On thermodynamic closures for two-phase flow with interfacial area concentration transport equation

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Abstract

The present work is a part of a modelling of forest fires fighting by aerial means. In this paper, we study different kind of closures for modelling two-phase flows with an almost "infinite range" of scales. Since theories like homogenization are not, in this case, relevant for obtaining the equivalent medium equations, the averaging method has been preferred. The variables are averaged by convolution with a smooth kernel with compact support, as the equations are non-linear, new quantities are defined in order to obtain the equations satisfied by averaged quantities; the entropy production is determined and closures or phenomenological equations are obtained using the second principle of thermodynamics. Main features of this work are, firstly a derivation in this framework of a balance equation for the interfacial area concentration and secondly, since this introduces a new unclosed variable: the mean velocity of interfaces, extended irreversible thermodynamics is used to obtain the general form of the appropriate closures equations.
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Keywords: Adiabatic multiphase flow; Interfacial area concentration; Extended irreversible thermodynamics

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1. Introduction

This paper is a part of a study on the modelling of forest fires fighting with aerial means, especially by dropping of fluids from aircrafts. Two main kinds of attacks are possible. The first one is direct attack which consists in dropping directly water on fire. In this case three main effects are expected: the blast effect due to the impact of fluid on the ground, the direct interaction of drops with flames (stopping the radiative transfer and cooling the flames) and the cooling of fuel (vegetation) in order to stop pyrolysis. The determination of the drop size distribution is a key for understanding and improving this kind of attack. The second kind of attack is the so called "indirect attack". A chemical additive is added to water. The fluid is dropped ahead of the fire front, the chemical playing the role of a barrier, the fire can thus be stopped to protect a spot or deviated to a place where ground fighting is easier. The ground distribution of fluid is the parameter which determines the effectiveness of the dropping. This part of the study is mainly concerned with the second kind of dropping. Dropping in Europe are made from an altitude of about 40 m while in North America the altitude is at least 100 m. During the dropping, the fluid goes out of the tank and will be partly atomized in the case of European dropping and totally atomized in the case of American dropping. In both case the production of interface is very important and an Eulerian simulation should take into account this fact. The first results on the subject have been provided by the USDA forest service in 70s (Swanson and Helvig, 1973, 1974). In these reports, valuable information was provided, as: mechanisms of atomization, many results of real dropping experiments, and a simple Lagrangian model, named PATSIM, has been developed in order to foresee ground pattern of dropping. The aim in the present paper is to provide a first attempt to model this kind of atomization in an Eulerian way. Let us notice that during the atomization process the Weber number is very high. We will neglect forces due to interfacial tension.

In this paper the spatial averaging method, with thermodynamic closure, will be used. This method consists of three steps: firstly balance relations at microscopic level are written in distributional form so that the jump conditions at interface appear as source term in the equations, secondly the equations are convoluted with a smooth kernel with compact support. As the equations are non-linear, new quantities are defined so that averaged equations take the form of balance relations. Thirdly, an entropy is constructed, and closure relations are defined such that the second thermodynamic principle is satisfied. In this paper, only the question of interaction between phases is addressed, and closure relations for this force are deduced assuming that both fluids are incompressible and that the equivalent medium is isothermal. However, consequences of extended thermodynamic are explored and a closure relation implying memory is obtained. In order to take into account the interface production, interfacial area concentration and interfacial velocity are defined by spatial averaging for being consistent with the averaging method. Then an evolution equation is deduced for these quantities. As the interfacial velocity is a non-closed quantity, closure relations for this velocity are obtained. Let us finally point out that from the general derivation obtained by thermodynamics, we will try systematically to recover empirical closures.

The paper is divided as follows. The second paragraph is devoted to the recall of the balance equations at microscopic level in distributional sense, and then these equations are averaged and new quantities are defined as interaction between phases. In the third paragraph the inter-

facial area concentration and the interfacial velocity are defined, an equation of evolution for these quantities is derived. Then in the fourth paragraph the entropy balance relation is derived and the production of entropy is obtained, by writing that this source term must be positive for all processes, some closure relations are obtained.

