdot(\rho_\sigma \mathbf{u}_\sigma^\star) = 0, \tag{76}$$

$$
\begin{aligned}
\frac{\partial\left(\rho_\sigma \mathbf{u}_\sigma\right)}{\partial t} &+ \nabla\cdot\left[\left(1-\alpha_\sigma\right)\rho_\sigma \mathbf{u}_\sigma^\star \otimes \mathbf{u}_\sigma^\star + \alpha_\sigma\,\rho_\sigma \mathbf{u}^\star \otimes \mathbf{u}^\star\right.\\
&+ \left.\alpha_\sigma \rho_\sigma \mathbf{u}_{\alpha(\sigma)}^\star \otimes \mathbf{w}_\sigma^\star + \alpha_\sigma \rho_\sigma \mathbf{w}_\sigma^\star \otimes \mathbf{u}_{\alpha(\sigma)}^\star\right] =\\
&- \nabla\left(\rho_\sigma e_\sigma\right) + \rho_\sigma \mathbf{g}_\sigma - \frac{1}{\tau_m}\,\rho_\sigma \mathbf{w}_\sigma^\star\\
&+ \nabla\cdot\left\{d_\sigma\,\alpha_\sigma \rho_\sigma e_\sigma \tau_m\left[\nabla \mathbf{u}_{\alpha(\sigma)}^\star + \nabla \mathbf{u}_{\alpha(\sigma)}^{\star T}\right]\right\}.
\end{aligned}
\tag{77}
$$

The proposed lattice Boltzmann method involves additional lattice parameters and a proper strategy is needed in order to tune them. First of all, the constraints must be defined. The lattice grid size δx, the viscosity of the components ν_σ and the viscosity of the ideally coupled mixture ν_m are considered input data of the problem. The internal energies for the components e_σ can be freely tuned, since the energy equations are not solved. In particular, assuming $r_\sigma = e_\sigma/c^2$, the parameters r_σ can be set in such a way as to reproduce the exact pressure gradients in the momentum equations. On the other hand, the local stability analysis of the lattice Boltzmann model suggests that $r_\sigma = 1/3$ is the optimal value for improving the stability [51]. In the low Mach number limit and when ideal gases are considered, both the effects of density gradients and of the pressure gradients are negligible. For this reason, stability will be preferred and the parameters will be accordingly selected.

Let us consider first the ideally non-interacting configuration, i.e when $1/\tau_m \to 0$. Let us define $\omega_\sigma^0 = \delta t^0/\tau_\sigma^0$ the dimensionless frequency for the generic component and c_σ^0 the lattice velocity for the generic component. Since the dimensionless frequency must be set in such a way as to respect the stability criterion $0 \leq \omega_\sigma^0 \leq 2$, the problem is to define c_σ^0 and τ_σ^0 in order to recover the desired lattice grid size δx and the kinematic viscosity for the single component ν_σ. The following formulas hold:

$$
\tau_\sigma^0 = \frac{\left(2-\omega_\sigma^0\right)}{6\left(\omega_\sigma{}^0\right)^2}\,\frac{\delta x^2}{\nu_\sigma},
\tag{78}
$$

$$
c_\sigma^0 = \frac{\delta x}{\tau_\sigma^0\,\omega_\sigma^0} = \frac{6\,\omega_\sigma^0}{\left(2-\omega_\sigma^0\right)}\,\frac{\nu_\sigma}{\delta x}.
\tag{79}
$$

The lattice velocities must be all identical, i.e. $c_\sigma^0 = c^0$, because all the mixture components are computed on the same lattice. This introduces a new constraint for the dimensionless frequencies. Let us label with a the component of the mixture characterized by the smallest viscosity, such that $\nu_\sigma^0 \geq \nu_a^0$. The condition $c_\sigma^0 = c^0$ implies:

$$
\omega_\sigma^0 = \frac{2\,\nu_a\,\omega_a^0}{\nu_\sigma\left(2-\omega_a^0\right) + \nu_a\,\omega_a^0} \leq \omega_a^0.
\tag{80}
$$

Selecting ω_a^0 in such a way that $0 \leq \omega_a^0 \leq 2$, then all the other dimensionless frequencies will follow from the previous condition and they will be also $0 \leq \omega_\sigma^0 \leq 2$. In particular the previous condition implies that the discretization time steps for all the components will be identical $\delta t^0 = \tau_\sigma^0\, \omega_\sigma^0 = \tau_a^0\, \omega_a^0$.

Since both binary mixture species evolve on the same lattice, if they have significantly different molecular weights, this can increase the computational time because the time step size is dictated by the lighter species, making the simulation very slow. Moreover, significantly different molecular weights widen the spectrum of the dimensionless relaxation frequencies, i.e. $\omega_\sigma^0 \ll \omega_a^0$ or, equivalently, $\omega_\sigma^0 \to 0$. Recalling Eqs. (78, 79), it is evident that very small dimensionless frequencies for the proposed tuning strategy can induce inaccurate numerical solutions. For this reason, this strategy is successful only for moderately different molecular weights. However, this is an open issue for all multi-species lattice Boltzmann models which use a single lattice.

We can proceed in a similar way for the ideally miscible configuration. Let us define $\omega_m^0 = \delta t^0/\tau_m^0$ as the dimensionless frequency for the ideally miscible configuration. The following formulas hold:

$$\tau_m^0 = \frac{(2-\omega_m^0)}{6\,(\omega_m^0)^2}\,\frac{\delta x^2}{\nu_m}, \tag{81}$$

$$c_\sigma^m = \frac{\delta x}{\tau_m^0\,\omega_m^0} = \frac{6\,\omega_m^0}{(2-\omega_m^0)}\,\frac{\nu_m}{\delta x}. \tag{82}$$

In this case, the lattice velocities are naturally identical $c_\sigma^m = c^m$ and the same happens for the discretization time steps $\delta t^m = \tau_m^0\,\omega_m^0$. For an intermediate degree of coupling, the generalized expression of the discretization time step can be assumed as:

$$\delta t = \chi_H\,\delta t^0 + \epsilon\,\delta t^m = \chi_H\,\tau_a^0\omega_a^0 + \epsilon\,\tau_m^0\,\omega_m^0. \tag{83}$$

This allows us to calculate the intermediate values of the dimensionless frequencies:

$$\omega_\sigma = \delta t\,\frac{\chi_H}{\tau_\sigma^0} = \omega_\sigma^0\,\chi_H\,\left[\chi_H + \epsilon\,\theta\right], \tag{84}$$

$$\omega_m = \delta t\,\frac{\epsilon}{\tau_m^0} = \omega_m^0\,\epsilon\,\left[\chi_H/\theta + \epsilon\right], \tag{85}$$

where θ is defined as

$$\theta = \frac{\nu_\sigma}{\nu_m}\,\frac{(2-\omega_m^0)}{(2-\omega_\sigma^0)}\,\frac{\omega_\sigma^0}{\omega_m^0}. \tag{86}$$

The discussed strategy for setting the microscopic parameters allows us to reproduce the correct viscosities for the components in the ideally non-interacting limit and for the mixture in the ideally miscible limit.

The coupling strength ϵ can be tuned by considering the mutual diffusion coefficient. Since the tuning strategy based on mixture viscosity was considered in this paper, some constraints for selecting the mutual diffusivity must exist. First, since the internal energies for the discrete model are the same, i.e. $e_a = e_b$, in order to reduce the computational efforts by using a single lattice, the Eqs. (18, 19) for the diffusion velocity become the same. Consequently, it is not possible to distinguish between diffusion coefficient and modified diffusion coefficient, i.e. $D = D^*$, or between Schmidt number and modified Schmidt number, i.e. $Sc = Sc^*$. In this case, the diffusion coefficient simply reads

$$D_c(\epsilon) = \frac{\nu_m}{\epsilon}, \tag{87}$$

and, consequently, the Schmidt number becomes

$$Sc(\epsilon) = \frac{\nu_c(\epsilon)}{D_c(\epsilon)} = \epsilon\,(1-\epsilon)\,\frac{\sum_\sigma x_\sigma\,\nu_\sigma}{\nu_m} + \epsilon^2. \tag{88}$$

In this case, the mutual diffusion coefficient is bounded from below, $D \geq \nu_m$. The situation is the opposite of that discussed for the linearized models. The viscous coupling allows us to freely tune the critical mixture viscosity $\nu_c(\epsilon)$ and, in particular, to recover the ideally miscible configuration. But the mutual diffusion coefficient cannot be smaller than the effective viscosity for ideally coupled mixture, ν_m. Also a constraint holds for the Schmidt number.

On the other hand, as previously done for the continuous model, Hamel's function χ_H can be set in order to obtain the desired critical viscosity. In this case the previous correlations must be taken into account for the computation of the critical viscosity given by Eq. (31) in the discrete model:

$$\nu_c(\epsilon) = \frac{d_a\,c_a^2\,x_a\,\nu_a}{\chi_H\,d_a^0\,(c_a^0)^2 + \epsilon\,d_a^m\,(c_a^m)^2\,\nu_a/\nu_m} + \frac{d_b\,c_b^2\,x_b\,\nu_b}{\chi_H\,d_b^0\,(c_b^0)^2 + \epsilon\,d_b^m\,(c_b^m)^2\,\nu_b/\nu_m}, \tag{89}$$

where the discrete lattice parameter depends on the coupling strength, namely

$$d_\sigma = 1 - \left(1 - d_\sigma^0\right)\,\chi_H^2 - \left(1 - d_\sigma^m\right)\,\epsilon^2 - \frac{\chi_H\,\epsilon}{2}\,\left(\omega_\sigma^0\,\theta + \omega_m^0/\theta\right), \tag{90}$$

and $d_\sigma^0 = 1 - \omega_\sigma^0/2$ and $d_\sigma^m = 1 - \omega_m^0/2$ are the limiting cases for ideally non-interacting components and for ideally miscible components respectively.

