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A common description for surface-tension and diffusion in two-component flows

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Abstract

This report tries to develop a common representation for diffusion and surface tension in two components newtonian flows. First, we make some preliminary considerations: (1) the Navier-Stokes equations of a fluid with two immiscible components is derived from the single components equations.In this case, the surface tension arrives from external consideration; (2) we shortly review the Cahn-Hilliard and Allen-Cahn equations describing a phase separation process, the surface-tension being considered as the terminal part of this process; and (3) we report a conservative representation of the surface tension and some of its variants.

Second, we reinterprete the conservative form of the single components as also valid for miscible fluids. Making an hypothesis common in diffusion phenomena, we make the surface tension naturally appear from the single phase equations. An anti-diffusive term connected with the surface tension is put in evidence. Compatibility with diffusion is discussed.

Contents

1   Introduction

The original aim of this work was the derivation of a conservative sharpening algorithm for free-surface flows. It is very common, to explain free-surface smearing, to invoque errors from the discretisation, from the convection or the temporal numerical schemes. The result is that the surface smearing is fighted with numerical schemes adaptation. Our feeling is that the core of the problems lies inside the set of solved equations. We therefore want to derive a set of equations whose numerical resolution should naturally give a sharp interface when it is due. Our basis is the Navier-Stokes equation for isothermal incompressible Newtonian homogeneous flows. However, most of the derivations are performed without using that the phase densities are constant. The paper is organised as follows. We derive rigorously the unclosed equations of a flow composed of two immiscible fluids. This derivation in some sense generalizes and simplifies the first part of the derivation performed by Zhang and Prosperetti [1] in their fundamental paper in 1993. This derivation allows us to introduce the notations and some of the issues. It is based on the use of generalised functions which are relatively touchy to manipulate. Second, we discuss the usual form given to the surface tension, with a particular emphasis on the usefulness of the conservative form. We present and discuss the Cahn-Hilliard and Allen-Cahn equation which were built to represent the separation of two immiscible fluids. We discuss the link between these equations and the surface tension. Third, we postulate a momentum equation of a single component fluid, having a constant density but not necessarily occupying all the local volume, introducing the concept of intrinsic volume fraction, that is a volume fraction having sense before any averaging process. This methodology is widely discussed and used in Drew and Passman [2]. The postulated equation is constrained to revert to the normal equation for the single component flow and to the equation derived at the beginning of the paper for immiscible two component flows. We discuss the constraints on the different terms of the postulated equation. We show how the postulated equation lead both to the appearance of an additional force in the overall momentum equation and to a drift term in the single phase transport equation, both terms being clearly related one to the other. We show that in the case of immiscible fluids, the force can be interpreted as a generalized surface tension, the drift term being related with the Cahn-Hilliard or Hallen Cahn equation. Conversely, we show that for miscible fluids, the drift term correspond to the classical diffusion while the force, being quadratic in the (already quite small) diffusion coefficient can usually be neglected. These considerations are possible thanks to the introduction of an initially not completely defined

length parameter. The two previous interpretations of the additional terms correspond to two different choices for the expression of this length.

2   System of equations for two immiscible fluids

We consider the case of two immiscible fluids A and B occupying different complementary parts of a shared common domain Ω. The volume occupied by the fluid A (resp. B)is characterised by the unit value of its support function "a" (resp. "b"). We will also use the subscript "a" (resp. "b") for all and any physical quantity related to fluid A (resp. B). Stating that "a" and "b" are support functions means that they can take only the discrete values 0 and 1. The property that together the two fluids occupy all the available volume is mathematically stated as:

$$a + b \;=\; 1 \;\text{ in } \Omega, \tag{1}$$

The property that the two fluids are immiscible is mathematically stated as:

$$ab \;=\; 0 \;\text{ in } \Omega, \tag{2}$$

The volume occupied by each phase is driven by the evolution of its boundary

$$\partial_t a + u_{\Gamma_a} \cdot \nabla a \;=\; 0 \;\text{ in } \Omega, \tag{3}$$

$$\partial_t b + u_{\Gamma_b} \cdot \nabla b \;=\; 0. \;\text{ in } \Omega, \tag{4}$$

where u_{Γ_a} is the velocity of the boundary of phase A.

We restrict our analysis to cases in which mass exchange can occur only through an existing interface. For example, a bubble of phase A cannot spontaneously appear in the bulk of phase B from nothing. It can however collapse and disappear if care is taken for the dependance of mass exchange with surface curvature. The interface velocity has two components, one is the fluid velocity, the other is related to the local mass transfer rate $\dot{m}_{ab} = -\dot{m}_{ba}$ between the phases of respective densities ρ_a and ρ_b.

$$\partial_t a + \left(u_a - \frac{\dot{m}_{ab}}{\rho_a} n_a\right) \cdot \nabla a \;=\; 0 \;\text{ in } \Omega, \tag{5}$$

$$\partial_t b + \left(u_b - \frac{\dot{m}_{ba}}{\rho_b} n_b\right) \cdot \nabla b \;=\; 0. \;\text{ in } \Omega, \tag{6}$$

where n_a (resp. n_b) is a regular vector field whose trace on the surface Γ_A (resp. Γ_B) coincide with the surface normal oriented towards the interior of

A (resp. B). This convention is opposite to the usual one but allows to have formally n_a and ∇a oriented in the same direction.

From equation 1 one has that

$$\partial_t(a+b) \quad = \quad 0 \quad \text{in } \Omega, \tag{7}$$

$$\nabla(a+b) \quad = \quad 0 \quad \text{in } \Omega \tag{8}$$

One should also get easily convinced that $n_a \cdot \nabla a = n_b \cdot \nabla b$ so that

$$\dot{m}_{ab} n_a \cdot \nabla a + \dot{m}_{ba} n_b \cdot \nabla b = 0, \tag{9}$$

but in general, in presence of mass transfer between fluids of different density, none of u_a, u_b and u_{Γ_a} coincide on the interface. Moreover, u_{Γ_a} and u_{Γ_b} must have only the normal component conciding on the interface.

