

Macroscopic theory of multicomponent flows: irreversibility and well-posed equations

Preprint of the paper published in *Physica D: Nonlinear Phenomena* 225, 94-111 (2007)

A.W. Vreman

Vreman Research
Godfried Bomansstraat 46
7552 NT Hengelo
The Netherlands

e-mail: bert@vremanresearch.nl

Abstract

A theory of the coupling of N systems of Navier-Stokes equations is presented. A set of coupled Navier-Stokes systems contains the macroscopic equations of motion for flows of N different material components or phases. Six postulates are formulated from which three Navier-Stokes sets are derived for arbitrary N . The main postulate concerns the second law of thermodynamics, which the present sets satisfy for arbitrary N . Appropriate averages of velocities and pressures distribute the entropy production of each irreversible interaction process among the interacting components. Increase of entropy is guaranteed if the interaction terms are suitably constructed from positive definite operators. A strictly irreversible mass exchange law, expressed in the difference of Gibbs free energies of the exchanging components, is proposed. Furthermore, the characteristic velocities of each Navier-Stokes set are investigated, since their realness is required for well-posedness of the equations. For both Navier-Stokes sets with multiple pressures, the characteristic velocities are proven to be real for arbitrary N . The canonical problem of complex characteristics of incompressible multiphase flows with a single pressure is revisited. A solution to this problem is found by generalizing the standard algebraic Stokes drag law to a positive definite operator acting on the velocity difference. In this way, effects of forces due to nonuniform velocity fields can be incorporated. A generalized drag law is specified by the inclusion of m -th order spatial derivatives of the velocity differences between two components ($m \geq 2$). The characteristic polynomial is derived for an arbitrary number of components and its roots, the characteristic velocities, appear to be real. This is rigorously proven for two- and three-phase flows, and conjectured to be true for flows with four or more components. The linear stability of the incompressible one-dimensional case is also investigated. Ill-posedness of the standard case leads to an infinite growth rate for infinitely short waves. In the case of generalized drag, this growth rate becomes finite for even and neutral for odd m .

Keywords: Entropy inequality; Multicomponent flows; Multiphase flows; Intrafacial interaction terms; Well-posedness; Characteristic velocities.

1 Introduction

The equations which describe the motion of a single Newtonian fluid are the well-known Navier-Stokes equations. These equations are formulated from basic physical principles: conservation of mass, momentum and energy. The total entropy of the system increases, consistent with the second law of thermodynamics. If the fluid is incompressible the total kinetic energy decreases due to viscous dissipation. The equations linearized around constant fields are also mathematically well-posed in the sense of Hadamard. Well-posedness means existence and uniqueness of solutions and continuous dependence of the initial data in appropriate spaces. Linear equations are well-posed only if the eigenvalues (characteristic velocities) are real.

Many flows in nature and technology contain more than one component or phase. Flows relevant in chemistry, biology or geophysics, for example, are often multicomponent or multiphase flows. Whereas there is general consensus about the structure of the Navier-Stokes equations for a single fluid, basic issues concerning the structure of the equations of motion for multiphase flows are much less resolved. A macroscopic theory usually assumes that the motion of the N components are governed by a collection of N interacting Navier-Stokes systems, which we call a Navier-Stokes set. The individual systems are connected through forcing terms which represent the exchange of mass, momentum and energy [15, 28, 21, 11, 9, 17, 24]. As the number of unknowns is larger than the number of equations, closure assumptions have to be made in order to complete the theory. At this point numerous problems arise. The most fundamental and unresolved problems are the violation of the second law of thermodynamics and model equations which are mathematically ill-posed [25, 10, 11, 24]. Let us consider these two important fundamental problems in more detail.

First, it is unclear whether there exists a multicomponent description which guarantees the second law of thermodynamics to be satisfied for arbitrary N [28, 11, 27]. In fact complete descriptions which satisfy the entropy law are known for $N = 2$ only [26, 3, 6, 22]. These works define a pressure for each phase and an additional equation which relates the difference of these pressures to the evolution of the volume fraction [7, 14, 21]. The present paper will present several Navier-Stokes sets which satisfy the second law of thermodynamics for arbitrary N . Appropriate averages of velocities and pressures [6] will be incorporated to distribute the entropy produced by the irreversible interaction processes among the participating phases.

The second fundamental problem is that even the linearized versions of many systems of equations for two-phase flows are known to be ill-posed in the sense of Hadamard [25, 10, 11, 24], that is the equations linearized around a constant field have complex valued characteristic velocities. The initial boundary-value problem cannot be solved; it is ill-posed in the sense of Hadamard. The problem already occurs in the most simple system of multiphase flow: a system describing inviscid flows by two incompressible components, different velocities, equal pressures and interaction through the Stokes drag force. Complex characteristics correspond to an infinitely large growth rate for large wavenumbers ($k \rightarrow \infty$) and occur if the velocities of two components are unequal.

The introduction of standard viscous terms makes the canonical case with one pressure well-posed [2]. Another possibility is to use two pressures and to prescribe the pressure difference by an additional equation. Ramshaw and Trapp [25] found real characteristics in case the pressure difference acts as a surface tension. Ransom and Hicks [26] and Baer and Nunziato [3] proved that the characteristics are real if the pressure difference is governed by the volume fraction, or equivalently, if the evolution of the volume fraction is controlled by the pressure difference. Nevertheless the fundamental question remains (e.g. [11]): why is the most simple formulation, incompressible flow, single pressure and drag force ill-posed? Why should viscosity necessarily play a fundamental role, except within the drag law? Or why should a pressure relaxation term that controls the volume fraction be essential? It is important to realize that the canonical problem is not always ill-posed; it is well-posed if the velocities of the two phases are equal. Unlike the approaches mentioned above, we investigate regularizations which vanish for equal velocities by expressing the additional terms in velocity differences. As the drag law is expressed in velocity differences it may be natural to regard these terms as extensions to standard drag, provided that they do not distort the dissipative property of the drag law. This condition is ensured by a formulation of interaction terms in terms of positive definite operators.

It will be shown that the canonical problem of complex characteristics can be solved by a modification of the drag law. It is remarked that the standard Stokes drag law holds for isolated bodies and assumes an undisturbed free-stream velocity. To circumvent this limitation it is proposed to include high-order derivatives of velocity differences into the drag, which physically represents the effects of a non-uniform flow on embedded particles. In case of second-order derivatives, the extension may be interpreted as a

generalization of the so-called Faxen force. Care is taken to ensure that the extension does not violate the entropy law.

The property of real characteristics does not exclude the occurrence of instabilities, which may represent physical Kelvin Helmholtz instabilities [25, 23, 16]. It is known that the linear growth-rate of the inviscid instability of the hyperbolic tangent velocity profile is finite and that the corresponding wavelength is seven times the shear layer thickness [19]. As a consequence, the instability of the wake of a particle in multiphase flow will have a fundamental wavelength of a few particle diameters, provided the particle Reynolds number is sufficiently high. Apparently, in a macroscopic description, physical instabilities may occur at very short wavelengths.

In practical situations, not only real characteristics, but also a non-positive growth rate for sufficiently large spatial wave numbers k is desirable. Positive growth rates for $k \rightarrow \infty$ can be prevented by the inclusion of surface tension [25]. Viscosity does not always prevent positive growth rates for $k \rightarrow \infty$, but only if the phases are compressible [2]. Likewise, multiple pressure approaches may allow positive growth rates for infinite k in the incompressible limit [23]. There is thus sufficient reason to subject the present Navier-Stokes sets to a linear stability analysis, in which we will focus on the limit $k \rightarrow \infty$.

The organization of the present paper is as follows. Six postulates will be formulated and the basic formulation of a Navier-Stokes set will be given (section 2). From these postulates, three possibilities for the reversible (nozzling) parts of the interaction terms are naturally found in section 3, from which three different Navier-Stokes sets will emerge (A-C). The generalized drag law, expressed as a positive definite operator, will be formulated in section 4. In section 5 we will consider more irreversible processes and prove that the second law of thermodynamics is satisfied for suitable forms of the interactions terms. The issue of well-posedness will be investigated in section 6 and will turn out that the three Navier-Stokes sets have real characteristics. The linear stability analysis will be performed in section 7. The incorporation of external forces, an entropy increasing mass exchange law, and pressure jumps will be postponed until section 8, to keep the derivations and equations in previous sections more concise and clear. Finally, section 9 contains concluding remarks.

2 Basic formulation

In this section we present general concepts and the basic formulations of Navier-Stokes sets. Six postulates are used to arrive at formulations of macroscopic equations for a mixture of N constituents:

1. The sum of all volume fractions ϕ_i equals one; vacuum regions do not occur.
2. The amount of mass, momentum, angular momentum and total energy are globally conserved in arbitrarily small domains without external influences.
3. The total entropy of the mixture increases in the absence of external influences; the second law of thermodynamics holds.
4. Locally, there is no change of variables for any component if all the velocity, temperatures and pressure variables, u_i , T_i and p_i , are in equilibrium and the variables are not subject to an external force or heat source. Equilibrium at a given location x occurs if

$$u_1 = u_2 = \dots = u_N, \tag{1}$$

$$T_1 = T_2 = \dots = T_N, \tag{2}$$

$$p_1 = p_2 = \dots = p_N, \tag{3}$$

are constant in a neighbourhood of x . [In the macroscopic view unequal volume fractions ϕ_i and unequal material densities ρ_i should be allowed in a state of equilibrium [24]. The definition of equilibrium changes if pressure jumps between components are included (section 8)].

5. The equations for component i reduce to the standard Navier-Stokes equations at location x in case $\phi_i = 1$ in a neighbourhood of x .
6. The equations are Galilean invariant.

The conventional approach to obtain equations for multiphase flows is to introduce linear volume or ensemble averages of physical quantities, such as the phase velocities (e.g. [10]). The approach is complicated due to closure problems arising from averaging across time-dependent interfaces and from

the averaging of nonlinear terms. For more than two components the geometrical problems become even more complicated. Therefore, a different strategy is pursued in the present paper; the second law of thermodynamics and other basic principles are postulated and then equations satisfying these principles are derived by mathematical reasoning.

However, before we will continue with this approach, the meaning of the entropy postulate is explained in terms of the conventional averaging approach. For this purpose we consider the dissipation of kinetic energy, which is the dominant nonlinear contribution to the growth of entropy, in most cases. The entropy law and dissipation inequality of total kinetic energy are formally equivalent in case of isothermal multicomponent flow with incompressible material densities. The nonlinearity of kinetic energy implies that part of the kinetic energy will be in microscale fluctuations. We define a local volume average \bar{q} of a quantity q by by a spatial convolution filter operation $\int G_\Delta(x-y)q(y)dy$ with $\int G_\Delta(z)dz = 1$, where G_Δ is a function of top-hat or Gaussian shape, for example. The filter removes the scales smaller than the filterwidth Δ . In particulate flows Δ should be larger than a few times the particle diameter at least. This averaging operator conserves integrals [29] which means $\int \bar{q} = \int q$. We also define the microscopic functions $X_i(x)$, where $X_i(x)$ equals 1 if component i is present precisely at x and 0 otherwise [10, 11]. The macroscopic volume fractions are subsequently defined by $\phi_i = \overline{X_i}$, the filtered velocities \hat{u}_i are defined by $\hat{u}_i = \overline{X_i u_i} / \phi_i$ and conservation of mass is expressed by

$$\frac{\partial \phi_i}{\partial t} + \nabla \phi_i \hat{u}_i = 0 \quad (4)$$

for constant material densities.

The total kinetic energy in all scales is denoted by K_1 for constant ρ_i and is rewritten as:

$$\begin{aligned} \int \sum_i \frac{1}{2} \rho_i X_i u_i \cdot u_i dV &= \int \sum_i \frac{1}{2} \overline{\rho_i X_i u_i \cdot u_i} dV \\ &= \int \sum_i \frac{1}{2} \rho_i \phi_i \hat{u}_i \cdot \hat{u}_i dV + K_4, \end{aligned} \quad (5)$$

where the three integrals define K_1 , K_2 and K_3 , respectively. The integration domain of these integrals, V , represents the entire flow domain. In the absence of external influences, The physical total kinetic energy K_1 equals K_2 , because the local volume average, denoted by the 'bar', is a conservative linear operator [29]. K_2 is decomposed in $K_3 + K_4$, where K_3 represents the kinetic energy in scales larger than Δ and $K_4 = K_2 - K_3$ represents the kinetic energy in scales smaller than Δ . In fact K_4 is the integral of the sum of half the traces of the Reynolds stress tensors.