Finally, the compatibility condition for the discrete model must be also modified. Considering $\epsilon = 1$ in the system of discrete equations, analogous to the previous system of equations (22), and applying the difference between the equations of the system, the compatibility condition is obtained:

$$\frac{e}{\nu_m} \left(u_a^x - u_b^x \right) = g_a^x - g_b^x. \tag{91}$$

If the forcing terms due to the external field are such as $g_a^x = g_b^x$, then the solution of the system of equations is unique, i.e. $u_a^x = u_b^x$. Let us model a mixture affected by a given forcing term $\rho\, g^x$. For the mixture, where only the total value of the pressure gradient is known, the splitting of the forcing term among the momentum equations for the components can be made by means of the previous compatibility condition. If the source term for a generic species is called $\rho_\sigma\, g_\sigma^x$, then the compatibility condition for the discrete model prescribes that $\rho_\sigma\, g_\sigma^x = x_\sigma\, \rho\, g^x$, i.e. the splitting of the forcing term must be made on the basis of the mass concentrations. The difference with the continuous model is due to the fact that for the lattice Boltzmann model $e_\sigma = e = c^2/3$, while for the continuous model $e_\sigma = e\, y_\sigma / x_\sigma$. This feature of the discrete lattice model is a consequence of the stability constraint which has been assumed but it can be easily overcome by considering different values for the internal energy of each species, i.e. $r_\sigma \neq 1/3$. In the next section, the designed lattice model will be numerically solved.

IV. DISCUSSION OF NUMERICAL RESULTS AND CONCLUSIONS

Some numerical results are reported for the suggested discrete lattice model in this section. First, some carefully conducted benchmarking computations were performed to verify the transport coefficients of the proposed lattice Boltzmann model. Because the proposed model is isothermal, diffusivity and kinematic viscosity are the only involved transport phenomena. Theoretical predicted equations (87, 89) were compared with simulation results reported in Table I. In particular, simulated diffusivity values were obtained using the transient method presented in [16]. In the case where u^x is a constant and $D_c(\epsilon)$ is also a constant, a solution of Eq. (38), describing a decaying sine wave flowing along in the x direction with velocity u^x, is given as

$$x_a[x, t] = x_a^0 + (x_a' - x_a^0)\, \exp\left[-k^2\, D_c(\epsilon)\, t\right]\, \sin\left[k\, (x - u^x\, t)\right], \tag{92}$$

where x_a^0 is the constant averaged concentration of the a species, x_a' is the maximum value of the initial perturbation applied to concentration (in the reported simulations $x_a' > x_a^0$ was adopted) and $1/k$ is the wave length of the perturbation. Since periodic boundary conditions were used, the ratio between the computational domain length along x axis and the wave length was an integer. Assuming $u^x = 0$, the numerical diffusivity can be measured by considering the sine wave maximum decay, namely

$$(D_c)_M = \frac{1}{k^2\, t}\, \ln\left\{\frac{x_a[\pi/(2\,k), 0] - x_a^0}{x_a[\pi/(2\,k), t] - x_a^0}\right\}. \tag{93}$$

Similarly we can proceed for the mixture kinematic viscosity. Simulated mixture kinematic viscosity values were obtained considering a plane Poiseuille flow. If the accelerations due to the external force field are the same $g_a^x = g_b^x = g^x$ and prevail over the effects due to the concentration gradients, then it is possible to assume $a_\sigma^x \approx \rho_\sigma\, g^x$. Assuming g^x is a constant, a solution of Eq. (41), describing a parabolic flow between infinitely extended parallel planes, is given as

$$u_\nu^x[y] = \frac{g^x}{2\,\nu_c(\epsilon)}\,(H - y)\,y, \tag{94}$$

where H is the distance between the parallel planes. The numerical kinematic viscosity can be measured by considering the fluid flow maximum velocity, namely

$$(\nu_c)_M = \frac{H^2\, g^x}{8\, u_\nu^x[H/2]}. \tag{95}$$

Recalling the results reported in Table I, a good agreement between theoretical predictions and simulation results was found for both diffusivity and mixture kinematic viscosity. From this table, it is evident that, in the proposed model, the cross-collision relaxation time can be tuned to recover the desired mutual diffusivity or the desired mixture kinematic viscosity, but not both independently.

The numerical results reported in Table I refer to one dimensional case because analytical solutions useful for benchmarking are available. However some doubts emerge regarding the fact that the suitability of suggested tuning strategy for the relaxation time constants could be valid only for the one dimensional case. These doubts were dispelled by considering the two dimensional randomly generated porous medium reported in Fig. 1. The actual calculations were done by using periodic boundary conditions. A given pressure gradient induced the flow of some binary mixtures through the porous medium. Each mixture was characterized by a different coupling strength.

When the coupling strength is very small, the two species independently evolve according to their kinematic viscosities. When the coupling strength increases, i.e. when the cross collisions become important, the slower species try to slacken the other species and vice versa. At the end, the result is that the two species are characterized by velocities much more similar in comparison with the results for the non-interacting configurations. In the ideally coupled case, i.e. when the cross collisions prevail the mixture viscous velocity or equivalently the barycentric velocity, is enough to characterize the mixture fluid flow.

From the physical point of view, we can imagine that each mixture is made of components characterized by different diffusivity but the same kinematic viscosity (if considered alone). From the kinetic point of view, this test case allows us to evaluate the effects of increasing cross collisions for species characterized by the same self collisions. Moreover, the effectiveness of the tuning strategy for the relaxation time constants can be easily verified because the mixture viscous velocity u_ν^x should be linear with respect to the coupling strength. In fact, if in the low Reynolds number limit the fluid flow for each species is essentially controlled by the critical viscosity ν_c and the latter was tuned according to Eq. (42), then the mixture viscous velocity should be consequently linear.

As an example, the mixture viscosity was assumed to be linear with the coupling strength, but any other experimental formula can be included. In particular, strong experimental evidences exist that cross collisions effect the effective mixture viscosity. In fact, a very popular experimental formula for the mixture kinematic viscosity is [48]:

$$\nu_c^r = \frac{x_a\, \nu_a}{1 + F_{a\,b}\, y_b / y_a} + \frac{x_b\, \nu_b}{1 + F_{b\,a}\, y_a / y_b}, \tag{96}$$

where $F_{a\,b}$ and $F_{b\,a}$ are positive corrective factors. In particular the experimental data show that the effective kinematic viscosity for the mixture is smaller than the averaged viscosity based on the mass concentrations of the components $\nu_m^r \leq \sum_\sigma x_\sigma \nu_\sigma$. This allows us to understand the importance of the previously discussed drawback of the linearized models, which forces us to consider a fixed mixture viscosity equal to the averaged viscosity based on the mass concentrations [25, 26]. Fortunately, the proposed tuning strategy overcomes this drawback.

In Tabs. II and III and in Fig. 2 some numerical results are reported which have been calculated by the proposed discrete lattice model. In the first case A, the fully-coupled mixture viscosity is set equal to the mass averaged kinematic viscosity $\nu_m = \sum_\sigma x_\sigma \nu_\sigma$ while, in the second case B, it is set equal to a lower value $\nu_m \leq \sum_\sigma x_\sigma \nu_\sigma$. Hamel's function has been set in such a way that the linear equation (42) holds also for the discrete critical viscosity given by Eq. (89). As it is evident by considering Fig. 2, the viscous velocity effectively shows a linear dependence on the coupling strength. Although the mathematical suitability of the previous strategy has been previously deduced for the one dimensional case only, the two dimensional numerical results confirm that it is generally effective and it allows us to recover the desired behavior of the viscous velocity with respect to the coupling strength for more complex computational domains.

Taking into account the previous results, some conclusions can be summarized.

1. In the present paper, a new lattice Boltzmann model for binary mixtures has been proposed on the basis of the two-fluid kinetic theory developed by Hamel [42, 43]. A proper correction of the discrete lattice effects allows us to exactly recover the performance of the continuous model with second order accuracy in both time and space [49, 50]. This substantially corrects and improves a previously proposed two-fluid lattice Boltzmann model for binary mixtures, based on a linearized cross collisional operator [25, 26]. In this way, the theoretical foundation of this model is well established in the framework of the kinetic theory. The lattice model thus naturally inherits all the properties and all diffusion driving mechanisms of the original kinetic model. This is in contrast to previous lattice Boltzmann models for mixtures [15, 17–22], which are not directly based on the fundamental physics of continuous kinetic equations. These models rely on pseudo-potential interactions [17, 18] or heuristic free energies [15, 19–22] to produce the requisite mixing.

2. This kinetic model implies two fundamental ways to couple the velocity fields of the mixture components: a *force coupling*, based on a source term in the momentum equations proportional to the diffusive velocity, and a *viscous coupling*, based on an additional term in the effective stress tensor proportional to the deformation of the barycentric velocity field. Both coupling mechanisms derive from cross collisions. Unfortunately, the macroscopic mutual diffusivity and the mixture kinematic viscosity cannot be independently tuned because only a single cross-collision relaxation time is available. Tuning strategies based on diffusivity and on mixture kinematic viscosity have been discussed. It is worth pointing out that in this model the viscous relaxation

process and the diffusion process are inseparable. This is not because of a mysterious "equilibrium velocity" as it happens for the previous single-fluid models [17, 18], but only because of cross collisions, as it should be. The viscous relaxation process and the diffusion process can be decoupled by linearizing the cross collisional operator [25, 26], in order to freely vary the Schmidt number. This is practical but unphysical because experimental evidences suggest that cross collisions effect the mixture kinematic viscosity, which in fact cannot be constantly assumed equal to the mass averaged kinematic viscosity [48].

3. Hamel's model automatically includes both single-fluid and two-fluid approaches, because both self collisions and cross collisions are described by BGK-like structures. It can be considered the ideal forerunner of all linearized models, like the Sirovich's model, but it does not require any hypothesis on cross collisions because there is no local approximation of the collisional operators. For this reason, the whole range of the coupling strength is naturally included. In order to reproduce the ideally miscible configurations, the *viscous coupling* is more effective than the *force coupling* because there is no need to consider infinitely strong forcing terms in the momentum equations. Recently a two-fluid model with similar characteristics has been suggested [28], but the lattice collisional operator has been conjectured without any theoretical foundation and the Chapman-Enskog asymptotic analysis has been reported only for a linearized approximation. In particular there is no discussion concerning how cross collisions effect the mixture kinematic viscosity.