Each fluid is supposed to follow the behaviour dictated by the Navier-stokes equations. That is:

$$\partial_t(\rho_a u_a) + \nabla \cdot (\rho_a u_a u_a) + \nabla P_a + \nabla \cdot \tau_a \quad = \quad \rho_a g + F_a \quad \text{in } \Omega_a(t) \tag{10}$$

$$\partial_t(\rho_b u_b) + \nabla \cdot (\rho_b u_b u_b) + \nabla P_b + \nabla \cdot \tau_b \quad = \quad \rho_b g + F_b \quad \text{in } \Omega_b(t) \tag{11}$$

where P_a is the phase A pressure, τ_a the extra-stress tensor, g the gravity acceleration and F_a the resultant of the internal forces, usually null for homogeneous phases, so it will be omitted hereafter. We do not forget the mass conservation equations, degenerating for constant phase density in the divergence free constraints:

$$\partial_t \rho_a + \nabla \cdot (\rho_a u_a) \quad = \quad 0 \quad \text{in } \Omega_a(t) \tag{12}$$

$$\partial_t \rho_b \nabla \cdot (\rho_b u_b) \quad = \quad 0 \quad \text{in } \Omega_b(t) \tag{13}$$

The problem is that the velocities u_a and u_b (but also the densities ρ_a and ρ_b) have no physical meaning respectively in domain Ω_b and Ω_a. But, because we want to write formulas valid in all the domain considered, we must extend u_a and u_b to the whole domain. The extension can be arbitrary but we want to be authorised to use the standard derivation rules like $\nabla \cdot (a u_a) = u_a \cdot \nabla a + a \nabla \cdot u_a$ taking into account that a is a discontinuous function. For this reason, we choose to consider regular extensions of u_a and u_b, preserving at least their continuity and the continuity of their divergence. Similarly, ρ_a and ρ_b are extended possibly by a constant value all over the domain. Note that we can not extend the divergence free property of u_a and u_b to the entire domain when the interface has closed contours, but we will not need it. Nevertheless, by continuity, if u_a and u_b are divergence-free in their respective domains, they are also divergence free on the interface. We could be however faced with mathematical inconsistency in pathological cases with discrete change

in the topology on the interface, such as the disappearance of a bubble or droplet.

The variables can be interpreted on a phase basis or on a global basis. The relation between them can be made by use of the "a" and "b" variable. The global variables can therefore be expressed in terms of the phase variables as follows.

$$\rho = a\rho_a + b\rho_b \tag{14}$$
$$P = aP_a + bP_b \tag{15}$$
$$u = au_a + bu_b \tag{16}$$
$$\rho u = a\rho_a u_a + b\rho_b u_b \tag{17}$$
$$\rho uu = a\rho_a u_a u_a + b\rho_b u_b u_b \tag{18}$$

These equations simply state that the density is the phase A density when $a = 1$ and is the phase B density when $a = 0$, and so on for pressure, momentum and ρuu. Specifically, u is really the velocity associated to ρ.

These definitions lead to additional properties that can prove useful for the derivation of the unclosed terms. For example:

$$\rho a = a\rho_a \tag{19}$$
$$\rho ua = a\rho_a u_a \tag{20}$$
$$a = a^2 \tag{21}$$

Moreover, all properties should remain true inverting "a" and "b".

The domain of the single phase Navier-Stokes equations can be extented to all Ω simply by multiplying by the respective support functions.

$$a\partial_t(\rho_a u_a) + a\nabla \cdot (\rho_a u_a u_a) + a\nabla P_a + a\nabla \cdot \tau_a = a\rho_a g \text{ in } \Omega \tag{22}$$
$$b\partial_t(\rho_b u_b) + b\nabla \cdot (\rho_b u_b u_b) + b\nabla P_b + b\nabla \cdot \tau_b = b\rho_b g \text{ in } \Omega \tag{23}$$

and

$$a\partial_t\rho_a + a\nabla \cdot (\rho_a u_a) = 0 \text{ in } \Omega \tag{24}$$
$$b\partial_t\rho_b + b\nabla \cdot (\rho_b u_b) = 0 \text{ in } \Omega \tag{25}$$

Turning back to the volume equation 5,6, multiplying by the respective density, we obtain the single phase density equations. It comes:

$$\rho_a\partial_t a + \rho_a u_a \cdot \nabla a = m_{ab}n_a \cdot \nabla a \tag{26}$$
$$\rho_b\partial_t b + \rho_b u_b \cdot \nabla b = m_{ba}n_b \cdot \nabla b \tag{27}$$

Summing the volume conservation equations and the mass conservation equations extended to the entire domain, we get the conservative form of the progress and regress variable equations:

$$\partial_t(\rho_a a) + \nabla \cdot (a\rho_a u_a) = \dot{m}_{ab} n_a \cdot \nabla a \tag{28}$$

$$\partial_t(\rho_b b) + \nabla \cdot (b\rho_b u_b) = \dot{m}_{ba} n_b \cdot \nabla b \tag{29}$$

Summing these two equations, we recognize the global mass conservation term. That is:

$$\partial_t \rho + \nabla \cdot (\rho u) = 0, \tag{30}$$

which is quite conforting.

To simplify the notation, we define the phase source term as:

$$\dot{S} = \dot{m}_{ab} n_a \cdot \nabla a \tag{31}$$

Therefore:

$$\partial_t(\rho_a a) + \nabla \cdot (a\rho_a u_a) = \dot{S} \tag{32}$$

$$\partial_t(\rho_b b) + \nabla \cdot (b\rho_b u_b) = -\dot{S} \tag{33}$$

The term $\dot{S}$ is here a scalar distribution with support on the interface. However equations 32 and 33 are valid even for variable distributions of the volume fractions. We introduce the notation: $\delta\frac{1}{\rho} = \frac{1}{\rho_b} - \frac{1}{\rho_a}$. Summing the original volume equations, we get a new time independent equation:

$$(u_a - u_b) \cdot \nabla a = \delta\frac{1}{\rho}\dot{S}, \tag{34}$$

equivalent to

$$\nabla \cdot u - [a\nabla \cdot u_a + b\nabla \cdot u_b] = \delta\frac{1}{\rho}\dot{S}, \tag{35}$$

or,using the incompressibility of each separated flow, if applicable:

$$\nabla \cdot u = \delta\frac{1}{\rho}\dot{S}. \tag{36}$$

Note that for the first time, we have broken the symmetry of representation.