The second law of thermodynamics prescribes that in a closed system (no external forces) K_1 decreases as a function of time. By analogy to the theory of the Reynolds stress tensor in single-phase flows, we assume that the scales smaller than Δ draw kinetic energy from the scales larger than Δ . As this transfer of energy is global, it does not necessarily exclude local backscatter of energy from small to large scales (see below). Due to the energy transfer from large to small scales the decrease of K_1 implies a decrease of K_3 , or in terms of entropy: the global increase of the physical entropy implies the increase of entropy in the averaged scales.

The heuristic arguments above indicate that the variables in the present continuum approach may be interpreted as local volume averages of the microscopic physical velocities, temperatures, pressures and densities. In fact, a positive dissipation axiom of macroscopic variables has been proven to lead to useful macroscopic equations for a wide range of complex physical phenomena, such as the description of elasticity and the behavior of polymers [20]. It is therefore interesting to adopt such an axiom as a starting point for the complex task of modeling multicomponent flows and investigate which models emerge. As we mentioned before, closed models for multicomponent flows that satisfy the second law existed for $N = 2$ only. In the present paper models that satisfy the second law are constructed for arbitrary N .

The theory, naturally evolving from the postulates above starts to formulate Navier-Stokes equations for each component i ,

$$\frac{\partial \rho_i \phi_i}{\partial t} + \nabla \cdot \rho_i \phi_i u_i = 0, \quad (6)$$

$$\frac{\partial \rho_i \phi_i u_i}{\partial t} + \nabla \cdot \rho_i \phi_i u_i u_i^T + \nabla \phi_i p_i + \nabla \cdot \phi_i \sigma_i = M_i, \quad (7)$$

$$\frac{\partial \rho_i \phi_i \tilde{e}_i}{\partial t} + \nabla \cdot \phi_i (\rho_i \tilde{e}_i + p_i) u_i - \nabla \cdot u_i \cdot \phi_i \sigma_i + \nabla \cdot \phi_i q_i = E_i, \quad (8)$$

where ρ_i , ϕ_i , u_i , p_i refer to the material densities, volume fractions, velocity vectors and pressures, respectively. Each total energy \tilde{e}_i is the sum of internal energy e_i and $\frac{1}{2}u_i \cdot u_i$. The viscous forces and heat-transfer acting inside component i are denoted by the viscous stress tensor σ_i and viscous heat flux q_i . The right-hand sides of the equations represent the interactions between components; M_i denotes the momentum and E_i the energy received from the other components. To keep the present formulation relatively concise, incorporation of body forces and nonzero right-hand sides of the continuity equations, corresponding to mass exchange, will be postponed until section 8.

For further reasons of transparency of the present equations, we did not write the macroscopic variables with an overbar. Reynolds stresses, which occur from the viewpoint of (mass-weighted) averaging and represent the effect of scales smaller than Δ , are not explicitly visible in the equations, but their effect is incorporated. The standard Boussinesq hypothesis is adopted, which means that our viscosity represents the sum of the molecular viscosity and an eddy-viscosity (see [29, 30] for a discussion of suitable eddy-viscosities). The thermal diffusivity is interpreted in a similar manner. More advanced models may be chosen instead of simple eddy-viscosity models, for example mixed models or adjoint filtered models [29]. These models are able to predict local backscatter of energy from small to large scales, while remaining globally dissipative. A positive global dissipation is essential in order to get stable large-eddy simulations of single-phase flows (e.g. [31]). Robustness of numerical computations is a practical reason to impose the global entropy law, also on equations for multiphase mixtures.

The equations (6-8) are local conservation laws for mass, momentum and total energy, respectively. The left-hand sides of reduce to the standard Navier-Stokes equations if the volume fraction ϕ_i equals one. The occurrence of ϕ_i reflects that the mass fluxes, momentum fluxes and energy fluxes through an infinitesimal volume face are multiplied with the local concentration of the component.

Postulates 1 and 2 are satisfied if we impose the following additional equations [28, 21, 6, 22]:

$$\sum_i \phi_i = 1, \quad (9)$$

$$\sum_i M_i = 0, \quad (10)$$

$$\sum_i \int E_i dV = 0, \quad (11)$$

where the last constraint holds in case of appropriate boundary conditions, e.g. zero flux of energy through the boundaries of the flow domain. Equation (11) is less severe than the conventional multiphase energy constraint, which omits the integral (see the references just mentioned). There does not seem to be a physical reason to enforce strict locality on the total energy exchange terms. However, we do impose a local constraint on the momentum exchanges. This is based on the third law of Newton, which leads to a pointwise balance of interaction forces, actually in a pairwise manner,

$$M_i = \sum_j M_{ij}, \quad M_{ij} = -M_{ji}, \quad (12)$$

where M_{ij} represents the force exerted by component j upon component i . We note that definition (12) implies equation (10). Equation (10) is not suitable for the modeling of surface tension. However, surface tension effects can be taken account by including pressure jumps and additional source terms in the momentum equation. This will be discussed in section 8.

The conservation of angular momentum requires a symmetric tensor σ_i , for which we assume the Newtonian form, $2\mu_i S u_i$, where μ_i represents the viscosity of phase i . The symbol S is the tensor valued strain operator, defined by

$$S w = \frac{1}{2} \nabla w + \frac{1}{2} (\nabla w)^T - \frac{1}{3} (\nabla \cdot w) I, \quad (13)$$

where w is a three-dimensional velocity vector. To simplify the equations a bulk-viscosity of $-\frac{2}{3}\mu_i$ is assumed. This assumption is not essential; increase of entropy can also be proven for larger bulk-viscosities. The viscous heat flux q_i is given by the Fourier law, $-\kappa_i \nabla T_i$, where κ is the heat-conductivity and T_i the temperature of component i . We also assume that each material density ρ_i is related to p_i and T_i , through thermodynamic equations of state. If component i is incompressible, the equation of state can be replaced with the assumption of constant ρ_i .

To find which possible structural forms of the interaction terms M_i and E_i satisfy the postulates, we start to consider the second law of thermodynamics. The entropy η_i of a component is defined by the Gibbs identity (see [3, 26, 6, 22]),

$$T_i D_i \eta_i = D_i e_i - \frac{p_i}{\rho_i^2} D_i \rho_i, \quad (14)$$

where

$$D_i = \frac{\partial}{\partial t} + u_i \cdot \nabla \quad (15)$$

denotes the material derivative with respect to u_i .

The second law of thermodynamics holds if the spatial integral of the weighted sum of all individual entropies increases. For appropriate boundary conditions this means (Appendix 1):

$$\begin{aligned} & \sum_i \int \frac{\partial \rho_i \phi_i \eta_i}{\partial t} dV = \\ & \sum_i \int \left(\frac{\phi_i \mu_i}{T_i} |Su_i|^2 + \frac{\phi_i \kappa_i}{T_i^2} \|\nabla T_i\|^2 \right) dV + \\ & + \sum_i \int \frac{1}{T_i} (p_i D_i \phi_i - u_i \cdot M_i + E_i) dV \geq 0, \end{aligned} \quad (16)$$

where $|Su_i|^2 = 2Su_i : Su_i$ and $\|\nabla T_i\|^2 = \nabla T_i \cdot \nabla T_i$. The first and second term at the right-hand side are analogous to the standard single-phase entropy production terms caused by viscosity and temperature. The last integral in equation (16) represents the contribution of the interaction terms to the entropy production. It is evident that a possible violation of the entropy law arises from this integral.

It is convenient to split the interaction terms into reversible and irreversible parts. For the following physical clarification we assume that there is no external force; the entire system is closed. Then the irreversible terms represent the effects of a deviation from the state of equilibrium. According to the definition of the state of equilibrium (postulate 4), nonequilibrium implies nonzero differences between velocities, temperatures or pressures. Therefore, appropriate models for the irreversible interaction terms are relaxation laws which irreversibly drive the flow towards equilibrium. The relaxation laws will be expressed in the differences just mentioned. Additional terms are needed to ensure that the flow remains in rest if it is in a state of equilibrium (postulate 4). Since these terms may be nonzero, even if the flow is in equilibrium, they are not allowed to alter the entropy. We call these terms the reversible parts of the interaction terms. The volume fraction does not occur in the definition of the state of equilibrium, which implies that the reversible parts can depend on the differences between or gradients of volume fractions. Their dependence on gradients of volume fractions is the reason to label these terms "nozzling terms".

The reversible parts are denoted by a single prime and will be specified in section 3, whereas the irreversible parts, denoted by double primes, will be specified in sections 4 and 5. The split reads:

$$M_i = M_i' + M_i'', \quad (17)$$

$$E_i = E_i' + E_i'', \quad (18)$$

where both M_i' and M_i'' satisfy equation (10) and both E_i' and E_i'' satisfy equation (11).

The last integral in (16) shows that appropriate expressions of $D_i \phi_i$ in terms of pressure differences may lead to quadratic forms of pressure differences, which always increase the entropy. This supports the historical choice (reviewed in the Introduction) to supplement two-pressure two-phase systems with with an equation that relates the change in volume fraction to the pressure difference. The sets B and C in this paper employ equations for the volume fraction of the form:

$$D_i \phi_i = F_i' + F_i'', \quad (19)$$

$$\sum_i F_i' = Y, \quad \sum_i F_i'' = 0, \quad (20)$$

$$Y = \sum_j (u_j \cdot \nabla \phi_j). \quad (21)$$

The variable Y is needed to ensure that the sum of volume fractions remains one. It implies that the expressions above define $N - 1$ independent evolution equations for the volume fractions.

Sets B and C apply the equations (19) and the continuity equations (6) simultaneously. It is instructive to consider the incompressible continuity equations (ρ_i is constant), which read

$$\frac{\partial \phi_i}{\partial t} + \nabla \cdot \phi_i u_i = 0, \quad (22)$$

and constraint (9) implies a continuity equation for the mixture,

$$\sum_i \nabla \cdot \phi_i u_i = 0. \quad (23)$$

Like incompressible single-component flows, incompressible multicomponent flows do not require equations of state. In the next section, set A will appear to have a single pressure P and a Poisson equation for P can be derived from condition (23). The solutions of the incompressible equations of sets B and C (multiple pressures) also satisfy (22), and combining (19) and (22) implies the additional equations

$$F'_i + F''_i = -\phi_i \nabla \cdot u_i. \quad (24)$$

F''_i will be expressed in pressure differences, such that (24) can be used to obtain the extra $N - 1$ pressures and (22) to prescribe the incompressible volume fractions. The precise definition of F''_i will be specified in section 5, where the physical meaning of (24) and thereby the meaning of (19) will be clarified.

3 Interaction terms: Nozzling

In this section we will present the interaction terms M'_i , E'_i and F'_i for three Navier-Stokes sets. Expression (16) shows that these interactions do not contribute to entropy if

$$E'_i = -p_i D_i \phi_i + u_i \cdot M'_i, \quad (25)$$

whereas constraints (9-11) are satisfied if

$$\sum_i D_i \phi_i = \sum_i u_i \cdot \nabla \phi_i, \quad (26)$$

$$\sum_i M'_i = 0, \quad (27)$$

$$\sum_i (-p_i D_i \phi_i + u_i \cdot M'_i) = 0. \quad (28)$$

Equation (26) is equivalent to zero $\sum \partial \phi_i / \partial t$, which follows from equation (9).