4. This paper shows that it is possible to develop a two-fluid model within the framework of the standard lattice Boltzmann method and there is no need to consider the finite-difference lattice approach in order to achieve this goal. This evidence contradicts some statements of a recent paper [27]. Although the first proposed two-fluid model did not satisfy the species continuity equation [25, 26], it is easy to solve the problem by considering the technique recently suggested to properly take into account the effects due to the external force field [49, 50]. In this case, the velocity correction must satisfy a slightly more complicated formula, given by Eq. (75).

5. Although more discrete distribution functions are considered, they should share the same discretization of the space space. In fact if the same spatial discretization and

the same time step are considered, then all the distribution functions can share the same lattice velocity $c_\sigma = c$. This congruence condition is simply a numerical trick, but it is very useful in order to reduce the demand of computational resources by including different relaxation dynamics on the same discretization of the phase space. However, since this strategy forces us to consider values of the relaxation parameters different from the optimal ones required by stability, the universality of this strategy could be uncertain.

An improvement of the proposed lattice Boltzmann model for binary mixing could be the development of a multiple-relaxation-time model for the cross-collisional operator [52, 53]. In this way, it should be possible to independently tune both mutual diffusivity and mixture kinematic viscosity, which differs from the elementary mass averaged kinematic viscosity according to the experimental data, but simultaneously preserving the fact that both phenomena derive from cross collisions.

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APPENDIX A: CHAPMAN-ENSKOG ASYMPTOTIC ANALYSIS OF HAMEL'S MODEL

An asymptotic analysis will be performed to recover the macroscopic equations for the lower-order moments, which derive from the simplified kinetic models defined by the previous Eqs. (3, 4, 5, 8). The Chapman-Enskog expansion technique [31] is very popular and it essentially consists of expanding the velocity distribution function in terms of a small parameter (Knudsen number) but not the macroscopic moments. It is well known that the Chapman-Enskog expansion can bring in solutions which are simply nonexistent [2, 3], when the equations beyond the Navier-Stokes system are considered. Since in this case we limit our interest to the transport coefficients involved in the Navier-Stokes system, the Chapman-Enskog expansion will be considered.

Let us expand the velocity distribution function in terms of a small parameter K, which is proportional to the Knudsen number Kn:

$$f_\sigma = f_\sigma^{(0)} + K\, f_\sigma^{(1)} + K^2\, f_\sigma^{(2)} + ... \,, \tag{A1}$$

and to proceed in the same way for the partial derivatives:

$$\frac{\partial}{\partial t} = K\, \frac{\partial}{\partial t^{(1)}} + K^2\, \frac{\partial}{\partial t^{(2)}} + ..., \tag{A2}$$

$$\frac{\partial}{\partial x_i} = K\, \frac{\partial}{\partial x_i^{(1)}} + ..., \tag{A3}$$

$$\frac{\partial}{\partial v_i} = K\, \frac{\partial}{\partial v_i^{(1)}} + ... \tag{A4}$$

The expansion of the gradient which involves the microscopic velocity (A4) is quite unusual, but it is equivalent to the common practice of considering the effects of the external force field of the first order in the Knudsen number. Both of these approaches simplify the asymptotic analysis but nonetheless allow us to recover the correct source term in the momentum equation due to the external force field. Substituting the previous expansions in the kinetic model, given by Eqs. (3, 4, 5), a coupled hierarchy system of equations in the powers of K is obtained and the first elements of this system are:

$$f_\sigma^{(0)} = (1-\alpha_\sigma)\, f_\sigma^e + \alpha_\sigma f_{\sigma(m)}^e, \tag{A5}$$

$$\frac{\partial f_\sigma^{(0)}}{\partial t^{(1)}} + \mathbf{v}\cdot\nabla^{(1)} f_\sigma^{(0)} + \mathbf{g}_\sigma\cdot\nabla_{\mathbf{v}}^{(1)} f_\sigma^{(0)} = -\frac{1}{\alpha_\sigma\, \tau_m} f_\sigma^{(1)}, \tag{A6}$$

$$\frac{\partial f_\sigma^{(0)}}{\partial t^{(2)}} + \frac{\partial f_\sigma^{(1)}}{\partial t^{(1)}} + \mathbf{v}\cdot\nabla^{(1)} f_\sigma^{(1)} + \mathbf{g}_\sigma \cdot \nabla_{\mathbf{v}}^{(1)} f_\sigma^{(1)} = -\frac{1}{\alpha_\sigma\, \tau_m} f_\sigma^{(2)}, \tag{A7}$$

where $0 \leq \alpha_\sigma = \tau_\sigma / (\tau_\sigma + \tau_m) \leq 1$. The effects of the external force field in Eq. (A7), which involves the terms $O(K^2)$, can be neglected. This practice is based on the fact that the non-equilibrium distribution function does not differ too much from the equilibrium distribution with regard to the microscopic velocity, in the fluid regime limit [33]. Equation (A6) can be simplified by means of the first order of the expansion (A5):

$$-\nabla_{\mathbf{v}}^{(1)} f_\sigma^{(0)} = (1-\alpha_\sigma)\,\frac{f_\sigma^e}{e_\sigma}\,(\mathbf{v}-\mathbf{u}_\sigma) + \alpha_\sigma \frac{f_{\sigma(m)}^e}{e_\sigma}\,(\mathbf{v}-\mathbf{u}), \tag{A8}$$

where the scale index of the velocity has been omitted $\mathbf{v}^{(1)} \rightarrow \mathbf{v}$ because it is the dummy variable in the next integrals. Finally the system of equations becomes:

$$\begin{aligned} \frac{\partial f_\sigma^{(0)}}{\partial t^{(1)}} \;&+\; \mathbf{v}\cdot\nabla^{(1)} f_\sigma^{(0)} = -\frac{1}{\alpha_\sigma\, \tau_m} f_\sigma^{(1)} \\ &+\; (1-\alpha_\sigma)\,\frac{f_\sigma^e}{e_\sigma}\,\mathbf{g}_\sigma\cdot(\mathbf{v}-\mathbf{u}_\sigma) + \alpha_\sigma \frac{f_{\sigma(m)}^e}{e_\sigma}\,\mathbf{g}_\sigma\cdot(\mathbf{v}-\mathbf{u}), \end{aligned} \tag{A9}$$

$$\frac{\partial f_\sigma^{(0)}}{\partial t^{(2)}} + \frac{\partial f_\sigma^{(1)}}{\partial t^{(1)}} + \mathbf{v}\cdot\nabla^{(1)} f_\sigma^{(1)} = -\frac{1}{\alpha_\sigma\, \tau_m} f_\sigma^{(2)}. \tag{A10}$$

Recovering the macroscopic equations for the moments of the velocity distribution function requires multiplying the previous equations by the collisional invariants and then performing the integration over the microscopic velocity. Since the previous equations are coupled, this procedure will be useful only if the integral equations are decoupled. In the single-fluid BGK model, it is easy to demonstrate that the higher-order terms due to the expansion of the distribution function, i.e. $f^{(\xi)}$ for $\forall\, \xi \geq 1$, do not effect the moments of the collisional invariants. In this case, since the first order of the expansion $f_\sigma^{(0)}$ is a linear combination of Maxwellian functions, which in general does not yield a Maxwellian function, this property does not hold anymore. The following similar conditions can be derived:

$$\int m_\sigma \sum_{\xi=1}^{\infty} K^\xi\, f_\sigma^{(\xi)}\, d\mathbf{v} = \int m_\sigma \left[f_\sigma - f_\sigma^{(0)} \right]\, d\mathbf{v} = 0, \tag{A11}$$

$$\begin{aligned} \sum_\sigma \frac{1}{\alpha_\sigma\, \tau_m} \int m_\sigma \mathbf{v} \sum_{\xi=1}^{\infty} K^\xi\, f_\sigma^{(\xi)}\, d\mathbf{v} \;&=\; \sum_\sigma \frac{1}{\alpha_\sigma\, \tau_m} \int m_\sigma \mathbf{v} \left[f_\sigma - f_\sigma^{(0)} \right]\, d\mathbf{v} \\ &=\; \frac{1}{\tau_m} \sum_\sigma \rho_\sigma\, (\mathbf{u}_\sigma - \mathbf{u}) = 0, \end{aligned} \tag{A12}$$

$$\sum_\sigma \frac{1}{\alpha_\sigma\,\tau_m}\int \frac{1}{2}\,m_\sigma\,\mathbf{v}^2\sum_{\xi=1}^{\infty} K^\xi\, f_\sigma^{(\xi)}\,d\mathbf{v} \;=\; \sum_\sigma \frac{1}{\alpha_\sigma\,\tau_m}\int \frac{1}{2}\,m_\sigma\,\mathbf{v}^2\left[f_\sigma - f_\sigma^{(0)}\right]\,d\mathbf{v}$$

$$= \frac{1}{2\,\tau_m}\sum_\sigma \rho_\sigma\left(\mathbf{u}_\sigma^2 - \mathbf{u}^2\right) \geq 0. \tag{A13}$$

Particularly, the condition (A13) can be easily proved by remembering that the sum of the kinetic energies of the components must be greater than or equal to the barycentric kinetic energy because of the deformation energy. This consideration allows us to assume that each term of the series is positive and can be upper bounded by the right hand side of the property (A13). Since the previous relations must be satisfied for any small value of the parameter K, finally we obtain:

$$\int m_\sigma f_\sigma^{(\xi)}\,d\mathbf{v} = 0, \tag{A14}$$

$$\sum_\sigma \mathbf{q}_\sigma^{(\xi)} = 0, \tag{A15}$$

$$\sum_\sigma \varphi_\sigma^{(\xi)} \ll \frac{1}{2\,\tau_m}\sum_\sigma \rho_\sigma\left(\mathbf{u}_\sigma^2 - \mathbf{u}^2\right), \tag{A16}$$

for any ξ-th perturbation ($\xi \geq 1$) of the velocity distribution function, where $\mathbf{q}_\sigma^{(\xi)}$ and $\varphi_\sigma^{(\xi)}$ are moments of the considered perturbation:

$$\mathbf{q}_\sigma^{(\xi)} = \frac{1}{\alpha_\sigma\,\tau_m}\int m_\sigma \mathbf{v} f_\sigma^{(\xi)}\,d\mathbf{v}, \tag{A17}$$

$$\varphi_\sigma^{(\xi)} = \frac{1}{2\,\alpha_\sigma\,\tau_m}\int m_\sigma \mathbf{v}^2 f_\sigma^{(\xi)}\,d\mathbf{v}. \tag{A18}$$

This means that the higher-order terms of the expansion for the distribution function can effect the moments of the collisional invariants for each species in such a way that the previous relations must hold.