Operating in a similar way, we will show how the global momentum equation can be retrieved from the momentum equation of both phases. Now, we incorporate the multipliers under the differential operators in equations 10,11 to get:

$$\partial_t(a\rho_a u_a) + \nabla \cdot (a\rho_a u_a u_a) + \nabla(aP_a) + \nabla \cdot (a\tau_a) = a\rho_a g$$
$$+ (\rho_a u_a)\partial_t a + (\rho_a u_a u_a) \cdot \nabla a + P_a \nabla a + \tau_a \cdot \nabla a \tag{37}$$

$$\partial_t(b\rho_b u_b) + \nabla \cdot (b\rho_b u_b u_b) + \nabla(bP_b) + \nabla \cdot (b\tau_b) = b\rho_b g$$
$$+ (\rho_b u_b)\partial_t b + (\rho_b u_b u_b) \cdot \nabla b + P_b \nabla b + \tau_b \cdot \nabla b \tag{38}$$

or, substituting with the progress and regress variable equations multiplied respectively by u_a and u_b:

$$\partial_t(a\rho_a u_a) + \nabla \cdot (a\rho_a u_a b u_a) + \nabla(aP_a) + \nabla \cdot (a\tau_a) = a\rho_a g +$$
$$u_a \cdot \dot{S} + P_a \nabla a + \tau_a \cdot \nabla a \qquad (39)$$
$$\partial_t(b\rho_b u_b) + \nabla \cdot (b\rho_b u_b u_b) + \nabla(bP_b) + \nabla \cdot (b\tau_b) = b\rho_b g$$
$$-u_b \cdot \dot{S} + P_b \nabla b + \tau_b \cdot \nabla b \qquad (40)$$

Direct averaging of these equations may lead to the Eulerian two-phase flow approach. Summing these two equations, we recognise on the left hand side the momentum equation for the global variables:

$$\partial_t(\rho u) + \nabla \cdot (\rho u u) + \nabla P + \nabla \cdot \tau = \rho g + (u_a - u_b) \cdot \dot{S}$$
$$+ (P_a - P_b)\nabla c + (\tau_a - \tau_b) \cdot \nabla c \, (41)$$

that is:

$$\partial_t(\rho u) + \nabla \cdot (\rho u u) + \nabla P + \nabla \cdot \tau = \rho g + F \qquad (42)$$

and

$$F = (u_a - u_b) \cdot \dot{S} + (P_a - P_b)\nabla a + (\tau_a - \tau_b) \cdot \nabla a. \qquad (43)$$

It means that the interpretation of a two-phases reactive flow as a unique flow leads to the appearance of a force term in the momentum equation localised on the interface. We will show that this force vanishes if we make the necessary hypothesis that the impulse and τ are continuous accross the interface, in a reference frame linked to the interface. If we assume that τ is continuous across the frontier then the former equation simplifies to:

$$F = (u_a - u_b) \cdot \dot{S} + (P_a - P_b)\nabla a. \qquad (44)$$

To keep invariance properties, we can assume that the tangential velocity is continuous accross the interface. Therefore, using equation 34:

$$(u_a - u_b) = \delta \frac{1}{\rho} m_{ab} \cdot n_a \qquad (45)$$

so that

$$(u_a - u_b) \cdot \dot{S} = \delta \frac{1}{\rho} \dot{m}_{ab}^2 \cdot \nabla a \qquad (46)$$

The pressure drop can be estimated considering that the total impulse is conserved through the interface in a reference frame linked to the interface. Considering that the interface velocity is $u_a - \dfrac{\dot{m}_{ab}}{\rho_a} \cdot n_a$, or equivalently $u_b - \dfrac{\dot{m}_{ba}}{\rho_b} \cdot n_b$ we have:

$$P_a + \rho_a[u_a - (u_a - \frac{\dot{m}_{ab}}{\rho_a} \cdot n_a)]^2 \quad = \quad P_b + \rho_b[u_b - (u_b - \frac{\dot{m}_{ba}}{\rho_b} \cdot n_b)]^2 \quad (47)$$

$$P_a - P_b \quad = \quad \rho_a[\frac{\dot{m}_{ab}}{\rho_a}]^2 - \rho_b[\frac{\dot{m}_{ba}}{\rho_b}]^2 \quad\quad (48)$$

leading to:

$$P_a - P_b = \dot{m}_{ab}^2 \delta \frac{1}{\rho} \quad\quad\quad\quad\quad\quad\quad\quad\quad\quad (49)$$

Combining this result with 46, then $F = 0$, that is:

$$\partial_t(\rho u) + \nabla \cdot (\rho uu) + \nabla P + \nabla \cdot \tau \quad = \quad \rho g. \quad\quad\quad (50)$$

In presence of a surface tension σ when at least one of the phases is liquid, the pressure difference is incremented by $\sigma(\nabla \cdot n_a)$ and $F = -\sigma(\nabla \cdot n_a)\nabla a+ < n_a, \nabla a > \nabla_S\sigma$, where ∇_S is the surface gradient. It should be noted that the surface tension does not appear in the single phase equations but only in the global one, defining an interface condition. It is also normal that the surface tension could not be completely directly derived from the single phase equations because the mere existence and the intensity of the surface tension is independent from the physical quantities present in the single phase equations. In other words, given the physical properties necessary to write their respective Navier-stokes equations, on can decide independently if they will be miscible or immiscible.