To find suitable solutions for the equations (26-28), we start from the definition of equilibrium (postulate 4). If M'_i was zero then a nonuniform volume fraction in a constant pressure field would generate a change of momentum and consequently cause inequilibrium [24]. This undesirable feature can be prevented if so-called nozzling terms are subtracted from $\nabla \phi_i p_i$. More specifically, the pressure terms minus M'_i should be independent of gradients of volume fractions in case of equilibrium. A natural way to achieve this is to require that the effective pressure term equals $\phi_i \nabla p_i$ in case all pressures are the same. Therefore, we require that the nozzling terms (M'_i) are equal to $p_i \nabla \phi_i$ in case of N equal pressures. Two straightforward choices to satisfy this requirement are:

$$M'_i = P \nabla \phi_i, \quad (29)$$

and

$$M'_i = p_i \nabla \phi_i - Z, \quad (30)$$

$$Z = \frac{1}{N} \sum_j p_j \nabla \phi_j, \quad (31)$$

where P represents an unknown pressure in the first case and Z is required to achieve $\sum M'_i = 0$ in the second case.

Once M'_i is known, substitution into equations (26-28) allows us to find solutions for $D_i \phi_i$, and, finally, E'_i can be calculated from equation (25). Next, we will show how three Navier-Stokes sets (A-C) emerge. The reversible parts of the resulting interaction terms have been summarized in Table 1. The trivial solution, zero $D_i \phi$, M'_i and E'_i , which, after the restrictive assumption of constant volume fraction, satisfies postulate 4 and equations (26-28) is not included in Table 1.

Set	M'_i	E'_i	F'_i	M'_{ij}
A	$P\nabla\phi_i$	$-P\partial\phi_i/\partial t$	$-$	$P(\nabla\phi_i - \nabla\phi_j)/N$
B	$p_N\nabla\phi_i$	$p_N u_i \nabla\phi_i$	0	0 ($j < N$) $p_N \nabla\phi_i$ ($j = N$)
$i = N$	$p_N \nabla\phi_N$	$p_N(u_N \nabla\phi_N - Y)$	Y	$-p_N \nabla\phi_j$ ($j < N$) 0 ($j = N$)
C	$p_i \nabla\phi_i - Z$	$p_i U \nabla\phi_i - u_i Z$	$(u_i - U) \nabla\phi_i$	$(p_i \nabla\phi_i - p_j \nabla\phi_j)/N$

Table 1: Nozzling interaction terms for three Navier-Stokes sets $A-C$, where $P = \sum p_i/N$, $U = \sum u_i/N$, $Y = \sum u_i \nabla\phi_i$ and $Z = \sum p_i \nabla\phi_i/N$. Set A is the only set with a single pressure. The momentum exchange M'_i equals $\sum_j M'_{ij}$, which relies on constraint (9), which implies $\sum \nabla\phi_j = 0$.

Substituting the first nozzling definition (29) into equation (28) yields

$$\sum_i \left(-p_i \left(\frac{\partial\phi_i}{\partial t} + u_i \cdot \nabla\phi_i \right) + u_i \cdot P \nabla\phi_i \right) = 0. \quad (32)$$

This is equivalent to

$$\sum_i (P - p_i) D_i \phi_i = 0, \quad (33)$$

since the sum of all $\partial\phi_i/\partial t$ equals zero. The most straightforward way to satisfy (33) is to assume a single pressure ($p_i = P$). Equation (25) implies

$$E'_i = -P \frac{\partial\phi_i}{\partial t}, \quad (34)$$

which leads to set A , the only set with a single pressure. The assumption of one pressure is a traditional choice in the field of multicomponent flows. Although P can be interpreted as some average of all pressures, it is not necessarily the actual pressure on the interfaces between components. The use of only one pressure is less restricting than it seems, as the influence of pressure differences can be represented by additional interaction forces [24]. Another option is to include multiple pressures explicitly, which is the case for the following two sets, B and C .

Set B follows from the notion that equation (33) can also be satisfied by $F'_i = 0$ for each i with $p_i \neq P$. As we do not yet incorporate irreversibility, $F'_i = 0$ implies $D_i \phi_i = 0$, according to definition (19). Condition (20) implies that $F'_i = 0$ is allowed for $N - 1$ components only. Then equation (33) implies that P must be equal to the pressure of the remaining component, say $P = P_N$. Equations (20) and (25) produce

$$F'_N = Y, \quad (35)$$

$$E'_i = p_N u_i \cdot \nabla\phi_i \quad (i < N), \quad (36)$$

$$E'_N = p_N (u_N \cdot \nabla\phi_N - Y). \quad (37)$$

The nozzling terms in set B are equivalent to those proposed by Baer-Nunziato [3] in case $N = 2$.

The third set (C) is based on the second definition of nozzling, equation (30). It does not assume a single pressure P , but a single convection velocity U for the equations of volume fractions, which implies

$$F'_i = (u_i - U) \cdot \nabla\phi_i. \quad (38)$$

Condition (28) reduces to

$$\sum_i p_i U \cdot \nabla\phi_i = \sum_i u_i \cdot Z, \quad (39)$$

which can only be satisfied by $U = \sum u_j/N$, which follows from substituting the definitions of U and Z and subsequent interchange of the summations on the right-hand side of equation (39). The reversible energy exchange terms in set C result from (25),

$$E'_i = p_i U \cdot \nabla \phi_i - u_i \cdot Z. \quad (40)$$

It appears that condition (33) also admits $P = \sum p_i/N$. For $N = 2$, set C is recovered with nozzling terms equivalent to those proposed by Ransom and Hicks [26]. For larger N , the choice $P = \sum p_i/N$ leads to a set with $F'_i = Y/N$. Analysis of the corresponding volume fraction equations shows that the set is ill-posed and thus inferior to set C .

All reversible (or nozzling) interaction terms in the Navier-Stokes sets have been collected in Table 1. The nozzling terms always allow a decomposition in pairwise terms, for example $M'_i = \sum_j M'_{ij}$ with $M'_{ij} = -M'_{ji}$, consistent with Newton's third law. Explicit forms of M'_{ij} for the sets $A - C$ are presented in Table 1. After the substitution of $p_i = P$, the M'_{ij} of set C reduces to M'_{ij} of set A . Theoretically, the pairwise decomposition is possible for E'_i and F'_i as well; there are $\frac{1}{2}N(N-1)$ pairwise unknowns, while they have to satisfy than $N-1$ independent equations only.

Cases A and C have symmetric nozzling terms, i.e. the mathematical form of these terms is the same for each component i . The terms are asymmetric in Set B , with the exception of M'_i , which has the same structure for each i . Including asymmetry in M'_i as well, more asymmetric sets, can be found. For example, suppose that M'_i equals $p_i \nabla \phi_i$ for $i < N$ and $p_N \nabla \phi_N - NZ$ for $i = N$. Then equations (25) and (38) with U replaced by u_N yield an asymmetric set that convects each volume fraction with u_N .

Set B is asymmetric in the sense that one particular component is governed by structurally different equations than the other components. At first sight, set B is suitable for the case in which one continuous component (number N) carries $N-1$ species of suspended solid particles. The volume fraction equations of the $N-1$ dispersed components all convect with their own velocity, which is consistent with the solid surface of the particles. The alternative asymmetric example mentioned at the end of the previous paragraph may be suitable to $N-1$ types of compressible bubbles dispersed in an incompressible fluid (component N).

However, also the sets with symmetric nozzling terms (A and C) allow asymmetry in the sense that each component has its own thermodynamic equation of state, viscosity and heat-conductivity. Also specific interactions between two components i and j , detailed in the next section, can be chosen asymmetric. For example, if component i is dispersed and component j is continuous, the drag force exerted by these components on each other may depend on the viscosity of component j only. Complications of this choice arise in cases where two components can be both dispersed and continuous. This is the case for a gas-liquid flow in which the liquid contains gas bubbles and the gas carries liquid drops. Then both viscosities effectively influence the drag; a strictly symmetric formulation is possibly the most logical choice.

It is interesting that if we subtract the nozzling term M'_i from the pressure term $\nabla \phi_i p_i$, a form of the pressure term derived from the conventional averaging approach results, for example $\phi_i \nabla P$ in case A . In contrast to some averaging theories, the present viscous terms in the momentum equations do not have nozzling corrections in M_i , because $\nabla \cdot \phi \sigma_i$ satisfies postulates 3 and 4 by itself, while $\nabla \phi_i p_i$ without nozzling would violate postulates 3 and 4. We stress that the volume averaging method is useful to interpret the present equations, but that it is not the starting point of the present derivation. We have chosen to start from the six postulates formulated above rather than exerting conventional averaging methods as far as possible. The present postulates are consistent with the present forms of pressure and viscous terms.

Perhaps somewhat unexpected, the harmonic average $U = \sum u_j/N$ occurs in the nozzling terms of set C , and not the weighted average

$$U_m = \left(\sum_j \rho_j \phi_j u_j \right) / \left(\sum_j \rho_j \phi_j \right). \quad (41)$$

Rewriting of the volume fraction equations of set C shows that each ϕ_i is convected with U . We therefore interpret U as the average velocity of the interfaces at a certain location. Consider a two-phase flow with one stagnant phase (on average) and one moving phase (on average). In this case the velocity of the interface is unlikely to be proportional to the volume fraction of the moving phase. We therefore conclude that the average velocity of the interface is probably better approximated by U than by U_m .

On the other hand, the volume average velocity of the mixture (not to be confused with the velocity of the interface) should depend on the volume fractions and material densities. The weighted average

(41) is an appropriate definition for the mixture velocity, including the mixture modeled by set C . This is deduced from the summation of the Navier-Stokes equations (6-8) over i . In fact the mixture satisfies a single system of Navier-Stokes equations (Powers, 2004), with complicated unknown constitutive equations. One classic modeling strategy, different from the present approach, is to prescribe the constitutive equations of the mixture, without modeling the evolution equations for the velocities of the separate components. A famous example of such a constitutive equation is the Einstein viscosity [12, 11], an expression for the effective viscosity μ_{eff} of a dilute two-phase flow with small particles, $\mu_{eff} = (1 + \frac{5}{2}\phi_{part})\mu_{fluid}$. It follows from an association between the physical dissipation of the suspension and the dissipation of an imaginary 'effective' fluid. The increase of the effective viscosity with volume fraction sheds another light on the present approach and its postulate of the second law of thermodynamics. Imposing the second law leads to a formula for the total dissipation of the mixture (section 5). The parameters of the following interaction laws can possibly be validated using the classic results of effective viscosity.

4 The generalized drag law

In this section we specify the irreversible momentum exchange terms M_i'' , defined by $\sum_j M_{ij}''$ with $M_{ij}'' = -M_{ji}''$. According to the explanation in section 2, the irreversible exchange M_{ij}'' depends on the velocity difference $u_i - u_j$. This immediately guarantees the Galilean invariance of M_{ij}'' .

We turn to an operatorial definition and write,

$$M_{ij}'' = -A_{ij}(u_i - u_j). \quad (42)$$

The dissipation inequality of kinetic energy will be satisfied if each A_{ij} is positive definite,

$$\int w \cdot A_{ij} w \, dV \geq 0, \quad w \neq 0. \quad (43)$$

Section 5 will clarify how the positive definiteness of A_{ij} is useful to prove the entropy law.

To define a positive definite A_{ij} , we consider the standard algebraic drag law supplemented with suitable second- and third-order derivatives:

$$\begin{aligned} A_{ij}w &= a_{ij1}w - \nabla \cdot (a_{ij2}Sw) \\ &+ \nabla \cdot ((a_{ij3}\nabla \cdot Sw) w^T) - \frac{1}{2}w\nabla \cdot (a_{ij3}\nabla \cdot Sw), \end{aligned} \quad (44)$$

where the coefficients may be functions of space and time and should satisfy $a_{ijk} = a_{jik} \geq 0$ for $k \leq 2$ and $a_{ij3} = -a_{ji3}$. The term with a_{ij2} is a linear second-order extension, whereas the last line represents a nonlinear third-order extension. For the coefficients a_{ijk} we propose the proportionality relation,

$$a_{ijk} \sim \phi_i \phi_j. \quad (45)$$

The incorporation of the volume fractions ensures postulate 5.