Multiplying Eq. (A9) by the collisional invariants and integrating over the microscopic velocity, the following equations are recovered:

$$\frac{\partial \rho_\sigma}{\partial t^{(1)}} + \nabla^{(1)}\cdot\left(\rho_\sigma \mathbf{u}_\sigma\right) = \alpha_\sigma\,\nabla^{(1)}\cdot\left(\rho_\sigma \mathbf{w}_\sigma\right), \tag{A19}$$

$$\frac{\partial}{\partial t^{(1)}}\left[\rho_\sigma\left(\mathbf{u}_\sigma - \alpha_\sigma \mathbf{w}_\sigma\right)\right] \;+\; \nabla^{(1)}\cdot\left[\left(1-\alpha_\sigma\right)\rho_\sigma \mathbf{u}_\sigma\otimes\mathbf{u}_\sigma + \alpha_\sigma\,\rho_\sigma \mathbf{u}\otimes\mathbf{u}\right] =$$

$$-\;\nabla^{(1)}\left(\rho_\sigma e_\sigma\right) + \rho_\sigma \mathbf{g}_\sigma - \mathbf{q}_\sigma^{(1)}, \tag{A20}$$

$$
\begin{aligned}
\frac{\partial\left(\rho_\sigma e_\sigma^t\right)}{\partial t^{(1)}} \; &- \; \frac{\alpha_\sigma}{2}\frac{\partial}{\partial t^{(1)}}\left[\rho_\sigma\left(\mathbf{u}_\sigma^2-\mathbf{u}^2\right)\right]+\nabla^{(1)}\cdot\left[\rho_\sigma\left(e_\sigma^t\mathbf{u}_\sigma-\alpha_\sigma e_\sigma\mathbf{w}_\sigma\right)\right] = \\
&- \; \nabla^{(1)}\left[\rho_\sigma e_\sigma\left(\mathbf{u}_\sigma-\alpha_\sigma\mathbf{w}_\sigma\right)\right]+\frac{\alpha_\sigma}{2}\nabla^{(1)}\left[\rho_\sigma\left(\mathbf{u}_\sigma^2\mathbf{u}_\sigma-\mathbf{u}^2\mathbf{u}\right)\right] \\
&+ \; \rho_\sigma\left(\mathbf{u}_\sigma-\alpha_\sigma\mathbf{w}_\sigma\right)\cdot\mathbf{g}_\sigma-\varphi_\sigma^{(1)}, \qquad \text{(A21)}
\end{aligned}
$$

where $\mathbf{w}_\sigma=\mathbf{u}_\sigma-\mathbf{u}$ is the diffusion velocity for the generic species and $e_\sigma^t=e_\sigma+\rho_\sigma\mathbf{u}_\sigma^2/2$ is the specific total energy.

Let us consider the effects of the first-order perturbation on the continuity equation. Multiplying Eq. (A10) by the particle mass for the generic species, the following equation is recovered:

$$
\frac{\partial\rho_\sigma}{\partial t^{(2)}}=-\alpha_\sigma\,\tau_m\,\nabla^{(1)}\cdot\mathbf{q}_\sigma^{(1)}. \qquad \text{(A22)}
$$

In the derivation of the previous equation, Eqs. (A19, A20) have been applied. If Eq. (A19) and Eq. (A22) are summed, then the final result for the continuity equation is obtained:

$$
\frac{\partial\rho_\sigma}{\partial t}+\nabla\cdot\left(\rho_\sigma\mathbf{u}_\sigma\right)=\alpha_\sigma\,\nabla\cdot\left[\rho_\sigma\mathbf{w}_\sigma-\tau_m\,\mathbf{q}_\sigma^{(1)}\right]. \qquad \text{(A23)}
$$

According to the previously discussed definition given by Eq. (A17), the general property given by Eq. (A12) prescribes that the resultant of the vectors $\mathbf{q}_\sigma^{(1)}$, when all the mixture components are considered, must be zero, i.e. $\sum_\sigma\mathbf{q}_\sigma^{(1)}=0$. Since each parameter α_σ can be independently varied by setting the relaxation time constants for the corresponding species, the previous property implies that the vector $\mathbf{q}_\sigma^{(1)}$ cannot depend on α_σ. For this reason, the divergence involved in the right hand side of the previous equation does not explicitly depend on α_σ. In particular, if the relaxation time constants are properly set in such a way that $\alpha_\sigma=1$, then the considered model reduces to the single-fluid model with regard to the barycentric velocity. It has been shown by means of both Grad's moment method and the Chapman-Enskog expansion that this model satisfies the continuity equation [23, 47]. For this reason, it is possible to conclude that the $\mathbf{q}_\sigma^{(1)}=\rho_\sigma\mathbf{w}_\sigma/\tau_m$ and this result must be consider independent of the value of α_σ. For this reason, Eq. (A23) reduces to Eq. (9).

Let us proceed in the same way for the momentum equation. Multiplying the previously discussed Eq. (A10) by the particle momentum for the generic species and integrating over the microscopic velocity, the following equation is recovered:

$$
\begin{aligned}
\frac{\partial}{\partial t^{(2)}}\left[\rho_\sigma\left(\mathbf{u}_\sigma-\alpha_\sigma\mathbf{w}_\sigma\right)\right] \; &+ \; \alpha_\sigma\frac{\partial}{\partial t^{(1)}}\left(\rho_\sigma\mathbf{w}_\sigma\right) = \\
&- \; \nabla^{(1)}\cdot\left(\int m_\sigma\mathbf{v}\otimes\mathbf{v}\,f_\sigma^{(1)}\,d\mathbf{v}\right)-\mathbf{q}_\sigma^{(2)}. \qquad \text{(A24)}
\end{aligned}
$$

The previous Eqs. (A19, A20, A21) will be used to simplify the analysis of the effects of the first-order perturbation to the momentum equation and in particular to calculate the integrals in the right hand side of the previous equation. Because the moments are linear integral forms of the distribution function, Eqs. (A19, A20, A21) are essentially linear combinations of the Euler's equations for the generic species and for the single-fluid barycentric description. Unfortunately, these equations are nonlinear with regard to the moments and for this reason it is convenient to reformulate them in the following way:

$$\frac{\partial \rho_\sigma}{\partial t^{(1)}} + \nabla^{(1)} \cdot \left[\rho_\sigma \mathbf{u}_{\alpha(\sigma)}\right] = 0, \tag{A25}$$

$$\begin{aligned} \frac{\partial}{\partial t^{(1)}} \left[\rho_\sigma \mathbf{u}_{\alpha(\sigma)}\right] \; + \; \nabla^{(1)} \cdot \left[\rho_\sigma \mathbf{u}_{\alpha(\sigma)} \otimes \mathbf{u}_{\alpha(\sigma)} + \alpha_\sigma \left(1-\alpha_\sigma\right) \rho_\sigma \mathbf{w}_\sigma \otimes \mathbf{w}_\sigma\right] = \\ - \; \nabla^{(1)} \left(\rho_\sigma e_\sigma\right) + \rho_\sigma \mathbf{g}_\sigma - \mathbf{q}_\sigma^{(1)}, \end{aligned} \tag{A26}$$

$$\begin{aligned} &\frac{\partial}{\partial t^{(1)}} \left(\rho_\sigma e_\sigma\right) + \frac{1}{2} \frac{\partial}{\partial t^{(1)}} \left[\rho_\sigma \mathbf{u}_{\alpha(\sigma)}^2 + \alpha_\sigma \left(1-\alpha_\sigma\right) \, \rho_\sigma \mathbf{w}_\sigma^2\right] + \nabla^{(1)} \cdot \left[\rho_\sigma \, e_\sigma \mathbf{u}_{\alpha(\sigma)} \right. \\ &\left. + \, O\left(|\mathbf{u}_{\alpha(\sigma)}|^3\right)\right] = -\nabla^{(1)} \left[\rho_\sigma \, e_\sigma \mathbf{u}_{\alpha(\sigma)}\right] + \rho_\sigma \mathbf{g}_\sigma \cdot \mathbf{u}_{\alpha(\sigma)} - \varphi_\sigma^{(1)}, \end{aligned} \tag{A27}$$

where $\mathbf{u}_{\alpha(\sigma)} = \left(1-\alpha_\sigma\right) \mathbf{u}_\sigma + \alpha_\sigma \mathbf{u}$ is the linearly interpolated velocity. Because we are interested in the low Mach number limit, the terms which involve higher powers of the velocities can be neglected. Applying Eqs. (A25, A26, A27), the following expression can be recovered:

$$\begin{aligned} &- \int m_\sigma \mathbf{v} \otimes \mathbf{v} \, f_\sigma^{(1)} \, d\mathbf{v} = -\alpha_\sigma \, \tau_m \left[\rho_\sigma e_\sigma \nabla \cdot \mathbf{u}_{\alpha(\sigma)} - \mathbf{u}_{\alpha(\sigma)} \cdot \mathbf{q}_\sigma^{(1)} + \varphi_\sigma^{(1)} \right. \\ &\left. + \, \frac{1}{2} \, \alpha_\sigma \left(1-\alpha_\sigma\right) \frac{\partial \left(\rho_\sigma \mathbf{w}_\sigma^2\right)}{\partial t^{(1)}} \right] \mathbf{I} + \alpha_\sigma \, \tau_m \left[\rho_\sigma e_\sigma \nabla \mathbf{u}_{\alpha(\sigma)} + \rho_\sigma e_\sigma \nabla \mathbf{u}_{\alpha(\sigma)}^T \right. \\ &\left. - \, \mathbf{u}_{\alpha(\sigma)} \otimes \mathbf{q}_\sigma^{(1)} - \mathbf{q}_\sigma^{(1)} \otimes \mathbf{u}_{\alpha(\sigma)} + \alpha_\sigma \left(1-\alpha_\sigma\right) \frac{\partial}{\partial t^{(1)}} \left(\rho_\sigma \mathbf{w}_\sigma \otimes \mathbf{w}_\sigma\right) \right] . \end{aligned} \tag{A28}$$