From both momentum equations, we have derived only one global equation. So, it seems that we have lost some information. The equation $F = -\sigma(\nabla \cdot n_a)\nabla a+ < n_a, \nabla a > \nabla_S\sigma$, as well as the impulse equation seem to come from the postulate that the Navier-Stokes equation is still valid for two-phase flows adding F on the RHS being the resultant of the internal forces, which was considered a priori zero for the single phase flows. In the literature, some attempt has been made to state a second equation, for example keeping one of the single phase equations. The problem is that the additional equation introduces a new variable, say u_a, which requires an additional relationship. Our approach will be not to derive but to postulate a second relationship using the same set of variables, allowing to close it.

3   On surface tension and immiscibility

Surface tension is a an intrinsic property of a medium pair. It appears only when at least one of the media is a liquid. Surface tension is also defined for a solid. It is used to determine the angle formed by two immiscible fluids on a

wall. It is also strongly linked with the concept of wettability. Here, we want to deal only with two immiscible fluids. So, at least one of them is liquid, because gases are always miscibles. What is strange at first glance is that liquids can continuously change to gas on a temperature/pressure diagram if they "travel" around the triple point. So, there is a continuous transformation of most if not all fluid pair from immiscibility characterized by a positive surface tension coefficient to miscibility characterized by a positive diffusion coefficient. The problem is that the surface tension is present in the momentum equation while the diffusion coefficient is in the mass fraction equation. But both should be representative of the same physico-chemical property. We would like to find a uniformized representation of the phenomena, at least for simple cases.

3.1   Cahn-Hilliard equation

One tentative to represent the behaviour of two immiscible fluids has led to the famous Cahn-Hilliard equation. The Cahn-Hilliard equation describes the evolution of a conserved concentration field during phase separaton [1]. One of its simplest isotropic version is given below:

$$\partial_t c = \alpha \Delta[(1 - 2c)c(1 - c)] - \beta \Delta^2 c. \tag{51}$$

where α and β are positive parameters and c has the same meaning of our "a" or "b". The first term in the RHS is a second order contracting term, while the second one is a fourth order diffusing term. Given a representative length λ, of the region of intermediary values of c (i.e. the diffuse interface length), the first RHS term scales like λ^{-2} will the second one like λ^{-4}. Therefore for large λ, the contracting term dominates while for small λ the diffusing term dominates. That is, a wide interface will tend to contract while a thin one will tend to dilate. The equilibrium is obtained when λ^2 is order β/α.

For our initial concern, the derivation of a naturally contracting system of equation, such that the interface length is no more than one or two computational cells, the fourth order equation is out of range. In effect, the discretisation of a fourth order operator requires (in 1D) at least five point or informations up to two cells distant in both direction. This is incompatible with the maximum two-cells interface objective. It is also not possible to just set $\beta = 0$ because the strength of the contracting term would result unbounded for arbitrary small discretization.

However, the idea that phase separation, for which surface tension is the final residual trace, must be based on energy considerations is very attractive.

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[1]http://www.ctcms.nist.gov/ wcraig/variational/node9.html

3.2   Allen-Cahn equation

The technical burden associated with the Cahn-Hilliard equation fourth order term has been recognized for long, see for example Yang et al.[3]. The result is that many authors have chosen to represent the same physical phenomena with a more simple equation, the Allen-Cahn equation, almost surely derived for the same reason. Its simplest form is:

$$\partial_t c = -\alpha(1 - 2c)c(1 - c) + \beta\Delta c \qquad (52)$$

this time, the diffusive term is the classical laplacian, scaling like λ^{-2} while the contracting term is scalar and scales like λ^0 with the equilibrium still obtained when λ^2 is order β/α.

This equation is much more convenient and has also been studied and simulated in a huge number of papers. While the order of the equation is convenient, it is straighforward to see that the equation is non-conservative. This defect has been recognized and has generally been corrected by use of a non local Lagrange multiplier:

$$\partial_t c = -\alpha(1 - 2c)c(1 - c) + \beta\Delta c + \epsilon(t). \qquad (53)$$

Unfortunately, this solution is completely unfit for multi-purpose CFD codes. In effect, the Lagrange multiplier is simply redistributing mass (a small quantity everywhere) slightly dirtying all the computational domain, just to comply with the overall mass preservation, even if mass preservation is "non negociable" in this context. By the way, only the scalar term is not conservative and it would be very convenient to find out a similar one (with regards to the scaling in λ) which would be conservative.

Both equations are constructed on the same scheme. Evolution of the concentration arises from the balance of two contributions, a linear higher order diffusive one impeding the degeneracy due to a non-linear lower order contracting one. In the Cahn-Hilliard equation, one cannot just set $\beta = 0$ because only a Laplacian with a "negative" diffusivity is left, and a phase separation to an unbounded energy of the surface. This feature is not true setting the coefficient β to zero in the Allen-Cahn equation. In this case, the concentration simply decays to zero or one depending whether the initial value is greater or lower that one half. We can also see that in this case, most of the physico-chemical process tentatively modelled is lost. However, we think that this loss of physical meaning is simply caused by the absence of local mass conservation. So, we will try to derive a conservative term, however scaling with a power of λ more than minus one so that its integral effect will vanish in the limit of a discontinuous interface.

3.3   representation of the surface tension

The classical representation of the surface tension σ is, as already written:

$$F = -\sigma(\nabla \cdot n_a)\nabla a + <n_a, \nabla a> \nabla_S \sigma \tag{54}$$

with $\nabla_S = (\delta ij - n_i n_j)\partial_j$ is the surface gradient. The formule above is valid only if the condition $\nabla n^2 = 0$ holds on the interface. That is if field n coinciding with with the unit normal on the interface is prolonged to a unit field out of the interface, at least at the first order (slope of normal module is zero on the interface). Otherwise, the term $\nabla \cdot n_a$ has to be changed to $\nabla_S \cdot n_a$.