The high-order extensions to the drag law may be interpreted as effects of the subcontinuum scales on the interaction forces. These scales do not explicitly occur in continuum models (see section 2), but their effect on the exchange of momentum is included through second-order sink terms and third-order redistribution terms of the kinetic energy in macroscopic scales. Analysis of the nonlinear third-order term will show that simultaneous inclusion of the second-order terms can be useful. Rules for the relation between coefficients a_{ij2} and a_{ij3} will be given for the one-dimensional case (section 7).

Second-order terms in the operator A_{ij} are physically justified because of the existence of the so-called Faxen forces. The first term in the operator A_{ij} resembles the standard Stokes drag law, which has extensively been investigated for a single particle in a uniform flow. For a large amount of interacting particles in a surrounding flow, the assumptions for the Stokes law are not valid anymore. A classic correction term to include nonuniform effects of the flow on the motion of a single particle is the Faxen force, which is proportional to the Laplacian of the flow field at the particle location. The ratio between the Faxen and Stokes term is of the order $(d/l)^2$, where d is the particle diameter and l is a characteristic fluid length-scale. Due to the occurrence of the Laplacian operator in the Faxen force, l will be the characteristic length-scale of the fluid dissipation, that is the Kolmogorov length-scale, implying that the Faxen force may be as large as the Stokes drag, provided the particles and Kolmogorov wavenumber are sufficiently large.

Whereas the effect of the second-order extension is dissipative, the effect of the third-order terms is dispersive. This dispersion means that the travelling speed of Fourier wavenumber k of the velocity

difference w is decreased with $a_{ij3}w|k|^2$. In one dimension there is a similarity with the Korteweg-de Vries equation, which is Burger's equation plus a third-order derivative. The former equation has been proven to be useful in the description of many physical phenomena [8], including the behavior of pressure in gas-liquid columns, related to the motion of interfaces in two-phase flows [32, 5]. The equation regularizes wave discontinuities and admits soliton solutions.

The operator A_{ij} is positive definite indeed:

$$\int w \cdot A_{ij} w dV = \int \left[a_{ij1} \|w\|^2 + \frac{1}{2} a_{ij2} |Sw|^2 + \nabla \cdot (a_{ij2} w \cdot Sw) + \nabla \cdot \left(\frac{1}{2} \|w\|^2 a_{ij3} \nabla \cdot Sw \right) \right] dV \geq 0, \quad (46)$$

since the integral of the two divergence terms cancels for appropriate boundary conditions. The equality can be proven by substituting A_{ij} in the left-hand side and subsequently writing out left- and right-hand side. Use of index notation is convenient. It is evident now that the algebraic and second-order term dissipate kinetic energy, while the third-order terms only redistribute the kinetic energy.

In section 6 we will show that a third-order extension is more effective in stabilizing short waves than the second-order term, but quite surprisingly, more effective than a fourth-order term as well. Fourth- and fifth-order contributions to a positive definite A_{ij} are obtained if the Laplacian operator ∇^2 is put in front of $\nabla \cdot S$, whereas an example of a contribution with first-order derivatives is found by omitting $\nabla \cdot S$ in the third-order terms. Other positive definite operators are obtained if Sw is replaced with ∇w or $I\nabla \cdot w$.

It is remarked that the operator A_{ij} remains positive definite if a term proportional to

$$(u_i - u_j) \times (\nabla \times (u_i + u_j)), \quad (47)$$

is added to $A_{ij}(u_i - u_j)$. The additional term represents the so-called lift plus rotational force [11, 9, 24]. It does not affect positive definiteness, because its innerproduct with $(u_i - u_j)$ vanishes.

The added (or virtual) mass force (see the references just mentioned) is the force exerted on an accelerating particle due to the acceleration of surrounding fluid. A formal proof that this force satisfies the entropy inequality for all flows seems a hard task, which will not be undertaken in the present study. Instead, we will verify the inequality for simple flows including virtual mass effects. Consider a particle, represented by component 1, in a surrounding fluid, represented by component 2. Addition of the standard expression of virtual mass to the Stokes drag force in dilute flow ($\phi_1 \rightarrow 0$) leads to

$$M''_{12} = \frac{\phi_1 \rho_1}{\tau_p} (u_2 - u_1) + \frac{1}{2} \phi_1 \rho_2 (D_2 u_2 - D_1 u_1), \quad (48)$$

where τ_p is the Stokes response time of the particle. Although the second term may produce kinetic energy, the entropy law is satisfied for this flow if in the absence of external forces the production by the second term is not larger than the dissipation by the first term, *i.e.*

$$(u_1 - u_2) \cdot M''_{12} \leq 0. \quad (49)$$

This condition is satisfied in the case of a uniform surrounding flow at least. Dropping viscous terms at the left-hand side of (7), the momentum equations reduce to:

$$\rho_1 D_1 u_1 = M''_{12} / \phi_1, \quad (50)$$

$$\rho_2 D_2 u_2 = -M''_{12} / \phi_2. \quad (51)$$

Pressure gradients vanish, because the flow is uniform and there are no external forces. Since M''_{12} is proportional to ϕ_1 , we find $D_2 u_2 \ll D_1 u_1$ if $\phi_1 \rightarrow 0$ and thus $D_2 u_2$ can be ignored in equation (48). Thus $D_1 u_1$ can simply be solved after substitution of (48) into (50). Subsequently M''_{12} can be calculated and contribution (49) to the kinetic energy becomes

$$-\frac{\phi_1 \rho_1}{\tau_p} \left(1 - \frac{\rho_2}{2\rho_1 + \rho_2} \right) \|u_1 - u_2\|^2, \quad (52)$$

which is always negative. In this example the energy production reduces the amount of dissipation, but the total dissipation remains positive.

In practice added mass is often combined with drag, which is a viscous force. However, the first derivations of the added mass force assumed inviscid flow and neglected drag. If drag is omitted from

(48), the added mass force can be analyzed for nonuniform surrounding flows. In the following we present a perturbation analysis in terms of the small variable ϕ_1 to prove that the $O(\phi_1)$ term in the dissipation is zero, which means that, at leading order, the added mass effect is reversible in a nonuniform steady inviscid two-phase flow. The perturbation analysis assumes

$$u_i = \tilde{u}_i + O(\phi_1), \quad (53)$$

$$P = \tilde{P} + O(\phi_1), \quad (54)$$

where $P = p_1 = p_2$ (set A). Since the nature of the perturbation analysis requires that \tilde{u}_i and \tilde{P} do not depend on ϕ_1 , substitution of (53) in the continuity equations and ρ_i is constant leads to

$$\nabla \cdot \phi_1 \tilde{u}_1 = 0, \quad (55)$$

$$\nabla \cdot \tilde{u}_2 = 0. \quad (56)$$

Since $M''_{12} = O(\phi_1)$, the momentum equations provide

$$\rho_1 D_1 \tilde{u}_1 = -\nabla \tilde{P} + \frac{1}{2} \rho_2 (D_2 \tilde{u}_2 - D_1 \tilde{u}_1), \quad (57)$$

$$\rho_2 D_2 \tilde{u}_2 = -\nabla \tilde{P}, \quad (58)$$

which implies

$$D_1 \tilde{u}_1 = \tilde{u}_1 \cdot \nabla \tilde{u}_1 = -a \nabla P, \quad (59)$$

where $a = 3\rho_2 / (2\rho_1 + \rho_2)$ is constant. The similarity of the last two equations implies $\tilde{u}_1 = \sqrt{a} \tilde{u}_2$. Substitution of (53) into the dissipation $(u_1 - u_2) \cdot M''_{12}$ shows reversibility of the added mass force at leading-order:

$$\begin{aligned} \int (\tilde{u}_1 - \tilde{u}_2) \cdot \frac{1}{2} \rho_2 (D_2 \tilde{u}_2 - D_1 \tilde{u}_1) \phi_1 dV &= \\ \frac{1}{2} (1 - \sqrt{a}) (1 - a) \int \phi_1 \tilde{u}_1 \cdot \nabla \tilde{P} dV &= 0. \end{aligned} \quad (60)$$

The last equality follows from partial integration and equation (55). Consequently, although $u_1 - u_2$ and M''_{ij} are respectively $O(1)$ and $O(\phi_1)$, the integral of the dissipation $(u_1 - u_2) \cdot M''_{ij}$ is only $O(\phi_1^2)$ or less. In dilute flows ϕ_1 is less than 0.01.

The analysis above concerns two special cases. Thus it is only an indication and no general proof that after the inclusion of added mass the entropy law of macroscopic scales is still satisfied. If the total force violated the entropy law, perhaps due to added mass or any other nondissipative force, one could project the physical expression on a globally dissipative structure of the form of equation (46) with variable coefficients, using a least-square minimization. A similar procedure is applied in the successful dynamic subgrid model [13, 18]. Essentially, this model projects nondissipative expressions of the Reynolds subgrid stress on an eddy-viscosity. We finish the discussion of the added mass force with the remark that incorporation of this force into a model may lead to less accurate predictions if the surrounding flow is turbulent [4].

5 Interaction terms: Irreversibility

In addition to M''_{ij} defined in the previous section, we propose the following irreversible interaction terms:

$$F_i = \sum_j F''_{ij}, \quad E_i = \sum_j E''_{ij}, \quad (61)$$

$$F''_{ij} = a_{ij4} (p_i - p_j), \quad (62)$$

$$\begin{aligned} E''_{ij} &= -a_{ij1} u_{ij} \cdot (u_i - u_j) - a_{ij2} s_{ij} : S(u_i - u_j) \\ &\quad - p_{ij} F''_{ij} - a_{ij5} T_i T_j (T_i - T_j) \\ &\quad + \nabla \cdot u_i \cdot \left[a_{ij2} S(u_i - u_j) \right] \\ &\quad + \nabla \cdot u_i \cdot \left[\frac{1}{2} (u_i - u_j) a_{ij3} \nabla \cdot S(u_i - u_j) \right], \end{aligned} \quad (63)$$

where u_{ij} (p_{ij}) are weighted averages of the velocities (pressures) of phases i and j . The tensor s_{ij} is an average of the strain tensors s_i and s_j , where s_i is the notation for $S(u_i)$. The precise form of the

averages will be defined later on. The two coefficients a_{ij4} and a_{ij5} should be positive. Note that set A does not contain the terms with F''_{ij} .

The physical interpretation of the five terms in E''_{ij} is as follows. The work done by the first two terms of the generalized drag law causes an energy exchange, which is quantified by the first, second and fifth term in equation (63). The third term denotes the energy exchange due to the work done by the pressure when the volume fraction changes. The fourth term in equation (63) accounts for the heat exchange from component j to i due to macroscopic temperature differences. The last two terms are energy redistribution terms, already visible in derivation (46). They have the same form as the standard stress working term in the Navier-Stokes equation, the third term in equation (8). As a consequence they can be interpreted as augmentations to the standard appearance of σ_i in the energy equations. It is remarked that the last two terms in E''_{ij} have their origin in the kinetic energy equations. Therefore they do not appear in the equations of internal energy e_i , which is the total minus kinetic energy. This implies that generalized drag induces *locally* positive source-terms in the internal energy and entropy equations, as will be shown below. Thus, generalized drag law will never decrease the global minima of the internal energies, (or temperatures), which is intuitively consistent with physics.

The physical justification for F''_{ij} is less obvious. These terms represent another model for the closure problem in multiphase flows, which arises because there are $7N$ principal unknowns, ϕ_i , ρ_i , p_i , T_i and the three components of u_i . However there are only $6N + 1$ equations, directly related to physics: Equation (9), $5N$ equations represented by (6-8) and N equations of state. In set A the $N - 1$ missing equations are provided by the assumption that all pressures are equal. Sets B and C involve $N - 1$ independent additional equations for the volume fractions, which can also be interpreted as relations between pressures, due to the definition of F''_{ij} . To clarify this we consider two-component flows and set B with two incompressible densities (which replace the two equations of state). Then the pressure difference $p_1 - p_2$ is proportional to $D_1\phi_1$ and, as a consequence of the continuity equation, also proportional to $\nabla \cdot u_1$. In the case of dispersed flows, p_2 is the pressure of the surrounding flow, while p_1 becomes larger than p_2 if $\nabla \cdot u_1 < 0$. The model F''_{12} is consistent with physics, because $\nabla \cdot u_1 < 0$ corresponds to compression of the dispersed phase, which implies more interparticle collisions and therefore a higher dispersed pressure.