It is interesting to point out that the previous result does not depend on the external force field. If in both scalar and tensorial quadratic forms the effects due to the diffusion velocity are smaller than the effects due to the interpolated velocity, i.e.:

$$\mathbf{u}_{\alpha(\sigma)}^2 \; \gg \; \alpha_\sigma \left(1-\alpha_\sigma\right) \mathbf{w}_\sigma^2, \tag{A29}$$

$$\mathbf{u}_{\alpha(\sigma)} \otimes \mathbf{u}_{\alpha(\sigma)} \; \gg \; \alpha_\sigma \left(1-\alpha_\sigma\right) \mathbf{w}_\sigma \otimes \mathbf{w}_\sigma, \tag{A30}$$

then the time derivatives in the integral (A28) can be neglected. Some hypotheses can also be formulated about the effects due to the moments of the first-order perturbation.

According to the definition (A17), the property (A16) can be reformulated:

$$\sum_\sigma \varphi_\sigma^{(1)} \ll \frac{1}{2}\sum_\sigma \mathbf{q}_\sigma^{(1)} \cdot (\mathbf{u}_\sigma + \mathbf{u}) = \left[\sum_\sigma \mathbf{q}_\sigma^{(1)} \cdot \mathbf{u}_{\alpha(\sigma)}\right]_{\alpha_\sigma=1/2} . \tag{A31}$$

It is well known that both in fully-decoupled systems ($\alpha_\sigma = 0$) and in systems described by the single-fluid approach ($\alpha_\sigma = 1$), the moments of the perturbations do not effect the macroscopic equations. For the intermediate case ($\alpha_\sigma = 1/2$), the previous property demonstrates that the effects due to the lower-order moments of the perturbation prevail. As a first approximation, it is then reasonable to assume that the effects of $\varphi_\sigma^{(1)}$ can be neglected in Eq. (A28). Substituting the simplified form of Eq. (A28) into Eq. (A24) and adding the result to Eq. (A20), the momentum equation for the generic species is obtained:

$$\begin{aligned}
\frac{\partial(\rho_\sigma \mathbf{u}_\sigma)}{\partial t} &+ \nabla \cdot \left[(1-\alpha_\sigma)\,\rho_\sigma \mathbf{u}_\sigma \otimes \mathbf{u}_\sigma + \alpha_\sigma\,\rho_\sigma \mathbf{u} \otimes \mathbf{u}\right.\\
&+ \left.\alpha_\sigma \rho_\sigma \mathbf{u}_{\alpha(\sigma)} \otimes \mathbf{w}_\sigma + \alpha_\sigma \rho_\sigma \mathbf{w}_\sigma \otimes \mathbf{u}_{\alpha(\sigma)}\right] = \\
&- \nabla(\rho_\sigma e_\sigma^c) + \rho_\sigma \mathbf{g}_\sigma - \frac{1}{\tau_m}\,\rho_\sigma \mathbf{w}_\sigma \\
&+ \nabla \cdot \left\{\alpha_\sigma \rho_\sigma e_\sigma \tau_m \left[\nabla \mathbf{u}_{\alpha(\sigma)} + \nabla \mathbf{u}_{\alpha(\sigma)}^T\right]\right\},
\end{aligned} \tag{A32}$$

where e_σ^c is the corrected internal energy and its expression is:

$$e_\sigma^c = e_\sigma - \alpha_\sigma\,\mathbf{w}_\sigma \cdot \mathbf{u}_{\alpha(\sigma)} + \alpha_\sigma\,\tau_m\,e_\sigma \nabla \cdot \mathbf{u}_{\alpha(\sigma)}. \tag{A33}$$

In the derivation of Eq. (A33), it has been assumed that $|\mathbf{q}_\sigma^{(1)}| \gg |\mathbf{q}_\sigma^{(2)}|$ because the perturbations of the distribution function can be considered decreasing corrections of the previous terms in the expansion. In the small Mach number limit, the corrected internal energy coincides with the internal energy, which is the leading term. For this reason, Eq. (A32) reduces to Eq. (10).

APPENDIX B: CHAPMAN-ENSKOG ASYMPTOTIC ANALYSIS OF THE SUGGESTED LATTICE BOLTZMANN MODEL

The Chapman-Enskog asymptotic analysis will be used in order to design a lattice Boltzmann model which recovers the performance of the continuous Hamel's model with second order accuracy in both time and space. Particularly some corrections are needed for removing the unexpected discrete lattice effects. Let us start from the simple lattice Boltzmann model defined by Eqs. (69, 70). The macroscopic equations for the lower-order moments will be discussed. First, the left hand side of Eq. (69) is expanded by a Taylor series in δt up to the second order:

$$\frac{D_\lambda\,\varphi_\sigma^\lambda}{D\,t}+\frac{\delta t}{2}\,\frac{D_\lambda}{D\,t}\,\frac{D_\lambda\,\varphi_\sigma^\lambda}{D\,t}=-\frac{\chi_H}{\tau_\sigma^0}\left[\varphi_\sigma^\lambda-\varphi_\sigma^{e\,\lambda}\right]-\frac{\epsilon}{\tau_m^0}\left[\varphi_\sigma^\lambda-\varphi_{\sigma(m)}^{e\,\lambda}\right]+\frac{1}{\sqrt{e_\sigma}}\,\mathbf{k}_{\alpha(\sigma)}^\lambda\cdot\mathbf{g}_\sigma. \tag{B1}$$

Then let us expand the normalized velocity distribution function φ_σ^λ in terms of a small parameter K, which is proportional to the Knudsen number Kn. The procedure is the same as previously considered for the continuous model and it yields:

$$\varphi_\sigma^\lambda=\varphi_\sigma^{\lambda\,(0)}+K\,\varphi_\sigma^{\lambda\,(1)}+K^2\,\varphi_\sigma^{\lambda\,(2)}+\dots\,. \tag{B2}$$

We can analogously proceed for the partial derivatives given by Eqs. (A2, A3). In this case, it is better to define a substantial derivative for the generic microscopic velocity of the lattice, by grouping together terms with the same order of magnitude:

$$\frac{D_\lambda^{(1)}}{D\,t^{(1)}}=\frac{\partial}{\partial\,t^{(1)}}+\mathbf{v}^\lambda\cdot\nabla^{(1)}. \tag{B3}$$

Substituting the previous expansions in the simple model, a coupled hierarchy system of equations in the powers of K is obtained and the first elements of this system are:

$$\frac{D_\lambda^{(1)}\,\varphi_\sigma^{\lambda\,(0)}}{D\,t^{(1)}}=-\frac{1}{\alpha_\sigma\,\tau_m}\,\varphi_\sigma^{\lambda\,(1)}+\frac{1}{\sqrt{e_\sigma}}\,\mathbf{k}_{\alpha(\sigma)}^\lambda\cdot\mathbf{g}_\sigma, \tag{B4}$$

$$\frac{\partial\varphi_\sigma^{\lambda\,(0)}}{\partial\,t^{(2)}}+\frac{D_\lambda^{(1)}\,\varphi_\sigma^{\lambda\,(1)}}{D\,t^{(1)}}+\frac{\delta t}{2}\,\frac{D_\lambda^{(1)}}{D\,t^{(1)}}\,\frac{D_\lambda^{(1)}\,\varphi_\sigma^{\lambda\,(0)}}{D\,t^{(1)}}=-\frac{1}{\alpha_\sigma\,\tau_m}\,\varphi_\sigma^{\lambda\,(2)}. \tag{B5}$$

Particularly the last term in the left hand side of Eq. (B5) can be simplified by considering Eq. (B4):

$$\frac{\partial\varphi_\sigma^{\lambda\,(0)}}{\partial\,t^{(2)}}+d_\sigma\,\frac{D_\lambda^{(1)}\,\varphi_\sigma^{\lambda\,(1)}}{D\,t^{(1)}}=-\frac{1}{\alpha_\sigma\,\tau_m}\,\varphi_\sigma^{\lambda\,(2)}-\frac{\delta t}{2\,\sqrt{e_\sigma}}\,\frac{D_\lambda^{(1)}}{D\,t^{(1)}}\,\left[\mathbf{k}_{\alpha(\sigma)}^\lambda\cdot\mathbf{g}_\sigma\right],$$

where

$$d_\sigma = 1 - \frac{1}{2}\,\frac{\delta t}{\alpha_\sigma\,\tau_m} = 1 - \frac{\delta t}{2}\left(\frac{\chi_H}{\tau_\sigma^0} + \frac{\epsilon}{\tau_m^0}\right). \tag{B6}$$

Recovering the macroscopic equations requires multiplying the previous equations by the collisional invariants and then performing the integration over the microscopic velocity. Because Eq. (B4) is analogous to Eq. (A9) for the continuous model, the same results are obtained and the macroscopic equations (A19, A20) still hold for the simple model. The effects of the first-order perturbation on the continuity equation, involve the following sum:

$$\frac{1}{\alpha_\sigma\,\tau_m}\sum_{\lambda=0}^{8}\varsigma^\lambda\,\mathbf{v}^\lambda\,\varphi_\sigma^{\lambda\,(1)}, \tag{B7}$$

which is equivalent to the vector $\mathbf{q}_\sigma^{(1)}$ for the continuous model. Eq. (A15) can be easily generalized to calculate the previous quantity. Particularly since this quantity can not depend on α_σ, it can not depend on the discrete lattice effects and this means that it must coincide with the vector $\mathbf{q}_\sigma^{(1)} = \rho_\sigma\mathbf{w}_\sigma/\tau_m$. An equivalent way to obtain the same result is to suppose that the diffusion velocity $\mathbf{w}_\sigma$ is a first-order term with regard to the parameter K:

$$\frac{1}{\alpha_\sigma\,\tau_m}\sum_{\lambda=0}^{8}\varsigma^\lambda\,\mathbf{v}^\lambda\sum_{\xi=1}^{\infty}K^\xi\,\varphi_\sigma^{\lambda\,(\xi)} = \frac{1}{\tau_m}\,\rho_\sigma\,\mathbf{w}_\sigma\,K. \tag{B8}$$