The form of the surface tension is rather unusual, considering that it originates from a Gibbs potential. While it cannot be put under gradient form, it may be represented, as found out by several authors (find reference) under a divergential form, as follows:

$$F = -\nabla \cdot T \tag{55}$$
$$T = -\sigma <n_a, \nabla a> (\delta ij - n_i n_j). \tag{56}$$

This last formula can be given in a much more symmetrical form, using the identity matrix I:

$$T = G(I + n_a n_b) \tag{57}$$
$$G = -\sigma| <\nabla a, \nabla b> |^{0.5} \tag{58}$$

This formula may appear quite artificial but gives very interesting hints. First of all, it outlines the natural potential G associated to the surface tension. For the resolution of the momentum equation, the addition as it is of the potential in the surface tension allows the pressure to be continuous across a flat interface. This is a quite arbitrary choice, made almost surely for comodity. However, one can make another choice and incorporate G in the pressure. In this case, the surface tension becomes directly a tensorial product of the "natural" variables of the interface, n_a and ∇a, with σ being the overall strength of the phenomena.

Considering one of the two components of the tensorial product, that is $\frac{\nabla a}{|\nabla a|^{0.5}}$, we can have a look to what happens when the field "a" is connecting monotonically zero to one (in 1D) within a sharpening interval centred about zero. We have the following functional space properties:

1. $\frac{\nabla a}{|\nabla a|^{0.5}}$ tend to zero in L^p for $p \in [1, 2[$

2. $\frac{\nabla a}{|\nabla a|^{0.5}}$ is bounded in L^2 (in fact the L^2 norme is constant)

3. $\dfrac{\nabla a}{|\nabla a|^{0.5}}$ is unbounded in L^p for $p > 2$

4. $[\dfrac{\nabla a}{|\nabla a|^{0.5}}]^2$ is bounded in the dual space of $W_0^{1,1}$ and tends weekly to the Dirac mesure.

These properties are very specific and at the limit, the term will diseappear (in some sense) if present at the first power or will tend to the Dirac measure if present at the second power. We will show that this property will allow the surface tension to appear in the momentum equation and to disappear in phase transport equation when we consider the limit case of a discontinuous interface.

Another point which is worth noting is that whatever increasing derivable function Φ of "a" such that $\Phi(1) - \Phi(0) = 1$ can be used for the definition of F instead of "a".

For example, taking $\Phi = \dfrac{\rho^2}{\rho_a^2 - \rho_b^2}$, we have $\nabla \Phi = \dfrac{2\rho}{\rho_a + \rho_b}$ and

$$T \quad = \quad -\nabla G - \nabla \cdot \rho \frac{2\sigma}{\rho_a + \rho_b} < n_a, \nabla a > (\delta ij - n_i n_j), \qquad (59)$$

that is a potential part plus a convective transport with velocity $U = \sqrt{\dfrac{2\sigma}{\rho_a + \rho_b} < n_a, \nabla a >} n$ if we manage to give some meaning to the term under the square root.

This choice of Φ, making the density appear has already been made in the reference work of Blackbill and al. [4]. They noticed that doing this, the interface width was better conserved.

4 System of equations for two miscible fluids

In the first part of this document, we have derived the global Navier-stokes equations for density and momentum from the single phases equations in the limiting case of two immiscible phases. While the derivation can be made quite rigorous, the surface tension cannot be derived in this way but comes as an external consideration. To be naturally included in the final equation, the surface tension should be in some way included in the single phase equations. Looking at the Cahn-Hilliard and Allen-Cahn equations, we have seen that the surface tension can be interpreted as the terminal state of a phase separation process. However, these equations do not allow the phase separation to arrive at completion, having a small but non null asymptotic interface width. And unfortunately, degenerating the equations for zero asymptotic width leads to loss of physical meaning. What remains crucial in this approach is the energetic consideration: surface sharpens because doing this the fluid minimizes

its energy. In a momentum equation, this energy consideration should naturally appear as minus the gradient of this energy. By looking at a symmetrical form of the surface tension, we have obtained a good candidate for the form of this additional energy term, at least in its degenerated configuration. This is a start as it gives an asymptotic limit to be respected.

By postulating single phase equations in which mixing is allowed, we could insert the additional energy term which would be acting where mixing is effective. This is what we do now. We postulate a general form of the single phases momentum equations and try to eliminate the unknown terms by considerations on the overall momentum equation.

4.1 momentum equation

Our first postulate is that equations 22 and 23 are still valid for any volume fractions a and b and not only for unit support function.

$$\partial_t(a\rho_a u_a) + \nabla \cdot (a\rho_a u_a u_a) + \nabla(aP_a) + \nabla \cdot (a\tau_a) = a\rho_a g$$
$$+ (\rho_a u_a)\partial_t a + (\rho_a u_a u_a) \cdot \nabla a + P_a \nabla a + \tau_a \cdot \nabla a \quad (60)$$

$$\partial_t(b\rho_b u_b) + \nabla \cdot (b\rho_b u_b u_b) + \nabla(bP_b) + \nabla \cdot (b\tau_b) = b\rho_b g$$
$$+ (\rho_b u_b)\partial_t b + (\rho_b u_b u_b) \cdot \nabla b + P_b \nabla b + \tau_b \cdot \nabla b \quad (61)$$

We interpret the equations for the single phases has follows:

$$\partial_t(a\rho_a u_a) + \nabla \cdot (a\rho_a u_a u_a) + \nabla(aP_a) + \nabla \cdot (a\tau_a) = I_{ab} + a\rho_a g \quad (62)$$

$$\partial_t(b\rho_b u_b) + \nabla \cdot (b\rho_b u_b u_b)\nabla(bP_b) + \nabla \cdot (b\tau_b) = I_{ba} + b\rho_b g \quad (63)$$

where I_{ab} is the term of momentum exchange with the phase B.