For simplicity, we have chosen for algebraic relations between F''_{ij} and $p_i - p_j$ and between E''_{ij} and $T_i - T_j$. However, irreversibility can also be proven if these relations are replaced by positive definite operators acting on scalar functions. Boundary terms occurring in the integral of the definition of positive definiteness have to be treated with care. They will induce additional divergence or other globally conserved terms in E''_{ij} .

In the definition of the averaged quantities, there is freedom in the partition of the entropy production to the components i and j which are involved in the specific interaction processes M''_{ij} or F''_{ij} [6]. Considering the momentum exchange process first, the entropy production caused by the irreversible interaction force between i and j is divided into a fraction $b_{uij} \geq 0$, attributed to component j and a fraction $b_{uji} \geq 0$, attributed to component i . For each pair $i \neq j$, we have

$$b_{uij} + b_{uji} = 1. \quad (64)$$

A symmetric partition of entropy is achieved if $b_{uij} = b_{uji} = \frac{1}{2}$, which might be a suitable choice, in general. However, for specific cases of multicomponent flows with one or more dispersed phases, asymmetric options can also be defended. Consider the example of one carrier fluid (component N) embedding $N - 1$ species of solid dispersed particles. Baer and Nunziato [3], Bdzil et al. [6] and Powers [22] propose to attribute all the entropy production caused by the drag to the entropy of the fluid, maybe because the drag deforms the fluid and not the solids. This corresponds to $b_{uiN} = 1$ and $b_{uNi} = 0$ in the present model. These authors further propose that the pressure differences increase only the entropy of the solids. In the present example of $N - 1$ species of solid particles, the $N - 1$ solid components also exert forces on each other. The interaction force between two solid components i and j is likely to increase the entropy of both components, which can be achieved with, with for example the symmetric choice, $b_{uij} = \frac{1}{2}$ for $i \neq N$ and $j \neq N$.

The definitions of the averages needed for the partitions are:

$$u_{ij} = b_{uij}u_i + b_{uji}u_j, \quad (65)$$

$$p_{ij} = b_{pij}p_i + b_{pji}p_j, \quad (66)$$

$$s_{ij} = b_{sij}s_i + b_{sji}s_j, \quad (67)$$

equal to u_{ji} , p_{ji} and s_{ji} , respectively. The entropy partition coefficients b_{pij} and b_{sij} satisfy constraints like (64).

The total energy is conserved; constraint (11) is satisfied by E_i'' , since

$$\int (E_{ij}'' + E_{ji}'') dV = 0, \quad (68)$$

provided there are no fluxes through the boundaries of V . It holds because $E_{ij}'' + E_{ji}''$ reduces to the two divergence terms in equation (46) for $w = u_i - u_j$. Divergence terms express a conservation property for arbitrarily small volumes. It is possible to formulate a general operatorial formulation that satisfies both irreversibility and a pointwise constraint $E_{ij}'' = -E_{ji}''$ (Appendix 1).

Next, it will be shown that the exchange laws introduced above always satisfy the entropy law. It is noted that M_i' , E_i' and F_i' do not alter the entropy, by construction. The total entropy increase follows from expression (16), in which we have to prove the positivity of the last integral. Details of the proof are found in Appendix 1. We just mention that one step in the derivation is the determination of the internal energy equation. As an example we give the local increase of the internal energy of component i due to the algebraic part of the drag-law:

$$a_{ij1}(u_i - u_{ij}) \cdot (u_i - u_j) = a_{ij1} b_{uji}(u_i - u_j) \cdot (u_i - u_j) \geq 0. \quad (69)$$

The equality can simply be verified by substitution of (65) into the left-hand side. It illustrates that b_{uji} is approximately the fraction of internal energy attributed to e_i due to the algebraic part of the drag force between i and j . The derivation above is similar for the scalar relaxation law dependent on $p_i - p_j$, and the tensor relaxation law dependent on $s_i - s_j$, which is equal to $S(u_i - u_j)$. The term with a_{ij3} does not occur in the internal energy equations.

At each location, the increase of entropy due to the interactions between all components turns out to be positive (Appendix 1):

$$\begin{aligned} & \frac{1}{2} \sum_i \sum_j \left(\frac{a_{ij1}}{T_{uij}} \|u_i - u_j\|^2 + \frac{a_{ij2}}{T_{sij}} \frac{1}{2} |S(u_i - u_j)|^2 \right) \\ & + \frac{a_{ij4}}{T_{pij}} (p_i - p_j)^2 + a_{ij5} (T_i - T_j)^2 \geq 0. \end{aligned} \quad (70)$$

Set A does not have the term with a_{ij4} . To shorten the notation, equation (70) employs an average temperature

$$T_{uij} = \frac{T_i T_j}{b_{uij} T_i + b_{uji} T_j}, \quad (71)$$

also equal to T_{uji} . The definitions of T_{pij} and T_{sij} are analogous; u_i is replaced by p_i or s_i , while b_{uij} is replaced by entropy partition coefficients b_{pij} or b_{sij} , respectively. These coefficients allow us to control the entropy production of each interaction process separately.

In the incompressible isothermal limit the entropy law is equivalent to the decay law of the total kinetic energy K . It is important that such a decay inequality exists for incompressible Navier-Stokes sets, since the decay inequality of kinetic energy is the corner stone of analytical theories for the Navier-Stokes equations of a single fluid. Incompressibility and the total conservation of mass further imply $\sum \nabla \cdot (\phi_i u_i) = 0$.

Like the coefficients in M_{ij}'' , we assume that the coefficients a_{ij4} and a_{ij5} are proportional to $\phi_i \phi_j$. The occurrence of ϕ_i in a_{ij4} causes each ϕ_i to remain between zero and one for finite $p_i - p_j$. This can be shown by solving the volume fraction equations along the appropriate Lagrangian path $(t, x(t))$. The integration yields

$$\phi_i(t) = \phi_i(0) \exp \left(\int r_i(t', x(t')) dt' \right) \geq 0, \quad (72)$$

where r_i represents the right-hand side of the volume fraction equation divided by ϕ_i . If each ϕ_i is positive then constraint (9) evidently implies $\phi_i \leq 1$.

The present equations for mass, momentum, entropy and volume fraction turn out to be Galilean invariant. This implies that the equations of internal energy are also Galilean invariant, which is easily recognized from the Gibbs identities. In the construction of the present Navier-Stokes sets, differences or derivatives of velocities have consistently been used for the purpose of Galilean invariance. Postulate 6 is therefore valid. Finally, Appendix 2 proves that under certain conditions of the coefficients in the interaction laws zero M_i , E_i and F_i imply that the flow is in the equilibrium state of postulate 4.

6 Characteristic velocities

In this section we will show that the sets B and C have real characteristic velocities for general N in the inviscid limit. Set A will be shown to have real characteristics for appropriate extensions of the drag law, involving spatial derivatives of at least second order and applied to velocity differences. A rigorous analytical proof will be given for incompressible two- and three-phase flows. The physical meaning of well-posedness can be understood in view of its connection with linear stability theory, which will be clarified in the next section. The analysis in this and the next section will be restricted to one dimension. Generalizations to more dimensions are straightforward in some cases, but more complicated in other cases. We remark that one-dimensional descriptions of multiphase flows are not only of theoretical, but also of practical interest for several applications.

The occurrence of complex characteristics is one of the most foundational problems in theory of multiphase flows, since it makes the equations ill-posed. With well-posedness of the linearized equations in the sense of Hadamard we mean that the equations linearized around constant fields have real characteristic velocities. They can therefore be solved as an initial boundary value problem, following the characteristic directions [11, 24]. The characteristic velocities are the roots of a generalized eigenvalue problem. The characteristic polynomial is the determinant of the matrix $R = \lambda\tilde{A} - \tilde{B}$, where \tilde{A} is the coefficient matrix of all first-order time derivatives and \tilde{B} of all first-order spatial derivatives. Algebraic terms do not influence the characteristics. Well-posedness of the equations requires that the characteristic velocities are real; complex eigenvalues would imply infinitely growing instabilities. Formally, well-posedness also requires the set of eigenvectors to be complete, which is definitely true if all eigenvalues are different.

In the construction of matrix R for set A , we combine the nozzling term with the pressure term at the left-hand side of equation (7). This leads to $\phi_i \nabla P$ and implies that derivatives of volume fractions vanish in the (linearized) momentum equations. Unfortunately, the matrix R cannot be written as a triangular block-matrix. This complicates the determination of the eigenvalues corresponding to set A .

First, we will turn to the more simple analysis of Navier-Stokes sets B and C , without irreversible terms. Unlike set A , these sets allow the generalized eigenvalue problem to be written as the determinant of a *triangular* block-matrix:

$$\begin{vmatrix} R_{00} & 0 & 0 & \dots & 0 \\ R_{01} & R_{11} & 0 & \dots & 0 \\ R_{02} & 0 & R_{22} & \dots & 0 \\ \vdots & \vdots & \vdots & & \vdots \\ R_{0N} & 0 & 0 & \dots & R_{NN} \end{vmatrix}. \quad (73)$$

All $\nabla \phi_i$ terms in the linearized continuity, momentum and energy equations occur in the matrices R_{01} to R_{0N} . These submatrices do not have any effect on the eigenvalues, since the block-structure is triangular. The roots of the determinant above simply result from the $N + 1$ decoupled eigenvalue problems given by the matrices on the diagonal, R_{ii} . For $i \geq 1$ the problem is equivalent to the standard Euler equations with the real characteristics u_i and $u_i \pm c_i$ where c_i is the speed of sound of phase i .

In more detail, the first column of blocks in (73) represent $N - 1$ independent volume fractions and each following column $i + 1$ of blocks represents the density, velocity and pressure of phase i . The first row of blocks is derived from $N - 1$ independent equations for ϕ_i , while row $i + 1$ represents the continuity, momentum and internal energy equation for phase i . The total dimension of the determinant equals $4N - 1$.

The matrix R_{00} is different for set B and C . For set B this submatrix is also triangular and its $N - 1$ eigenvalues are determined by the diagonal elements, u_1 to u_{N-1} . In case of set C the matrix R_{00} is diagonal and contains $N - 1$ times the element $\lambda - U$, resulting in the additional characteristic velocity U with multiplicity $N - 1$. In case of $N = 2$, the eigenvector spaces of sets B and C have been proven to be complete, with exception of a set of velocities with measure zero [26, 3, 6].

We now turn to the most complicated case, set A (single pressure), in which the eigenvalue problem is non-triangular. The canonical problem of ill-posed equations for multiphase flows concerns this set in case $N = 2$ and the two components are incompressible. There is a pair of complex characteristics if $u_1 \neq u_2$. The compressible case has the same problem, but the analysis becomes more complicated there. As reviewed in the Introduction several solutions have been presented in literature to obtain real characteristics for example to include viscosity for one of the phases [2, 11]. Another solution is the inclusion of surface tension [25]. The solutions proposed in literature can be interpreted as additions of higher-order derivatives to the equations [23], second-order in the case of viscosity and third-order in the case of surface tension.

Here an alternative approach is considered; the incorporation of high-order spatial derivatives in terms of velocity *differences* between the phases. It respects the fact that ill-posedness without regularization does not occur if the phase velocities are equal. Another advantage is that the regularization term can simply be interpreted as a modification of the standard drag law.

To prove real characteristics with the approach of derivatives of velocity differences, we chose a fixed m and add to the incompressible momentum equations terms m -th order derivatives. According to Appendix 3 the characteristic velocities are the roots of a polynomial of degree $N - 1$:

$$Q(\lambda) = \sum_i (\phi_i \prod_{j \neq i} (\lambda - u_j)) = 0 \quad (74)$$

The challenge is to prove realness of the roots of $Q(\lambda)$ for general N .