Both approaches allow us to analyze the effects of the first-order perturbation on the continuity equation:

$$\frac{\partial\rho_\sigma}{\partial t^{(2)}} = -d_\alpha\,\alpha_\sigma\,\nabla^{(1)}\cdot(\rho_\sigma\,\mathbf{w}_\sigma) - \frac{\delta t}{2}\,\nabla^{(1)}\cdot(\rho_\sigma\,\mathbf{g}_\sigma)\,. \tag{B9}$$

Summing the previous equation with Eq. (A19), the continuity equation for the simple model is obtained:

$$\frac{\partial\rho_\sigma}{\partial t} + \nabla\cdot(\rho_\sigma\mathbf{u}_\sigma) = \frac{\delta t}{2}\,\nabla\cdot(\rho_\sigma\,\mathbf{l}_\sigma)\,, \tag{B10}$$

where $\mathbf{l}_\sigma = \mathbf{w}_\sigma/\tau_m - \mathbf{g}_\sigma$ is the difference between the acceleration due to the internal coupling force and the external force field.

Proceeding in the same way for the momentum equation. Multiplying Eq. (B6) by the particle momentum for the generic species and integrating over the microscopic velocity, the following equation is recovered:

$$\begin{aligned}\frac{\partial}{\partial t^{(2)}}\left[\rho_\sigma\left(\mathbf{u}_\sigma - \alpha_\sigma\mathbf{w}_\sigma\right)\right] \;+\; \alpha_\sigma\,\frac{\partial}{\partial t^{(1)}}\left(\rho_\sigma\mathbf{w}_\sigma\right) &= \frac{\delta t}{2}\,\frac{\partial}{\partial t^{(1)}}\left(\rho_\sigma\,\mathbf{l}_\sigma\right)\\ &-\;\frac{\delta t}{2}\,\nabla^{(1)}\cdot\left[\rho_\sigma\mathbf{u}_{\alpha(\sigma)}\otimes\mathbf{g}_\sigma + \rho_\sigma\mathbf{g}_\sigma\otimes\mathbf{u}_{\alpha(\sigma)}\right]\\ &-\;d_\sigma\,\nabla^{(1)}\cdot\left(\int m_\sigma\mathbf{v}\otimes\mathbf{v}\,f_\sigma^{(1)}\,d\mathbf{v}\right),\end{aligned} \tag{B11}$$

where the effects of the higher-order perturbations have been neglected. Applying Eqs. (A25, A26, A27) and supposing that the effects due to both scalar and tensorial quadratic forms of the diffusion velocity are smaller than the effects due to the linearly interpolated velocity, then:

$$\begin{aligned} -\int m_\sigma \mathbf{v}\otimes\mathbf{v}\, f_\sigma^{(1)}\, d\mathbf{v} \;&=\; \rho_\sigma\,(e_\sigma - e_\sigma^c)\,\mathbf{I} + \alpha_\sigma\,\tau_m\left[\rho_\sigma e_\sigma \nabla\mathbf{u}_{\alpha(\sigma)} + \rho_\sigma e_\sigma \nabla\mathbf{u}_{\alpha(\sigma)}^T\right.\\ &-\;\left.\rho_\sigma\mathbf{u}_{\alpha(\sigma)}\otimes\mathbf{w}_\sigma/\tau_m - \rho_\sigma\mathbf{w}_\sigma\otimes\mathbf{u}_{\alpha(\sigma)}/\tau_m\right]. \end{aligned} \tag{B12}$$

Considering the previous result, summing Eq. (B11) with Eq. (A20), the momentum equation for the simple model is obtained:

$$\begin{aligned} \frac{\partial\left(\rho_\sigma\mathbf{u}_\sigma\right)}{\partial t} \;&+\; \nabla\cdot\left[(1-\alpha_\sigma)\,\rho_\sigma\mathbf{u}_\sigma\otimes\mathbf{u}_\sigma + \alpha_\sigma\,\rho_\sigma\mathbf{u}\otimes\mathbf{u} + \alpha_\sigma\rho_\sigma\mathbf{u}_{\alpha(\sigma)}\otimes\mathbf{w}_\sigma\right.\\ &+\;\left.\alpha_\sigma\rho_\sigma\mathbf{w}_\sigma\otimes\mathbf{u}_{\alpha(\sigma)}\right] = -\nabla\left(\rho_\sigma e_\sigma\right) + \rho_\sigma\mathbf{g}_\sigma - \frac{1}{\tau_m}\,\rho_\sigma\mathbf{w}_\sigma\\ &+\;\nabla\cdot\left\{d_\sigma\,\alpha_\sigma\rho_\sigma e_\sigma\tau_m\left[\nabla\mathbf{u}_{\alpha(\sigma)} + \nabla\mathbf{u}_{\alpha(\sigma)}^T\right]\right\} + \frac{\delta t}{2}\,\frac{\partial}{\partial t}\left(\rho_\sigma\,\mathbf{l}_\sigma\right)\\ &+\;\frac{\delta t}{2}\,\nabla\cdot\left[\rho_\sigma\,\mathbf{u}_{\alpha(\sigma)}\otimes\mathbf{l}_\sigma + \rho_\sigma\,\mathbf{l}_\sigma\otimes\mathbf{u}_{\alpha(\sigma)}\right], \end{aligned} \tag{B13}$$

where $e_\sigma^c \approx e_\sigma$ has been assumed. Comparing Eqs. (B10, B13) with the macroscopic equations of the continuous Hamel's model given by Eqs. (9, 10), the discrete lattice effects are evident. Although the macroscopic equations of the simple model recover the equations of the continuous model when $\delta t \to 0$, the simple model would be only accurate up to the first order. For improving the accuracy up to the second order, some corrections are needed. A recently suggested method for recovering the correct hydrodynamic equations will be generalized for the mixtures [49].

Let us introduce the following corrected velocities:

$$\rho_\sigma\,\mathbf{u}_\sigma^\star = \sum_{\lambda=0}^{8} \varsigma^\lambda\,\mathbf{v}^\lambda\,\varphi_\sigma^\lambda + \rho_\sigma\,\mathbf{t}_\sigma^\star\,\delta t, \tag{B14}$$

where $\mathbf{t}_\sigma^\star$ is an auxiliary vector. Consequently the corrected barycentric velocity $\mathbf{u}^\star = \sum_\sigma x_\sigma\mathbf{u}_\sigma^\star$ is defined. Similarly the corrected equilibrium distribution function $\varphi_\sigma^{e\,\lambda\star}$ centered on the specific velocity $\mathbf{u}_\sigma^\star$ and the corrected equilibrium distribution function $\varphi_{\sigma(m)}^{e\,\lambda\star}$ centered on the barycentric velocity $\mathbf{u}^\star$ can be obtained. Let us introduce the following guessed lattice Boltzmann model:

$$\begin{aligned} \frac{D_\lambda\,\varphi_\sigma^\lambda}{D\,t} + \frac{\delta t}{2}\,\frac{D_\lambda}{D\,t}\,\frac{D_\lambda\,\varphi_\sigma^\lambda}{D\,t} \;&=\; -\;\frac{\chi_H}{\tau_\sigma^0}\left[\varphi_\sigma^\lambda - \varphi_\sigma^{e\,\lambda\star}\right] - \frac{\epsilon}{\tau_m^0}\left[\varphi_\sigma^\lambda - \varphi_{\sigma(m)}^{e\,\lambda\star}\right]\\ &+\;\frac{1}{\sqrt{e_\sigma}}\,\mathbf{k}_{\alpha(\sigma)}^{\lambda\star}\cdot\mathbf{g}_\sigma + \Theta_\sigma^{\lambda\star}, \end{aligned} \tag{B15}$$

where $\mathbf{k}^{\lambda\star}_{\alpha(\sigma)}$ is the generalization of Eq. (70) when the corrected velocities are considered. The additional corrective factor $\Theta^{\lambda\star}_{\sigma}$ is defined as

$$\Theta^{\lambda\star}_{\sigma} = \rho_\sigma \left[-\frac{\delta t}{\alpha_\sigma \, \tau_m} \, \frac{\mathbf{t}^{\star}_{\sigma} \cdot \mathbf{v}^{\lambda}}{e_\sigma} + \frac{\mathbf{T}^{\star}_{\sigma} : \left(\mathbf{v}^{\lambda} \otimes \mathbf{v}^{\lambda} - e_\sigma \, \mathbf{I} \right)}{2 \, e_\sigma^2} \right], \tag{B16}$$

where $\mathbf{T}^{\star}_{\sigma}$ is an auxiliary tensor. The previous corrections to the simple model do not effect the first term of the expansion, i.e. $\varphi^{\lambda\,(0)\,\star}_{\sigma} = (1-\alpha_\sigma)\,\varphi^{e\,\lambda\,\star}_{\sigma} + \alpha_\sigma\,\varphi^{e\,\lambda\,\star}_{\sigma(m)}$. Using the previous result, the definition of the diffusion velocity, Eq. (B14) and assuming that the additional term in the corrected velocities is of the first order in the Knudsen number because it is multiplied by the discretization time step, the property given by Eq. (B8) can be generalized:

$$\frac{1}{\alpha_\sigma \, \tau_m} \sum_{\lambda=0}^{8} \varsigma^{\lambda} \, \mathbf{v}^{\lambda} \sum_{\xi=1}^{\infty} K^{\xi} \, \varphi^{\lambda\,(\xi)\,\star}_{\sigma} = \left(\frac{1}{\tau_m} \, \rho_\sigma \, \mathbf{w}^{\star}_{\sigma} - \frac{1}{\alpha_\sigma \, \tau_m} \, \rho_\sigma \, \mathbf{t}^{\star}_{\sigma} \, \delta t \right) K. \tag{B17}$$

The previous corrections have been designed in such a way as to preserve the macroscopic Eqs. (A19, A20), if the velocities of the mixture components are redefined according to Eq. (B14). Proceeding in the usual way, the effects of the first-order perturbation on the continuity equation can be analyzed and summing this result to Eq. (A19) the final form of the continuity equation is obtained:

$$\frac{\partial \rho_\sigma}{\partial t} + \nabla \cdot \left(\rho_\sigma \mathbf{u}^{\star}_{\sigma} \right) = \delta t \, \nabla \cdot \left[\rho_\sigma \, \left(\mathbf{l}^{\star}_{\sigma}/2 + \mathbf{t}^{\star}_{\sigma} \right) \right]. \tag{B18}$$

The auxiliary vector can be set in such a way as to reproduce the performance of the continuous model with second order accuracy, i.e. $\mathbf{t}^{\star}_{\sigma} = -\mathbf{l}^{\star}_{\sigma}/2$.