The exchange term I_{ab} can be split into several parts: exchange by pressure, viscous effects or phase change. What is important is that the momentum exchange terms are opposite:

$$I_{ab} + I_{ba} = 0 \quad (64)$$

We make the very strong constructing hypothesis: $u_b - u_a$ and n_a are colinear and related through a dynamical equilibrium. That is:

$$u_b - u_u = U n_a \quad (65)$$

with U a functional depending on "a" but not explicitely on time. This hypothesis is quite strong but is commonly used in the usual description of diffusion phenomena. summing the two equations 62 and 63, we obtain:

$$\partial_t(\rho u) + \nabla \cdot (a\rho_a u_a b u_a + b\rho_b u_b b u_b) + \nabla P - \nabla \cdot (\mu \Pi u) - R_{ab} = \rho g$$

Here we have already used the following definitions:

1. $\rho = a\rho_a + b\rho_b$,

2. $u = \dfrac{a\rho_a u_a + b\rho_b u_b}{a\rho_a + b\rho_b}$,

3. $P = aP_a + bP_b$,

4. $\mu = a\mu_a + b\mu_b$.

Moreover, R_{ab} is defined by the relation

$$R_{ab} = \nabla \cdot (\mu\Pi u - a\mu_a \Pi u_a - b\mu_b \Pi u_b). \tag{66}$$

such that

$$R_{ab} = \nabla \cdot (a\mu_a \Pi \frac{b\rho_b(u_a - u_b)}{\rho} - b\mu_b \Pi \frac{a\rho_a(u_a - u_b)}{\rho}). \tag{67}$$

This term is neglected hereinafter. This is because it seems to be the reflect of a very simplified approximation of viscous effects rather than having a real proper physical meaning.

Now, we use the "well known" formula:

$$\rho uu = a\rho_a u_a u_a + b\rho_b u_b u_b - \frac{ab\rho_a\rho_b}{\rho}(u_b - u_a)(u_b - u_a) \tag{68}$$

and equation 65 to get:

$$\partial_t(\rho u) + \nabla \cdot (\rho uu) + \nabla P - \nabla \cdot (\mu\Pi u) = -\nabla \cdot \frac{ab\rho_a\rho_b}{\rho}U^2 n_a n_a$$
$$+ \rho g \tag{69}$$

Up to a pseudo-pressure term which can be included in the overall pressure, we retrieve the immiscible two-phase Navier-Stokes momentum equation with surface tension by choosing the following functional form:

$$U_1 = \pm\sqrt{\frac{\rho\sigma|\nabla a|}{ab\rho_a\rho_b}} \tag{70}$$

to obtain

$$\partial_t(\rho u) + \nabla \cdot (\rho uu) + \nabla P - \nabla \cdot (\mu\Pi u) = -\nabla \cdot (\sigma|\nabla a|n_a n_a) + \rho g \tag{71}$$

and

$$\partial_t(\rho u) + \nabla \cdot (\rho uu) + \nabla Q - \nabla \cdot (\mu\Pi u) = F + \rho g \tag{72}$$
$$F = \nabla \cdot (\sigma|\nabla a|(\delta_{ij} - n_a n_a)) \tag{73}$$

with $Q = P + \nabla(\sigma|\nabla a|)$.

The expression for U is rather complex. We can simplify it. Let Φ be primitive of $\alpha\frac{\rho_a\rho_b ab}{\rho}$ normaziled by the constant α such that $\Phi(1) - \Phi(0) = 1$. Then let use Φ instead of "a" in the definition of F. That is, we choose:

$$F_2 = \sqrt{\alpha\sigma|\nabla\phi|}. \tag{74}$$

Then F is retrieved by using the following alternative much simpler U

$$U_2 = \pm\sqrt{\alpha\sigma|\nabla a|}. \tag{75}$$

4.2 volume fraction equation

The volume fraction equations, when written in conservative form, remain valid for non discrete configurations:

$$\partial_t(\rho_a a) + \nabla \cdot (a\rho_a u_a) = \dot{S} \tag{76}$$

$$\partial_t(\rho_b b) + \nabla \cdot (b\rho_b u_b) = -\dot{S} \tag{77}$$

What we have to do is to relate each phase velocity to the global velocity and to the phase velocity difference:

$$u_a = u - \frac{b\rho_b}{\rho}(u_b - u_a) \tag{78}$$

$$u_b = u + \frac{a\rho_a}{\rho}(u_b - u_a) \tag{79}$$

so that

$$\partial_t(\rho_a a) + \nabla \cdot (a\rho_a u) - \nabla \cdot [\frac{ab\rho_a\rho_b}{\rho}(u_b - u_a)] = \dot{S} \tag{80}$$

$$\partial_t(\rho_b b) + \nabla \cdot (b\rho_b u) + \nabla \cdot [\frac{ab\rho_a\rho_b}{\rho}(u_b - u_a)] = -\dot{S} \tag{81}$$

or

$$\partial_t(\rho_a a) + \nabla \cdot (a\rho_a u) - \nabla \cdot [\frac{ab\rho_a\rho_b}{\rho}Un_a] = \dot{S} \tag{82}$$

$$\partial_t(\rho_b b) + \nabla \cdot (b\rho_b u) + \nabla \cdot [\frac{ab\rho_a\rho_b}{\rho}Un_a] = -\dot{S}. \tag{83}$$

If U is positive then the equation is diffusive while if U is negative, then the equation is contracting. Using the same choice as for the momentum equation (equation 75)with minus sign:

$$\partial_t(\rho_a a) + \nabla \cdot (a\rho_a u) + \nabla \cdot \frac{ab\rho_a\rho_b}{\rho}\sqrt{\alpha\sigma|\nabla a|}n_a = \dot{S} \tag{84}$$

In this last equation, the second order term $K = \nabla \cdot \frac{ab\rho_a\rho_b}{\rho}\sqrt{\alpha\sigma|\nabla a|}n_a$ has a clear anti-diffusive nature. It can be easily be seen that in the limit of a discrete interface, it is bounded in the dual space of H_0^1 and converges to zero weakly in the dual space of $W_0^{1,\infty}$. The dual nature of the equation will be subject to difficulties for discretisation.

We can also see that a simple expression for F leads to a complex one for K. Maybe we can find U such that both F and K have simple forms.