The characteristic velocity equals $\phi_2 u_1 + \phi_1 u_2$ in case $N = 2$. It is real and hereby the canonical incompressible problem for $N = 2$ has now been proven to be well-posed for an arbitrarily small extension of the drag law with m -th order derivatives and $m \geq 2$. It is remarked that the dimension of the generalized eigenvalue matrix R equals five if $m = 2$. The four 'missing' eigenvalues include two infinite characteristics related to the infinite speed of sound in incompressible flows. The other two eigenvalues do not occur because the system has become partially parabolic.

Surprisingly, the characteristic velocity for $N = 2$ is $\phi_2 u_1 + \phi_1 u_2$ and not $\phi_1 u_1 + \phi_2 u_2$. In the example of dilute two-phase flows, this means that the characteristic velocity is approximately the velocity of the dilute phase. Apparently, the partially hyperbolic character of the equations is related to the dilute phase in particular. Signals propagating with the velocity of the dilute phase are hardly affected by the diffusion induced by the second-order extension of the drag.

A rigorous proof of real characteristic velocities for $N = 3$ is found in Appendix 3. There is an indication that the polynomial has also real roots for $N \geq 4$. This indication is based on solutions obtained with MATLAB after substitutions of random values for the velocities and volume fractions in the coefficients of $Q(\lambda)$, with the constraints $\phi_i \geq 0$ and $\sum \phi_i = 1$. No counter example with complex roots was found. The challenge to find a general proof for $N \geq 4$ remains.

Numerical evidence suggests that the extended drag law also leads to well-posed equations in set A in case the phases are compressible. The compressible $N = 2$ case was investigated substituting arbitrary values for the velocities, densities, pressures and volume fraction. For each phase we assumed an ideal gas law, but the Mach numbers and ratios of specific heats were different for each phase. The system was based on two continuity, two momentum and two internal energy equations, while the dimension of the system was seven for $m = 2$. Then the MAPLE-package solved the generalized eigenvalue problem for several realizations. For each realization, all returned eigenvalues, five in total, were real.

7 Linear stability

This section analyzes the linear stability of a constant profile, focusing on the incompressible isothermal case in one dimension. We will consider set A in particular; but the linear stability of sets B and C is also shortly discussed. As argued in the Introduction, the linear instability of a limited range of wavelengths is related to the physical Kelvin-Helmholtz instability.

Arai [2] showed that, although inclusion of the standard viscous terms leads to well-posedness, all wavenumbers are amplified if the phases are incompressible. Compressibility was proven stabilize sufficiently short waves. Prosperetti and Jones [23] found that the incompressible two-fluid equations supplemented with fluid velocity derivatives of arbitrary even order are linearly unstable for $u_1 \neq u_2$. They also rewrote several models in terms of third-order derivatives, for example the proposal of surface tension by Ramshaw and Trapp [25], which stabilizes sufficiently high wavenumbers. Here we will concentrate on the effect of derivatives of velocity *differences*. It will be shown that derivatives of even order lead to finite linear growth-rates, in contrast to ill-posedness, which produces instabilities of infinite growth-rate for $k \rightarrow \infty$. It will also appear that, provided the even order is larger than four, the unstable growth-rate asymptotically goes to zero. The growth-rate can also be exactly zero for sufficiently short wavelengths, but then the order of derivatives should be odd.

The linear stability is investigated by assuming constant fields plus a perturbation of the form

$$\exp(i\omega t - ikx). \quad (75)$$

Here k is the real spatial wavenumber, ω is a complex number and $i^2 = -1$. The imaginary part of ω is the growth-rate corresponding to k . If it is positive, the perturbation wave is called unstable.

The issue of well-posedness, treated in the previous section, can be interpreted in the terms of linear stability of specific waves. More specifically, $w = \lambda_c k$ is a solution of the linear stability problem for each characteristic velocity λ_c and spatial wavenumber k . Ill-posedness of the equations (complex λ_c) implies that for each k the perturbation is unstable for plus or minus k .

For the incompressible equations of set A , supplied with the generalized drag law, the solutions for ω in the linear stability analysis are the roots of a determinant of dimension $2N$,

$$\begin{vmatrix} R_{11} & R_{12} & 0 \\ 0 & R_{22} & R_{23} \end{vmatrix}. \quad (76)$$

Appendix 4 specifies the matrices R_{ij} for $N = 3$.

In the following we analyze the case $N = 2$ in more detail. Then determinant (76) is of dimension four and after some algebra the eigenvalue problem expressed in $\lambda = w/k$ can be written as:

$$-a\lambda^2 + (b - q)\lambda + (q\hat{u} - c) = 0, \quad (77)$$

where $\hat{u} = \phi_2 u_1 + \phi_1 u_2$ is precisely the characteristic velocity from the previous section, while a , b and c are positive real coefficients, specified in Appendix 4. These coefficients are independent of the spatial wavenumber k . The coefficient q , proportional to k^{m-1} , represents the interaction force. The coefficient is complex for even and real for odd m , where m is the order of the derivative in the generalized drag law.

In case of a second-order drag extension ($m = 2$), there are two complex roots of equation (77). The reader is referred to Appendix 4 for more details. The real parts of the roots contain \hat{u} and represent velocities of travelling waves. One of the imaginary parts corresponds to a negative growth-rate proportional to $-k^2$, obviously reflecting the dissipative character of the second-order derivative of the velocity difference. The other imaginary part vanishes if $u_1 = u_2$. Otherwise, it is of indefinite sign and may therefore represent a potential instability. However, the unstable growth-rate would remain constant for $k \rightarrow \infty$, in contrast to the infinitely large growth-rate for ill-posed problems. In case of fourth-order derivatives the growth-rate of the potential instability converges to zero if $k \rightarrow \infty$.

However, not only fourth-order, also second-order derivatives may be able to produce an asymptotically zero maximum growth-rate for $k \rightarrow \infty$. For this purpose the coefficient of the drag derivative, d , should be allowed to approach infinity for $k \rightarrow \infty$. For example, assume $a_{ij2} = c_2 \rho_{ij} l^2 |\partial w / \partial x|$, where c_1 is a positive coefficient, ρ_{ij} is an average density and l is a length-scale, for example the length-scale of an averaging operator or the particle diameter in particulate flows. This expression is analogous to Prandtl's mixing length model or the three-dimensional Smagorinsky model frequently applied in large-eddy simulation. Here the model is applied to the velocity difference, w . It was remarked in the Introduction that for particle-fluid flows the physical instabilities may continue to the scale of a few times the particle diameter. This scale might be smaller than the smallest length-scale in a numerical simulation, an additional reason to use a nonlinear a_{ij2} , taking subgrid contributions to the drag-force into account.

In section 4 we also proposed a nonlinear third-order extension to the drag law. In one dimension and for constant a_{ij3} , the nonlinear terms in equation (44) reduce to

$$a_{ij3} \left(\frac{1}{2} w \frac{\partial^3 w}{\partial x^3} + \frac{\partial w}{\partial x} \frac{\partial^2 w}{\partial x^2} \right) \quad (78)$$

The second term of equation (78) vanishes in the present linearized problem, because it is the product of first and second-order derivatives. This expression shows that the linearized equations will contain a third-order derivative multiplied with w , the velocity difference of the constant fields. It is evident that the third-order term is not active if w equals zero ($u_i = u_j$). This is not a problem because instabilities can occur only in the case of unequal velocities. In case w is not zero, the third-order derivative entirely neutralizes all instabilities beyond a certain wavenumber (proven in Appendix 4). Apparently, dispersion of the velocity difference is quite effective to stabilize short waves in multicomponent flows. The success of the Korteweg-de Vries equation in predicting wave phenomena, could indicate that the present regularizing third-order term also leads to a more physical propagation of waves, in particular waves caused by velocity differences.

Although the second term in equation (78) does not play a role in the linear theory, a remark on this term added, because it contains a possibly negative diffusion coefficient $\partial w / \partial x$. A suitable combination of the second- and nonlinear third-order derivative terms, for example $a_{ij3} = c_3 \rho_{ij} l^2$ with $|c_3| < c_2$ guarantees a locally positive diffusion.

The linear stability results above are probably also true for larger N . We investigated the incompressible cases $N = 3$ and $N = 4$, by substituting arbitrary numerical values for the unperturbed state and assuming

$$q_{ij} = \phi_i \phi_j \sqrt{\rho_i \rho_j} q. \quad (79)$$

The determinants of dimensions $2N$ were calculated using MAPLE, where care had to be taken to control the machine precision errors. Then the roots of the determinant were determined for several values of q . The imaginary parts of the growth-rates were negative for sufficiently high real values of q , showing short wavelength stability for odd $m \geq 3$.

So far, we considered the linear stability of set A . Conclusions regarding the incompressible stability of sets B and C can be drawn by interpretation of the existing literature for $N = 2$. If ρ_i is constant, $D_i \phi_i = -\phi_i \nabla \cdot u_i$. In one dimension this implies that the pressure differences in set B are equivalent to the inclusion of standard viscous terms in $N - 1$ momentum equations. However, standard viscosity does not stabilize sufficiently short waves, unless the phases are compressible [2]. In the present terminology, standard viscosity means that R_{22} becomes a diagonal matrix with diagonal elements $\tilde{\lambda}_j - ik\mu_j$. Repetition of the analysis above for this case leads to a finite positive growth-rate for $k \rightarrow \infty$. Apparently, the inclusion of multiple pressures does not cause short-wave stability for the incompressible variant of set B . We mentioned before that the treatment of pressures in set C for $N = 2$ reduces to the Ransom and Hicks [26] approach, which allows short-wave instabilities in the incompressible case [23]. Note that we performed a *linear* stability analysis. Due to nonlinearity, linear instabilities may saturate after some time, in particular if the growth-rate is finite and the equations satisfy an entropy inequality, since that inequality implies an upperbound for the total kinetic energy in the incompressible case.

8 External forces, mass exchange and pressure jumps

In this section we address a few additional effects, such as body forces (for example gravity), mass exchange and pressure jumps. We propose a strictly irreversible relaxation form for the interfacial mass exchange. This relaxation term will be expressed in the Gibbs free energy [6],

$$z_i = e_i + \frac{p_i}{\rho_i} - \eta_i T_i. \quad (80)$$

The physical interpretation is as follows: component i gains mass at the expense of component j if component i has a larger Gibbs free energy than component j . The entropy increases during this exchange process. Examples of mass exchange are combustion or other chemical reactions in technological flows, or the evaporation of water and melting of ice in geophysical flows.

Considering body forces and mass exchange first, the following terms are added to the right-hand sides of the continuity, momentum and energy equations,

$$G_i = \sum_j G''_{ij}, \quad (81)$$

$$\tilde{M}_i = \rho_i \phi_i g_i + \sum_j \frac{1}{2} (u_i + u_j) G''_{ij}, \quad (82)$$

$$\tilde{E}_i = \rho_i \phi_i g_i \cdot u_i + \sum_j (e_{ij} + \frac{1}{2} u_i \cdot u_j + y_{ij}) G''_{ij}, \quad (83)$$

respectively. Here g_i represents the acceleration vector of the body force. The mass exchange between phases i and j is denoted with G''_{ij} , while

$$G''_{ij} = -a_{ij6} (z_i - z_j), \quad (84)$$

$$e_{ij} = e_i + e_j, \quad (85)$$

$$y_{ij} = b_{zij} (z_i - e_{ij}) + b_{zji} (z_j - e_{ij}), \quad (86)$$

A positive coefficient a_{ij6} proportional to $\phi_i \phi_j$ is assumed and b_{zij} denotes the entropy partition coefficient corresponding to the mass exchange process (similar to b_{uij} in section 5). Summation over i yields zero for the three interaction terms (81-83), because $G''_{ij} = -G''_{ji}$, $e_{ij} = e_{ji}$ and $y_{ij} = y_{ji}$.

As a consequence, the entropy contribution due to mass exchange equals:

$$\sum_i \int \frac{1}{T_i} (\tilde{E}_i - u_i \cdot \tilde{M}_i + (\frac{1}{2} u_i \cdot u_i - z_i) G_i) dV$$

$$\begin{aligned}
&= \sum_i \sum_j \int \frac{1}{T_i} (y_{ij} + e_i + e_j - z_i) G''_{ij} dV \\
&= \frac{1}{2} \sum_i \sum_j \frac{a_{ij6}}{T_{zij}} (z_i - z_j)^2 \geq 0.
\end{aligned} \tag{87}$$

To obtain the third line, the definitions of y_{ij} , G''_{ij} have been substituted into the expression on the second line, whereas T_{zij} is defined like (71). Increase of entropy can also be proven for a positive definite operatorial formulation of the mass exchange law.