Similarly the effects of the first-order perturbation on the momentum equation can be analyzed and summing this result to Eq. (A20) the final form of the momentum equation is obtained:

$$\begin{aligned}
\frac{\partial \left(\rho_\sigma \mathbf{u}_\sigma \right)}{\partial t} & + \nabla \cdot \left[(1-\alpha_\sigma) \, \rho_\sigma \mathbf{u}^{\star}_{\sigma} \otimes \mathbf{u}^{\star}_{\sigma} + \alpha_\sigma \, \rho_\sigma \mathbf{u}^{\star} \otimes \mathbf{u}^{\star} + \alpha_\sigma \rho_\sigma \mathbf{u}^{\star}_{\alpha(\sigma)} \otimes \mathbf{w}^{\star}_{\sigma} \right. \\
& + \left. \alpha_\sigma \rho_\sigma \mathbf{w}^{\star}_{\sigma} \otimes \mathbf{u}^{\star}_{\alpha(\sigma)} \right] = -\nabla \left(\rho_\sigma e_\sigma \right) + \rho_\sigma \mathbf{g}_\sigma - \frac{1}{\tau_m} \, \rho_\sigma \mathbf{w}^{\star}_{\sigma} \\
& + \nabla \cdot \left\{ d_\sigma \, \alpha_\sigma \rho_\sigma e_\sigma \tau_m \left[\nabla \mathbf{u}^{\star}_{\alpha(\sigma)} + \nabla \mathbf{u}^{\star T}_{\alpha(\sigma)} \right] \right\} \\
& + \frac{1}{2} \nabla \cdot \left[\delta t \, \rho_\sigma \, \mathbf{u}^{\star}_{\alpha(\sigma)} \otimes \mathbf{l}^{\star}_{\sigma} + \delta t \, \rho_\sigma \, \mathbf{l}^{\star}_{\sigma} \otimes \mathbf{u}^{\star}_{\alpha(\sigma)} \right. \\
& - \left. \alpha_\sigma \, \tau_m \, \rho_\sigma \, \left(\mathbf{T}^{\star}_{\sigma} + \mathbf{T}^{\star T}_{\sigma} \right) \right].
\end{aligned} \tag{B19}$$

The auxiliary tensor can be set in such a way to reproduce the performance of the continuous model with second order accuracy, i.e. $\mathbf{T}^{\star}_{\sigma} = \delta t \, \mathbf{u}^{\star}_{\alpha(\sigma)} \otimes \mathbf{l}^{\star}_{\sigma} / \left(\alpha_\sigma \, \tau_m \right)$. The previous results can be included in the definition of the corrective factor:

$$\Theta_\sigma^{\lambda\star} = \rho_\sigma \, \frac{\delta t}{2\,\alpha_\sigma\,\tau_m} \left[\frac{\mathbf{l}_\sigma^\star \cdot \mathbf{v}^\lambda}{e_\sigma} - \frac{\mathbf{u}_{\alpha(\sigma)}^\star \otimes \mathbf{l}_\sigma^\star : \left(\mathbf{v}^\lambda \otimes \mathbf{v}^\lambda - e_\sigma\,\mathbf{I}\right)}{e_\sigma^2} \right]. \tag{B20}$$

It is easy to verify that $\Theta_\sigma^{\lambda\star} = (1 - d_\sigma)\,\mathbf{k}_{\alpha(\sigma)}^{\lambda\star} \cdot \mathbf{l}_\sigma^\star/\sqrt{e_\sigma}$. Substituting this result into the corrected Eq. (B16), the final lattice Boltzmann model given by Eq. (72) is recovered. It is interesting to highlight that for non-interacting particles, i.e. when $1/\tau_m \rightarrow 0$, the discussed correction reduces to the well-known formula for the external force field [49].

[1] G.A. Bird, *Molecular Gas Dynamics and the Direct Simulation of Gas Flows*, Oxford University Press, Oxford (1994).

[2] C. Cercignani, *The Boltzmann Equation and its Application*, Springer, New York (1988).

[3] Y. Sone, *Kinetic Theory and Fluid Dynamics*, Birkhäuser, Boston (2002).

[4] J.H. Ferziger, H.G. Kaper, *Mathematical Theory of Transport Processes in Gases*, North-Holland, Amsterdam (1972).

[5] G.R. McNamara, G. Zanetti, "Use of the Boltzmann equation to simulate lattice-gas automata", Phys. Rev. Lett. **61**, 2332 (1988).

[6] Y.H. Qian, D. D'Humieres, P. Lallemand, "Lattice BGK models for Navier-Stokes equation", Europhys. Lett. **17**, 479 (1992).

[7] X. He, L.-S. Luo, "Theory of the lattice Boltzmann method: From the Boltzmann equation to the lattice Boltzmann equation", Phys. Rev. E **56**, 6811 (1997).

[8] H. Chen, S. Chen, W. H. Matthaeus, "Recovery of the Navier-Stokes equations using a lattice-gas Boltzmann method", Phys. Rev. A **45**, R5339 (1992).

[9] D. Wolf-Gladrow, *Lattice-Gas Cellular Automata and Lattice Boltzmann Models*, Lecture Notes in Mathematics Vol. 1725, Springer-Verlag, Berlin (2000).

[10] S. Succi, *The Lattice Boltzmann Equation for Fluid Dynamics and Beyond*, Oxford University Press, New York (2001).

[11] G.D Doolen (editor), *Lattice Gas Methods for Partial Differential Equations*, Addison-Wesley, New York (1990).

[12] S. Chen, G. D. Doolen, "Lattice Boltzmann method for fluid flow", Annu. Rev. Fluid Mech. **30**, 329 (1998).

[13] B. Manz, L.F. Gladden, P.B. Warren, "Flow and dispersion in porous media: lattice-Boltzmann and NMR studies", AIChE. J. **45**, 1845 (1999).

[14] Th. Zeiser, P. Lammers, E. Klemm, Y.W. Li, J. Bernsdorf, G. Brenner, "CDF-calculartion of flow, dispersion and reaction in a catalyst filled tube by the lattice Boltzmann method", Chem. Eng. Science, **56**, 1697 (2001).

[15] X. Shan, H. Chen, "Lattice Boltzmann model for simulating flows with multiple phases and components", Phys. Rev. E **47**, 1815 (1993).

[16] E.G. Flekkoy, "Lattice bhatnagar-gross-krook models for miscible fluids", Phys. Rev. E **47**, 4247 (1993).

[17] X. Shan, G. Doolen, "Multicomponent lattice-Boltzmann model with interparticle interaction", J. Stat. Phys. **81**, 379 (1995).

[18] X. Shan, G. Doolen, "Diffusion in a multicomponent lattice Boltzmann equation model", Phys. Rev. E **54**, 3614 (1996).

[19] E. Orlandini, W.R. Osborn, J.M. Yeomans, "A lattice Boltzmann model of binary-fluid mixtures", Europhys. Lett. **32**, 463 (1995).

[20] W.R. Osborn, E. Orlandini, M.R. Swift, J.M. Yeomans, J.R. Banavar, "Lattice Boltzmann study of hydrodynamic spinodal decomposition", Phys. Rev. Lett. **75**, 4031 (1995).

[21] M.R. Swift, E. Orlandini, W.R. Osborn, J.M. Yeomans, "Lattice Boltzmann simulations of liquid-gas and binary fluid systems", Phys. Rev. E **54**, 5041 (1996).

[22] A. Lamura, G. Gonnella, J.M. Yeomans, "A Lattice-Boltzmann model of ternary-fluid mixtures", Europhys. Lett. **45**, 314 (1999).

[23] V. Sofonea, R.F. Sekerka, "BGK models for diffusion in isothermal binary fluid systems", Physica A **299**, 494 (2001).

[24] Z. Guo, T.S. Zhao, "Discrete velocity and lattice Boltzmann models for binary mixtures of nonideal fluids", Phys. Rev. E **68**, 035302 (2003).

[25] L.-S. Luo, S.S. Girimaji, "Lattice Boltzmann model for binary mixtures", Phys. Rev. E **66**, R035301 (2002).

[26] L.-S. Luo, S.S. Girimaji, "Theory of the lattice Boltzmann method: Two-fluid model for binary mixtures", Phys. Rev. E **67**, 036302 (2003).

[27] A. Xu, "Finite-Difference Lattice Boltzmann Methods for binary fluids", arXiv:cond-mat/0406012, www.arxiv.org (2004).

[28] P.C. Fancin, P.C. Philippi, L.O.E. dos Santos, "Non-linear lattice-Boltzmann model for ideal miscible fluids", FGCS J. **20**, 945 (2004).

[29] K. Xu, "BGK-Based Scheme for Multicomponent Flow Calculations", J. Comput. Phys. **134**, 122 (1997).