Moreover, the simplicity of the expressions depend whether one chooses the volume fraction as in these derivations or the mass fraction used in diffusion phenomena. When the volume fraction is used, an interesting form is obtained by taking the function Φ as $\Phi = \dfrac{\rho^2}{\rho_a^2 - \rho_b^2}$ so that $\nabla\Phi = \dfrac{2\rho}{\rho_a + \rho_b}\nabla a$ and $U = \sqrt{\frac{1}{2}(\frac{1}{\rho_a} + \frac{1}{\rho_b})\rho\sigma\dfrac{|\nabla a|}{ab}}$. In this case, the surface tension is expressed in a similar way to the convective transport (up to the usual pressure term), and the density harmonic mean appears in the expression of the velocity difference.

4.3   Constant density non reactive components

From now on, we suppose the phase densities constant. Dividing equations 82,83 by their respective density, and summing, we get the velocity divergence: what we have to do is relate each phase velocity to the global velocity and to the phase velocity difference:

$$\nabla \cdot u = \nabla \cdot [(\rho_a - \rho_b)\frac{ab}{\rho}Un_a] + \delta\frac{1}{\rho}\dot{S} \tag{85}$$

and in absence of source term, the velocity u^* defined by:

$$u^* = u - (\rho_a - \rho_b)\frac{ab}{\rho}Un_a \tag{86}$$

is divergence-free.

Rewritting in this case the volume fraction equation in term of u^*, we get:

$$\partial_t a + \nabla \cdot (au^*) + \nabla \cdot (abUn_a). \tag{87}$$

This last formulation is adapted for algorithms which consider the volume fraction, such as the VOF algorithm in Starccm+. It allows to choose U such as to preserve a 1D profile and choose the global behaviour for arbitrary large mixing layer. In effect, the only constraint given by the surface tension in the limit case of a discrete interface is precisely the asymptotic behaviour of U. In this case, U must be scaling as $\sigma|\nabla a|^{0.5}$.

From the work published in [5], if we can put the contracting term in the form:

$$abUn_a = -u_0 H(\delta)F(a)n_a \tag{88}$$

with u_0 a constant characteristic speed when $H(\delta)$ is chosen in adimensional form, $\delta^{-1} = f'(a)|\nabla a|$ and $F' = f$, then equation 87 has a self-similar solution whose profile is the inverse of the function f (so f must be monotonous) and whose time evolution is controled by $u_0 H(\delta)$.

For example, choosing $F(a) = -\frac{ab}{2}$ we have $f' = 1$ and $\lambda^{-1} = |\nabla a|$. We have seen from the momentum equation that the scaling must be in $H(\lambda) = \lambda^{-0.5} = |\nabla a|^{0.5}$. Then the self-similar profile is the ramp function, and we have in fact supposed $U = -\frac{u_0}{2}|\nabla a|^{0.5}$.

This is the simplest solution for the volume fraction equation. In fact, there is no need for a more regular profile, because it converges in finite time to the discontinuous Heavyside function. Requiring to reach it in a very smooth way does not make too much sense. For the momentum equation, it comes:

$$\frac{ab\rho_a\rho_b}{\rho}U^2 n_a n_a = \frac{u_0^2}{4}\frac{ab\rho_a\rho_b}{\rho}|\nabla a|n_a n_a \tag{89}$$

A lengthy calculation gives:

$$\int_{a=0}^{a=1} \frac{ab}{\rho}\nabla a = \frac{\rho_a\rho_b}{(\rho_a - \rho_b)^2}[\frac{1}{2}(\frac{1}{\rho_a} + \frac{1}{\rho_b}) - \frac{\ln\rho_a - \ln\rho_b}{\rho_a - \rho_b}] \tag{90}$$

allowing to match u_0 to σ with the formula

$$\sigma = \frac{u_0^2}{4}\frac{(\rho_a\rho_b)^2}{(\rho_a - \rho_b)^2}[\frac{1}{2}(\frac{1}{\rho_a} + \frac{1}{\rho_b}) - \frac{\ln\rho_a - \ln\rho_b}{\rho_a - \rho_b}]. \tag{91}$$

Using the equation 87, and reinterpreting the contracting term in a transport term of phase A, we can characterize the velocity of contraction as $V_c = \frac{u_0}{2}\lambda^{-0.5}$ (and in fact, in this case, u_0 is not a velocity).

4.4 Diffusive asymptotic

As already said, we consider that diffusion and surface tension can be two expressions of the same fundamental cause, the Gibbs energy. From the form of the contracting term in equation 82, it is clear that when ∇G and ∇a are in the same direction, the "contracting" term becomes diffusive. Moreover, in the limit of small concentration of one phase and small concentration gradient, we should revert to something quite similar to the classical diffusion term with constant coefficient. Thus we dispose of another asymptotic behaviour for the determination of U. Care must be taken to make the difference between mass and volume fractions. From [6], we have the reference diffusion equation:

$$\partial_t a\rho_a + \nabla \cdot (a\rho_a u) - \nabla \cdot (\rho\mathcal{D}_{ab}\nabla\frac{a\rho_a}{\rho}) = \dot{S}. \tag{92}$$

or in case of constant densities:

$$\partial_t a\rho_a + \nabla \cdot (a\rho_a u) - \nabla \cdot (\mathcal{D}_{ab}\frac{\rho_a\rho_b}{\rho}\nabla a) = \dot{S}. \tag{93}$$

Equating equations 82and 93, it remains:

$$\nabla \cdot \frac{\rho_a \rho_b}{\rho}(\mathcal{D}_{ab}\nabla a - abUn_a) = 0. \tag{94}$$

This leads to search for U with an asymptotic form in case of small gradient:

$$U = \mathcal{D}_{ab}\frac{|\nabla a|}{ab}. \tag{95}$$

Reporting this value in the momentum equation , we obtain:

$$F = -\nabla \cdot \mathcal{D}_{ab}^2 \frac{\rho_a \rho_b}{\rho}\frac{|\nabla a|^2}{ab}n_a n_a. \tag{96}$$

For a Gaussian profile, or its primitive, which are fundamental solutions of the diffusion equation, it is easy to see that $\frac{|\nabla a|^2}{ab}$ is uniformly bounded for a dispertion greater than any stricly positive constant. In this case, which is always verified for diffusive phenomena after a short time laps, we can see that F scales with $\mathcal{D}_{ab}^2$. As usually $\mathcal{D}_{ab}$ is closely related with the viscosity, which is already considered as a small parameter, we have that F basically scales with the square of viscosity and should very often be neglected. However, its effect is devastant if one wants to investigate the initial behaviour of mixing phases initially separated. In this case, the term is critically unbounded and funfamentally unsuited for arbitrary small times.