It is remarkable that the present irreversible mass exchange law is expressed in a difference of variables of the exchanging phases. To the author's knowledge existing irreversible models of two-phase mass exchange can not be expressed as a relaxation term dependent on a thermodynamic variable difference [6, 22]. That such a irreversible form does exist improves the similarity between the modeling of mass exchange and the irreversible momentum and energy exchange processes, which were also based on positive definite relaxation operators.

Pressure jumps can be included through pressure differences. Below we present an example, suitable for sets B and C , inspired by the two-phase formulation by Baer and Nunziato [3], Bdzil [6] and Powers [22]. The pressure relaxation operators $C_{ij}(p_i - p_j)$ in the equations for the volume fractions are replaced by

$$C_{ij}(p_i - \beta_i + \beta_j - p_j), \tag{88}$$

where β_i represents a configurational stress or surface tension for component i . The references cited above give more information about the thermodynamic foundation of β_i and its specific form in the context of detonation theory. The term

$$-\frac{\beta_i}{\rho_i^2} D_i \phi_i \tag{89}$$

is added to the right-hand side of the thermodynamic Gibbs identity (14). The pressures p_i and p_j in the definition of the interfacial quantity p_{ij} have to be replaced by $p_i - \beta_i$ and $p_j - \beta_j$, respectively. After these modifications positive definite C_{ij} can again be proven to increase the entropy. The natural redefinition of the state of equilibrium (postulate 4) would prescribe equal $p_i - \beta_i$ for each i . Substitution of the state of equilibrium in the momentum equation prescribes that the momentum equation requires an additional term $-\nabla(\beta_i \phi_i) \neq 0$ to satisfy postulate 4. This term represents the surface tension force. The entropy law is not violated if the formulation of this term is in terms of Korteweg stresses (see [1]).

9 Conclusions

We have formulated three well-posed Navier-Stokes sets $A - C$, which for arbitrary N have been proven to satisfy the second-law of thermodynamics. In the derivation of these sets we have been guided by six plausible postulates. The Navier-Stokes sets can be regarded as macroscopic models for general multiphase flows consisting of N different components. As far as the author knows, this is the first general macroscopic flow theory which satisfies the second law of thermodynamics in the sense that the weighted sum of the individual entropies can analytically be shown to increase for arbitrary N . Appropriate averages of velocities and pressures have been considered to control the distribution of entropy production among the phases. To satisfy the entropy law, the interaction terms do not have to be algebraic. Irreversibility can be proven for interaction terms expressed in positive definite operators, following the example of the operatorial drag law proposed in the present paper. We also found an entropy increasing mass exchange law, dependent on Gibbs free energy differences.

The Navier-Stokes sets $A - C$ represent the case of single pressure, the case of multiple pressures with asymmetric equations, and the case of multiple pressures with symmetric equations, respectively. If we take $N = 2$, we recover standard two-phase formulations; set A becomes equivalent to the classic single pressure formulation, set B to the Baer and Nunziato approach, and set C to the Ransom and Hicks approach. It is remarked that the three sets do not necessarily exclude each other. For example, one may adopt equal pressures for N_1 components, where $N_1 < N$. There are more possible combinations which may enlarge the possibility to apply the present theoretical results to more specialistic scientific areas.

An extension of the drag law with high-order derivatives leads to well-posedness of the otherwise ill-posed set with a single pressure. For the incompressible one-dimensional case the characteristic velocities turn out to be roots of the polynomial of the degree $N - 1$ expressed by equation (74). Realness of roots

has been proven for two- and three-phase flows. There are indications (no rigorous proofs yet) that the roots are also real for $N \geq 4$.

We conclude that well-posedness can be achieved by the generalized drag law. For second-order derivatives the drag extension is part of the physical friction forces caused by the velocity difference between two components, somewhat similar to the Faxen force. It was known that well-posedness can be achieved by including a nonzero standard viscous stress, but this term is not an interaction force and acts everywhere in the flow. Using second-order derivatives the ill-posed infinite growth-rate for $k \rightarrow \infty$ becomes finite. For fourth-order derivatives, the growth-rate of the instability converges to zero for $k \rightarrow \infty$. Third-order derivatives of the velocity difference achieve even more; they entirely neutralize the instabilities of sufficiently short waves. The regularizing behavior of third-order derivatives is possibly related to the regularity of solutions of the Korteweg-de Vries equation.

Acknowledgements

The author acknowledges the University of Twente for use of their facilities, Cornelise Vreman-de Olde for a bright remark, and professor Prosperetti for comments and for sharing two chapters of his forthcoming book with G. Tryggvason and others.

Appendix 1: Entropy production

Entropy evolution equations can be derived in five steps: (a) derive the evolution equation for $\frac{1}{2}\rho_i\phi_i u_i \cdot u_i$ by multiplication of (7) with u_i , (b) subtract this equation from (8) to obtain the evolution equation for $\rho_i\phi_i e_i$, (c) derive $D_i e_i$, (d) apply the Gibbs identity to find $D_i \eta_i$, and (e) derive the evolution equation for $\rho_i\phi_i \eta_i$. The following identity for a scalar field f is frequently used,

$$\rho_i\phi_i D_i f + f G_i = \frac{\partial \rho_i\phi_i f}{\partial t} + \nabla \cdot (\rho_i\phi_i f u_i), \quad (90)$$

where G_i denotes the mass exchange term of component i , equal to the right-hand side of the continuity equation (6), which in general is not zero (section 8). The steps (a-c) lead to the internal energy equations:

$$\begin{aligned} \rho_i\phi_i D_i e_i &= \left(\frac{1}{2}u_i \cdot u_i - e_i\right)G_i + u_i \cdot M_i + E_i \\ &+ p_i \nabla \cdot \phi_i u_i + \sigma_i : \nabla u_i - \nabla \phi_i q_i. \end{aligned} \quad (91)$$

The continuity equations are rewritten:

$$\phi_i D_i \rho_i = -\rho_i(D_i \phi_i + \nabla \cdot \phi_i u_i) + G_i. \quad (92)$$

Substitution of the internal energy equation and the rewritten continuity equation into the Gibbs identity (14) yields $D_i \eta_i$ (step (d)). The evolution equation for $\rho_i\phi_i \eta_i$ is finally obtained by the application of (90), which leads to a term $\eta_i G_i$ at the right-hand side of the final entropy equations. Summation and partial integration and substitution of $G_i = 0$ delivers the entropy production formulated by equation (16), whereas the terms expressed in G_i lead to the entropy production due to mass exchange, equation (87).

To clarify the entropy production (70) and (87) we first take the entropy gained by component i due to the algebraic part of the momentum exchange process between i and j ; we divide equation (69) by T_i . Next, we add the entropy production gained by the component j due to the same process is (replace $b_{u_{ji}}$ with $b_{u_{ij}}$). The sum is equal to the first term in (70).

An alternative set of irreversible interaction laws is obtained if M''_{ij} , F''_{ij} and G''_{ij} are multiplied with a suitable average temperature, for example T_{uij} . It is logical to use drag coefficients a_{ijk} reversely proportional to T_{uij} , then. Note that an important physical parameter in models of interaction forces, the viscosity, is in general a function of temperature as well. After multiplication of M''_{ij} with T_{uij} , the effect of generalized drag on E''_{ij} can now be written as a very compact formula, $-u_{ij} \cdot M''_{ij}$, which implies strict locality of the total energy exchange terms, $E''_{ij} = -E''_{ji}$. Again, positive definite exchange laws guarantee a global increase of the total entropy. However, in contrast to E''_{ij} in section 5, the compact expression of E''_{ij} does not always induce locally positive changes of internal energies, which might lead to locally unphysical behavior of temperatures.

Appendix 2: Equilibrium

In this appendix, we investigate under which conditions zero values of the irreversible parts of the interaction terms imply the flow to be in the equilibrium state of postulate 4. Let us focus on the drag law and the state of equal velocities. Since $A_{ij} = A_{ji}$ and A_{ii} is irrelevant, the set of equations for N components contains $\frac{1}{2}N(N-1)$ independent operators A_{ij} . The equilibrium state of velocities, in which case all velocities are the same, implies zero total drag $M_i'' = 0$ for each component i . It appears that the nontrivial reverse statement is also true in most cases; zero total drag implies that all velocities are equal. To prove this we have to consider the system of N equations $M_i'' = 0$ with the N velocities as unknowns. Solutions of N equal velocities are the only solutions precisely then if the rank of the system equals $N-1$ (in the case of one spatial dimension). This seems to be true if no phase is isolated; for each component i there is at least one nonzero A_{ij} for some j .

We give a rigorous proof that the rank equals $N-1$ indeed for the case of nonzero algebraic drag laws, which means that each A_{ij} is a constant and nonzero if $i \neq j$. The coefficient matrix M for the problem $M_i'' = 0$ is an N^2 matrix with elements $m_{ij} = -A_{ij}$ if $i \neq j$, whereas

$$m_{ii} = \sum_{j \neq i} A_{ij}. \quad (93)$$

The matrix M is positive semidefinite, since it is symmetric and all eigenvalues are nonnegative, shown by standard Gershgorin theory. Since the state of equal velocities is a solution of $Mu = 0$, we know that the rank of M equals $N-1$ at most. Thus $\lambda = 0$ is a root of the characteristic polynomial of M . The rank of M is precisely $N-1$ if the algebraic multiplicity of the root $\lambda = 0$ equals one, or equivalently, the coefficient m_1 of the term $m_1\lambda$ in the characteristic polynomial is nonzero. This coefficient is given by

$$m_1 = - \sum_i \det(J_i), \quad (94)$$

where J_i is the principal minor of M , obtained by omitting both column and row i . Resorting to Gershgorin theory again, we find that all eigenvalues of M_i are strictly positive. Thus $\det(J_i)$ is strictly positive for each i . As a result $m_1 \neq 0$, which finishes the proof. The conditions for this result can be enlarged, but we do not elaborate on this.

Appendix 3: Well-posedness

We prove real characteristics for set A in case the incompressible momentum equations are extended with m -the order derivatives of velocity derivatives. These additional terms have the form $\partial\tau_{m-1,j}/\partial x$, where

$$\tau_{1,j} = \frac{\partial}{\partial x}(u_{j+1} - u_j), \quad j < N-1, \quad (95)$$

$$\tau_{i,j} = \frac{\partial}{\partial x}\tau_{i-1,j}, \quad i < m-1, \quad j < N-1. \quad (96)$$

In this way the higher-order terms can be considered as first-order derivatives of new variables, like Arai [2] did for the standard viscosity.

The incompressible, inviscid generalized eigenvalue problem with extended drag law takes the general form

$$\begin{vmatrix} R_{11} & R_{12} & 0 & 0 \\ 0 & R_{22} & R_{23} & 0 \\ 0 & R_{32} & 0 & 0 \\ 0 & 0 & 0 & R_{44} \end{vmatrix}. \quad (97)$$

The first column corresponds to the derivatives of ϕ_1 to ϕ_{N-1} , the second column to the derivatives of u_1 to u_N , the third column to the derivatives of p and $\tau_{m-1,j}$ and the final column to the derivatives of $\tau_{i,j}$ with $i \leq m-2$. The first row corresponds to the N continuity equations, the second row to the momentum equations the third row to the equations for $\tau_{1,j}$ and the last row to the equations for $\tau_{i,j}$ with $i \geq 2$. Note that the last row and column vanish if $m = 2$. For larger m the matrix R_{44} is simply the identity matrix of dimension $m-2$, hence the order of m does not influence the analysis, provided $m \geq 2$.