[30] Y.S. Lian, K. Xu, "A Gas-Kinetic Scheme for Multimaterial Flows and Its Application in Chemical Reactions", J. Comput. Phys. **163**, 349 (2000).

[31] S. Chapman, T.G. Cowling, *The Mathematical Theory of Non-Uniform Gases*, Cambridge

University Press, Cambridge (1970).

[32] S. Harris, *An Introduction to the Theory of the Boltzmann Equation*, Holt, Rinehart and Winston, New York (1971).

[33] X. He, X. Shan, G. D. Doolen, "Discrete Boltzmann equation model for nonideal gases", Phys. Rev. E **57**, R13 (1998).

[34] T. Abe, "Derivation of the Lattice Boltzmann Method by Means of the Discrete Ordinate Method for the Boltzmann Equation", J. Comput. Phys. **131**, 241 (1997).

[35] X. Shan, X. He, "Discretization of the velocity space in the solution of the Boltzmann equation", Phys. Rev. Lett. **80**, 65 (1998).

[36] I. Kolodner, "On the Application of the Boltzmann Equations to the Theory of Gas Mixtures", Ph.D. dissertation, New York University (1950).

[37] P.L. Bhatnagar, E.P. Gross, M. Krook, "A model for collision processes in gases. I. Small amplitude processes in charged and neutral one-component systems", Phys. Rev. **94**, 511 (1954).

[38] E.P. Gross, M. Krook, "Model for collision processes in gases: small-amplitude oscillations of charged two-Component systems", Phys. Rev. **102**, 593 (1956).

[39] L. Sirovich, "Kinetic modelling of gas mixtures", Phys. Fluids **5**, 908 (1962).

[40] E. Goldman, L. Sirovich, "Equations for gas mixtures", Phys. Fluids **10**, 908 (1967).

[41] T.F. Morse, "Kinetic model equations for a gas mixture",Phys. Fluids **7**, 2012 (1964).

[42] B.B. Hamel, "Boltzmann Equations for Binary Gas Mixtures", Ph.D. dissertation, Princeton University (1963).

[43] B.B. Hamel, "Kinetic model of binary mixtures", Phys. Fluids **8**, 418 (1965).

[44] B.B. Hamel, "Kinetic Model for Binary Gas Mixtures", Phys. Fluids **9**, 12 (1966).

[45] F.J. McCormack, "Construction of linearized kinetic models for gaseous mixtures and molecular gases", Phys. Fluids **16**, 2095 (1973).

[46] V. Garzó, A. Santos, J.J. Brey, "A kinetic model for a multicomponent gas", Phys. Fluids **1**, 380 (1989).

[47] P. Andries, K. Aoki, B. Perthame, "A Consistent BGK-Type Model for Gas Mixtures", J. Stat. Phys. **106**, 993 (2002).

[48] R.C. Reid, *The properties of gases and liquids*, McGraw-Hill, New York (1987).

[49] Z. Guo, C. Zheng, B. Shi, "Discrete lattice effects on the forcing term in the lattice Boltzmann

method", Phys. Rev. E **65**, 046308 (2002).

[50] I. Ginzbourg, P. M. Adler, "Boundary flow condition analysis for the three-dimensional lattice Boltzmann model", J. Phys. II France, **4**, 191-214 (1994).

[51] P. Lallemand, L.-S. Luo, "Theory of the lattice Boltzmann method: Dispersion, dissipation, isotropy, Galilean invariance, and stability", Phys. Rev. E, **61**, 6546-6562 (2000).

[52] D. dHumiéres, "Generalized lattice Boltzmann equations", in *Rarefied gas dynamics: theory and simulations* (ed. B. D. Shizgal & D. P. Weaver), Prog. Astronaut. Aeronaut. **159**, 450 (1992).

[53] D. dHumiéres, I. Ginzburg, M. Krafczyky, P. Lallemand, L.-S. Luo, "Multiple-relaxation-time lattice Boltzmann models in three dimensions", Proc. Royal Soc. London **A 360**, 437 (2002).

TABLE I: Comparison between theoretical and simulated results for both diffusivity and mixture kinetic viscosity. The subscript $(\cdot)_T$ means the theoretical values given by Eqs. (87, 89), while the subscript $(\cdot)_M$ means the measured values given by Eqs. (93, 95).

ϵ $[-]$	c $[m/s]$	$\tau_m = \tau_m^0/\epsilon$ $[s]$	$(D_c)_T$ $[m^2/s]$	$(D_c)_M$ $[m^2/s]$	$(\nu_c)_T$ $[m^2/s]$	$(\nu_c)_M$ $[m^2/s]$
0.949	136.33	1.6244×10^{-10}	1.0064×10^{-6}	1.0376×10^{-6}	3.7525×10^{-7}	3.7560×10^{-7}
0.905	131.56	1.7888×10^{-10}	1.0320×10^{-6}	1.0802×10^{-6}	3.5928×10^{-7}	3.5944×10^{-7}
0.860	127.22	1.9793×10^{-10}	1.0679×10^{-6}	1.1211×10^{-6}	3.4489×10^{-7}	3.4488×10^{-7}
0.815	123.28	2.2020×10^{-10}	1.1156×10^{-6}	1.1610×10^{-6}	3.3207×10^{-7}	3.3190×10^{-7}
0.771	119.72	2.4646×10^{-10}	1.1774×10^{-6}	1.2195×10^{-6}	3.2083×10^{-7}	3.2048×10^{-7}
0.726	116.50	2.7770×10^{-10}	1.2563×10^{-6}	1.2768×10^{-6}	3.1117×10^{-7}	3.1079×10^{-7}
0.681	113.61	3.1529×10^{-10}	1.3564×10^{-6}	1.3667×10^{-6}	3.0308×10^{-7}	3.0264×10^{-7}

TABLE II: Superficial velocities $S(u_\sigma^x)$ (averaged values over the whole porous medium) for single components of binary mixtures characterized by different coupling strengths. The critical viscosity is constantly equal to the averaged viscosity based on mass concentrations (case A).

(A) $\nu_c(\epsilon) = \sum_\sigma x_\sigma \nu_\sigma$

ϵ $[-]$	χ_H $[-]$	ν_c $[m^2/s]$	ω_a $[-]$	ω_b $[-]$	ω_m $[-]$	$S(u_a^x)$ $[mm/s]$	$S(u_b^x)$ $[mm/s]$
0.000	1.000	3.750×10^{-6}	1.200	1.462	0.000	3.690	8.305
0.001	0.999	3.750×10^{-6}	1.199	1.460	0.001	3.753	8.196
0.002	0.998	3.750×10^{-6}	1.198	1.459	0.002	3.811	8.115
0.004	0.996	3.750×10^{-6}	1.196	1.457	0.004	3.937	7.943
0.008	0.992	3.750×10^{-6}	1.192	1.452	0.008	4.121	7.702
0.016	0.984	3.750×10^{-6}	1.184	1.442	0.016	4.281	7.323
0.032	0.968	3.750×10^{-6}	1.168	1.422	0.032	4.626	6.617
0.064	0.936	3.750×10^{-6}	1.135	1.382	0.064	4.913	6.187
0.126	0.827	3.750×10^{-6}	1.069	1.302	0.129	5.148	5.848
0.250	0.746	3.750×10^{-6}	0.934	1.138	0.263	5.234	5.567
0.500	0.495	3.750×10^{-6}	0.647	0.788	0.549	5.315	5.475
0.749	0.246	3.750×10^{-6}	0.336	0.410	0.860	5.383	5.424
0.989	0.010	3.750×10^{-6}	0.015	0.018	1.185	5.418	5.418

TABLE III: Superficial velocities $S(u^x_\sigma)$ (averaged values over the whole porous medium) for single components of binary mixtures characterized by different coupling strengths. The critical viscosity varies according to the coupling strength with the assumed linear law (case B).

(B) $\nu_c(\epsilon) = (1-\epsilon)\sum_\sigma x_\sigma \nu_\sigma + \epsilon\, \nu_m \leq \sum_\sigma x_\sigma \nu_\sigma$

ϵ $[-]$	χ_H $[-]$	ν_c $[m^2/s]$	ω_a $[-]$	ω_b $[-]$	ω_m $[-]$	$S(u^x_a)$ $[mm/s]$	$S(u^x_b)$ $[mm/s]$
0.000	1.000	3.750×10^{-6}	1.200	1.462	0.000	3.690	8.305
0.001	0.999	3.749×10^{-6}	1.199	1.461	0.001	3.736	8.253
0.002	0.998	3.748×10^{-6}	1.199	1.460	0.002	3.776	8.173
0.003	0.997	3.747×10^{-6}	1.198	1.459	0.003	3.851	8.081
0.006	0.993	3.745×10^{-6}	1.195	1.458	0.005	3.977	7.903
0.012	0.987	3.739×10^{-6}	1.190	1.450	0.009	4.178	7.650
0.024	0.973	3.729×10^{-6}	1.180	1.437	0.019	4.523	7.226
0.048	0.946	3.708×10^{-6}	1.160	1.413	0.038	4.850	6.566
0.096	0.893	3.666×10^{-6}	1.119	1.362	0.077	5.217	6.250
0.191	0.791	3.582×10^{-6}	1.032	1.257	0.160	5.544	6.101
0.382	0.590	3.414×10^{-6}	0.839	1.021	0.349	6.043	6.261
0.573	0.398	3.246×10^{-6}	0.616	0.750	0.570	6.468	6.617
0.764	0.214	3.078×10^{-6}	0.361	0.440	0.827	7.099	7.174
0.994	0.006	2.875×10^{-6}	0.011	0.013	1.189	7.662	7.662

FIG. 1: The figure shows the randomly generated porous medium considered in the numerical calculations. Periodic boundary conditions in both directions are assumed. Only a single component for a generic mixture is considered. The species flow is due to a given pressure gradient. The lighter regions are characterized by higher velocities.

FIG. 2: Superficial velocity for the components of the binary mixtures flowing in a randomly generated porous medium. Two cases are considered: in the first case, the fully-coupled mixture viscosity is set equal to the mass averaged viscosity and in the second case it is set equal to the generic experimental viscosity.