4.5   Matching the asymptotics

We already dispose of two asymptotic trends, one for a contractive behaviour, the other for a diffusive behaviour. With the additional asumptions, both asymptotics are valid for both behaviours, and we can try to match them in a unique functional U. The two conditions that we need to fullfil are:

1. U behaves like $U_1 = (\alpha\sigma|\nabla a|)^{0.5}$ when $|\nabla a|$ is large

2. U behaves like $U_2 = \mathcal{D}_{ab}|\nabla a|$ when $|\nabla a|$ is small.

this can be restated n terms of a characteristic lenght λ as

1. U behaves like $(\alpha\sigma\lambda^{-1})^{0.5}$ when λ is small

2. U behaves like $\mathcal{D}_{ab}\lambda^{-1}$ when λ is large

Matching the asymptote can be done by defining abU in terms of λ and decide for a suitable shape to define λ. The first step can be fulfilled in a number of way, always requiring the introduction of a reference length λ_0.

For example:

$$abU \sim \frac{\lambda_0}{\lambda}\frac{1}{\sqrt{1 + \frac{\lambda_0}{\lambda}}} \tag{97}$$

and also taking into account the constants:

$$abU = \frac{\mathcal{D}_{ab}}{\lambda} \frac{1}{\sqrt{1 + \frac{\mathcal{D}_{ab}^2}{\alpha^2 \sigma^2 \lambda}}} \qquad (98)$$

the characteristic length $\lambda_0 = \dfrac{\mathcal{D}_{ab}^2}{\alpha^2 \sigma^2}$ being obtained equating U_1 and U_2.

The problem is that the characteristic shape may be quite different for sharp interfaces and for large diffusing interfaces. So, it may be wiser not to look for a representation with an explicit reference shape. With the shape condition relaxed, the two behaviours can be matched by making their harmonic sum:

$$U = \frac{U_1 U_2}{U_1 + U_2} \qquad (99)$$

or

$$U^2 = \frac{U_1^2 U_2^2}{U_1^2 + U_2^2} \qquad (100)$$

depending on whether we consider the phase equation or the momentum equation as more fundamental. And we can take:

$$U_1 = \pm \mathcal{D}_{ab} \frac{|\nabla a|}{ab} \qquad (101)$$

$$U_2 = \pm \sqrt{\frac{\rho \sigma |\nabla a|}{ab \rho_a \rho_b}} \qquad (102)$$

4.6   Diffusion control

The diffusion caused by thermal agitation is a phenomena fundamentally different of the diffusion or contraction caused by a chemical potential. While for two miscible liquids, we can suppose that mixing is mainly driven by this chemical potential, the argument cannot stand for a gas and a liquid, or when it is difficult to characterize a phase rather as a gas or a liquid. Therefore, a term corresponding to the diffusion caused by thermal agitation should be present in the phase equation. We will name improperly this term as thermal diffusion. The classical diffusion term, with a scaling in λ^{-2} would dominate the contraction term in $\lambda^{-1.5}$ for small values of λ and would forbid the creation of a sharp interface. That is why the diffusion term must be modified. The classical diffusion term is known to be a good approximation of the diffusion phenomena only for smooth enough diffusion fronts. For very sharp fronts, the diffusion term should scale like λ^{-1}. The same procedure used to connect the asymptotics of the contraction term can be used to derive a

diffusion term scaling like λ^{-1} for sharp fronts and like λ^{-2} for diffuse fronts. For example, we can construct the modified diffusion term by taking:

$$U_1 = \mathcal{D}_{ab}\frac{|\nabla a|}{ab} \tag{103}$$

$$U_2 = U_0 \tag{104}$$

where U_0 is a constant.

Writing back the force term in the momentum equation corresponding to this generalized diffusion term, it is easy to see that it tends to zero for arbitrarily small interfaces.

By playing with the different asyptotic behaviour of both the chemical contraction and the thermal diffusion separately for small and large characteristic size, we can have a good chance to reproduce a variety of physical phenomena. For example, we can state that the chemical potential decays fast enough such that it cannot compensate for the thermal diffusion for large characteristic length. In this case, on can expect a quite stable small volume fraction of each phase in the other one. One should be also able to reproduce complex phenomena related to the vapour pressure between a gas and a liquid. This can be realized by taking the generalized diffusion term just given from one side and a contraction term scaling λ^{-n} with n greater than 2 for large λ.

While there is a large choice for the expression of the velocity difference U, the contraction, diffusion and force terms are conveniently writen choosing U asymptotically proportional to some power of $\dfrac{|\nabla a|}{ab}$, this power being at most one half otherwise the corresponding force becomes of unbounded integrale.

5   Conclusion

We have derived a uniform representation of two-component fluids exhibiting either a surface tension at their interface or a diffusive mixing behaviour. This representation has also the potential to naturally represent dissolution and vapor pressure phenomena. We interpret the surface tension as arising from a defect of the quadratic transport term in the overall momentum equation. The equations found out degenerate to the usual equations written for the asymptotic behaviours but can be used when this asymptotic behaviour is perturbed. This is particularly interesting for the simulation of immiscible fluids in which the discetization introduces a continuous perturbation of the asymptotic behaviour. This more general set of equation enforces the stability and attractivity of the asymptotic behaviour, correcting naturally the errors introduced by numerics.

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