In the following we give the matrices R_{ij} for $N = 3$ and then it will be sufficiently clear which forms they have for other values of N . The matrices R_{11} and R_{12} in the continuity equations are

$$\begin{pmatrix} \lambda - u_1 & 0 \\ 0 & \lambda - u_2 \\ u_3 - \lambda & u_3 - \lambda \end{pmatrix}, \quad \begin{pmatrix} -\phi_1 & 0 & 0 \\ 0 & -\phi_2 & 0 \\ 0 & 0 & -\phi_3 \end{pmatrix}, \quad (98)$$

respectively. The momentum equations contains R_{22} and R_{23} ,

$$\begin{pmatrix} \tilde{\lambda}_1 & 0 & 0 \\ 0 & \tilde{\lambda}_2 & 0 \\ 0 & 0 & \tilde{\lambda}_3 \end{pmatrix}, \quad \begin{pmatrix} -\phi_1 & -a_{12} - a_{13} & -a_{13} \\ -\phi_2 & a_{12} & -a_{23} \\ -\phi_3 & a_{23} & a_{13} + a_{23} \end{pmatrix}, \quad (99)$$

respectively, where $\tilde{\lambda}_i = \rho_i(\lambda - u_i)$ and a_{ij} is a nonzero drag law coefficient in front of a derivative of order m . Finally, R_{32} equals

$$\begin{pmatrix} -1 & 1 & 0 \\ 0 & -1 & 1 \end{pmatrix}, \quad (100)$$

corresponding to the equations for $\tau_{1,j}$.

The general determinant (97) can be calculated for arbitrary N , by starting its development with R_{32} . The resulting characteristic equation reads

$$Q(\lambda)|R_{23}| = 0. \quad (101)$$

The determinant $|R_{23}|$ does not depend on λ and

$$Q(\lambda) = \begin{vmatrix} \lambda - u_1 & 0 & \dots & -\phi_1 \\ 0 & \lambda - u_2 & \dots & -\phi_2 \\ \vdots & \vdots & \dots & \vdots \\ u_N - \lambda & u_N - \lambda & \dots & -\phi_N \end{vmatrix}. \quad (102)$$

We rewrite

$$Q(\lambda) = \sum_i (\phi_i \prod_{j \neq i} (\lambda - u_j)). \quad (103)$$

The determinant $|R_{23}|$ should not vanish. For two components $|R_{23}|$ equals $-a_{12}$, which may be arbitrarily small, but nonzero. For $N = 3$ the determinant $|R_{23}|$ equals

$$-(a_{12}a_{23} + a_{13}a_{23} + a_{12}a_{13}), \quad (104)$$

which is unequal to zero if the three interaction coefficients a_{ij} have the same sign and at least two of them are nonzero.

The characteristic velocities are the roots of $Q(\lambda)$, a polynomial of degree $N - 1$. The challenge is to prove that these roots are real for general N . The proof is trivial for $N = 2$ and to prove real characteristics for $N = 3$ we rewrite $Q(\lambda)$ as $\lambda^2 + b\lambda + c$ with

$$b = -((1 - \phi_1)u_1 + (1 - \phi_2)u_2 + (1 - \phi_3)u_3), \quad (105)$$

$$c = \phi_1 u_2 u_3 + \phi_2 u_1 u_3 + \phi_3 u_1 u_2, \quad (106)$$

where we used $\sum \phi_i = 1$. The roots are real if $D = b^2 - 4c$ is nonnegative. To prove that this is always true we employ the Galilean invariance of the original problem; without loss of generality we may assume that $u_1 = 0$. The discriminant can then be written as

$$D = v_2^2 + v_3^2 + (4\xi_{23} - 2)v_2 v_3, \quad (107)$$

$$v_i = (1 - \phi_i)u_i, \quad (108)$$

$$\xi_{ij} = \frac{\phi_i \phi_j}{(1 - \phi_1)(1 - \phi_j)}. \quad (109)$$

Since $(1 - \phi_i) \geq \phi_j$ we find that ξ_{ij} is bounded between zero and one, hence

$$D \geq (|v_2| - |v_3|)^2 \geq 0. \quad (110)$$

This finishes the proof of the realness of characteristics for $N = 3$.

Appendix 4: Linear stability

This appendix contains the mathematical specifications referred to in section 7. First the matrices R_{ij} in definition (76) are specified for $N = 3$. Substituting $\lambda = \omega/k$ into expression (98) provides R_{11} and R_{12} , whereas R_{22} equals

$$\begin{pmatrix} \tilde{\lambda} - q_{12} - q_{13} & q_{12} & q_{13} \\ q_{12} & \tilde{\lambda} - q_{12} - q_{23} & q_{23} \\ q_{13} & q_{23} & \tilde{\lambda} - q_{13} - q_{23} \end{pmatrix}, \quad (111)$$

where $\tilde{\lambda} = \phi_i \rho_i (\lambda - u_i)$, and R_{23} equals

$$\begin{pmatrix} -\phi_1 \\ -\phi_2 \\ -\phi_3 \end{pmatrix}. \quad (112)$$

The constants q_{ij} are complex for even m and real for odd m . Algebraic source terms correspond to complex values of q_{ij} , independent of the wavenumber k .

Secondly, we specify the coefficients of the characteristic equation (77):

$$a = \phi_2^2 \phi_1 \rho_1 + \phi_1^2 \phi_2 \rho_2, \quad (113)$$

$$b = 2u_1 \phi_2^2 \phi_1 \rho_1 + 2u_2 \phi_1^2 \phi_2 \rho_2, \quad (114)$$

$$c = u_1^2 \phi_2^2 \phi_1 \rho_1 + u_2^2 \phi_1^2 \phi_2 \rho_2. \quad (115)$$

Thirdly, investigating the discriminant of polynomial (77), it is simply recognized that both roots are real in case q is real and $|q|$ is sufficiently high. For third order derivatives q can be written as $q = -k^2 d$, where d is the nonzero factor in front of the drag derivative. Consequently, the imaginary part of ω is zero and the wave is stable, provided k is sufficiently high. In fact this result extends to all odd $m \geq 3$.

Analyzing even orders of derivatives, we first consider $m = 2$. In that case q is complex and can be written as $q = ikd$, where d is the nonzero factor in front of the drag derivative. It can be verified by substitution that the following complex numbers are solutions of the quadratic polynomial (77) in the asymptotic limit $k \rightarrow \infty$:

$$\lambda_1 \sim \frac{b}{a} - \hat{u} - ik \frac{d}{a}, \quad (116)$$

$$\lambda_2 \sim \hat{u} + \frac{i}{kd} (a\hat{u}^2 - b\hat{u} + c) \quad (117)$$

For positive values of d and sufficiently high k , the maximum growth-rate approaches the imaginary part of λ_2 . It vanishes if $u_1 = u_2$, but otherwise it may be positive. Since $\omega = \lambda k$ and $\text{Im}(\lambda_2) \sim 1/k$, a potential instability leads to a constant growth-rate for $k \rightarrow \infty$. In case of fourth-order derivatives, k in the two roots should be replaced by $-k^3$ and d should be negative. Then the unstable growth-rate is not only finite, but converges to zero if $k \rightarrow \infty$. However, the exactly neutralizing behavior encountered for third-order derivatives is not reached.

References

- [1] D.M. Anderson, G.B. McFadden, A.A. Wheeler, Diffuse interface methods in fluid mechanics, *Ann. Rev. Fluid Mech.* 30, pp139-165 (1998).
- [2] M. Arai, Characteristics and stability analysis for two-phase flow equation systems with viscous terms, *Nucl. Sci. Eng.* 74, p77 (1980).
- [3] M.R. Baer, J.W. Nunziato, A two-phase mixture theory for the deflagration-to-detonation transition (DDT) in reactive granular materials, *Int. J. Multiphase Flow* 12, p861 (1986).
- [4] P. Bagchi, S. Balachandar, Effect of turbulence on the drag and lift of a particle, *Phys. Fluids* 15, p3496 (2003).
- [5] S. Banerjee, A.M.C. Chan, Separated flow models - I, *Int. J. Multiphase Flow* 6, p1 (1980).
- [6] J.B. Dzil, R. Menikoff, S.F. Son, A.K. Kapila and D.S. Stewart, Two-phase modeling of deflagration-to-detonation transition in granular materials: A critical examination of modeling issues, *Phys. Fluids* 11, p378 (1999).
- [7] M.M. Carroll, A. Holt, Static and dynamic pore-collapse for ductile porous materials, *J. Appl. Phys.* 43, p1626 (1972).
- [8] D.G. Crighton, Applications of KdV, *Acta Applicandae Mathematicae* 39, p39 (1995).
- [9] C. Crowe, M. Sommerfeld, M. Tsuji, *Multiphase flows with droplets and particles*, CRC Press LLC, Boston (1998).
- [10] D.A. Drew, Mathematical modeling of two-phase flow, *Ann. Rev. Fluid Mech.* 15, p261 (1983).
- [11] D.A. Drew, S.L. Passman, *Theory of multicomponent fluids*, Springer, New York (1998).
- [12] A. Einstein, Eine neue Bestimmung der Molek^uldimensionen, *Annalen der Physik* 19, 289 (1906).
- [13] M. Germano, U. Piomelli, P. Moin, W.H. Cabot, A dynamic subgrid-scale eddy-viscosity model, *Phys. Fluids A* 3, p1760 (1991).
- [14] M.A. Goodman, S.C. Cowin, A continuum theory for granular materials, *Arch. Ration. Mech. Anal.* 44, p249 (1972).
- [15] M. Ishii, *Thermo-fluid dynamic theory of two-phase flow*, Eyrolles (1975).
- [16] R.I. Issa, M.H.W. Kempf, Simulation of slug flow in horizontal and nearly horizontal pipes with the two-fluid model, *Int. J. Multiphase Flow* 29, p69 (2003).
- [17] R. Jackson, *The dynamics of fluidized particles*, Cambridge University Press (2000).
- [18] D.K. Lilly, A proposed modification of the Germano subgrid-scale closure method, *Phys. Fluids A* 4, p633 (1992).
- [19] A. Michalke, On the inviscid instability of the hyperbolic-tangent velocity profile, *J. Fluid Mech.* 19, p543 (1964).
- [20] H.C. Öttinger, *Beyond equilibrium thermodynamics*, Wiley & Sons, Inc., Hoboken New Jersey (2005)
- [21] S.L. Passman, J.W. Nunziato, E.K. Walsh, *A theory of multiphase mixtures*, Rational Thermodynamics, Springer-Verlag, Berlin, 2nd ed., pp286-325 (1984).
- [22] J.M. Powers, Two-phase viscous modeling of compaction of granular materials, *Phys. Fluids* 16, p2975 (2004).
- [23] A. Prosperetti, A.V. Jones, The linear stability of general two-phase flow models - II, *Int. J. Multiphase Flow*, 13, p161 (1987).
- [24] A. Prosperetti, G. Tryggvason, *Computational Methods for multiphase flow*, Cambridge University Press (2006).

- [25] J.D. Ramshaw, J.A. Trapp, Characteristics, stability, and short-wavelength phenomena in two-phase flow equation systems, *Nucl. Sci. Eng.* 66, p93 (1978).
- [26] V.H. Ransom, D.L. Hicks, Hyperbolic two-pressure models for two-phase flow, *J. Comp. Phys.* 53, p124 (1984).
- [27] R. Saurel, O. Lemetayer, A multiphase model for compressible flows with interfaces, shocks, detonation waves and cavitation, *J. Fluid Mech.*, 431, p239 (2003).
- [28] C. Truesdell, Thermodynamics of diffusion, Rational Thermodynamics, Springer-Verlag, Berlin, pp219-236 (1984).
- [29] A.W. Vreman, The adjoint filter operator in large-eddy simulation of turbulent flow, *Phys. Fluids* 16, p2012 (2004a).
- [30] A.W. Vreman, An eddy-viscosity subgrid-scale model for turbulent shear flow: Algebraic theory and application, *Phys. Fluids*, 16, p3670 (2004b).
- [31] B. Vreman, B. Geurts, H. Kuerten, Large-eddy simulation of the turbulent mixing layer, *J. Fluid Mech.* 339, p357 (1997).
- [32] L. van Wijngaarden, One-dimensional flow of liquids containing small gas bubbles, *Ann. Rev. Fluid Mech.* 4, p369 (1972).