Related works are for the thermodynamics part, the pioneering paper of Bataille and Kestin (1977) on the thermodynamics of mixture. One can find an interesting generalization to an extended irreversible thermodynamics modelling of two-phase flow with phase changes by Bilicki et al. (2002) with a few references therein. Local interfacial area concentration has been introduced (though in a different form) by Ishii (1975) and Delhaye (1976) and recent works on a transport equation for this quantity have been reported by Morel et al. (1999), Delhaye (2001), Lhuillier et al. (2000), Lhuillier (2003, 2004) and by Veynante and Vervisch (2002) within the context of combustion theory.

2. Averaged equations of the models

2.1. The microscopic equations

Averaging method is simpler to apply if the physical laws are written in such a way that the boundary or interface conditions between phases are incorporated in the balance equations (Fitremann, 1977; Marle, 1982). For sake of being self content, let us recall this writing. The two phases occupy the domains Ω_k with $k = 1, 2$. Each quantity $\tilde{\varphi}_k$ associated to phase k is prolonged by 0 outside its domain of definition, in such a way that the new prolonged quantity $\bar{\varphi}_k$ is now defined everywhere. However this quantity is no more a function but becomes a distribution and derivatives must be considered as distributional derivatives. The relations between the definitions of derivatives are given in the following relations (Estrada and Kanwal, 1980):

$$
\begin{aligned}
\frac{\partial \bar{g}}{\partial t} &= \overline{\frac{\partial g}{\partial t}} + \mathbf{W} \cdot \mathbf{n}_k \bar{g} \delta_I \\
\nabla \cdot \bar{\mathbf{g}} &= \overline{\nabla \cdot \mathbf{g}} - \bar{\mathbf{g}} \cdot \mathbf{n}_k \delta_I \\
\nabla \bar{g} &= \overline{\nabla g} - \bar{g} \mathbf{n}_k \delta_I
\end{aligned}
\tag{1}
$$

In (1), $\frac{\partial \bar{g}}{\partial t}$, $\nabla \bar{g}$, and $\nabla \cdot \bar{\mathbf{g}}$ are derivatives in the distributional sense while $\overline{\frac{\partial g}{\partial t}}$, $\overline{\nabla g}$, $\overline{\nabla \cdot \mathbf{g}}$ are functions derivatives; moreover $\mathbf{W}$ is the interfacial velocity, $\mathbf{n}_k$ is the exterior normal and δ_I is the interfacial Dirac distribution defined by $\langle \delta_I, \varphi \rangle = \int_I \varphi(\mathbf{x}) \, dS(\mathbf{x})$. Let us notice that $\chi_k \bar{g} = \bar{g}$, so that we can use the convenient notation:

$$
\nabla \cdot \bar{\mathbf{g}} = \chi_k \nabla \cdot \bar{\mathbf{g}} + \bar{\mathbf{g}} \cdot \nabla \chi_k = \overline{\nabla \cdot \mathbf{g}} - \bar{\mathbf{g}} \cdot \mathbf{n}_k \delta_I
$$

With these notations the balance equations are the following:
Balance of mass:

$$
\frac{\partial \bar{\rho}_k}{\partial t} + \nabla \cdot (\bar{\rho}_k \overline{\mathbf{V}}_k) + \bar{\rho}_k (\overline{\mathbf{V}}_k - \mathbf{W}) \cdot \mathbf{n}_k \delta_I = 0
\tag{2}
$$

In this framework, jump conditions on the interface I are naturally included.

Balance of momentum of the kth phase:

$$\frac{\partial}{\partial t}(\bar{\rho}_k \overline{\mathbf{V}}_k) + \nabla \cdot (\bar{\rho}_k \overline{\mathbf{V}}_k \otimes \overline{\mathbf{V}}_k) = \bar{\rho}_k \mathbf{g} - \nabla \bar{p}_k + \nabla \cdot \bar{\tau}_k - \bar{\rho}_k \overline{\mathbf{V}}_k (\overline{\mathbf{V}}_k - \mathbf{W}) \cdot \mathbf{n}_k \delta_I + (-\bar{p}_k \mathbf{n}_k + \bar{\tau}_k \cdot \mathbf{n}_k) \delta_I$$

(3)

The total stress tensor is denoted by $\overline{\Sigma}_k = -\bar{p}_k \mathbf{1} + \bar{\tau}_k$. The viscous stress tensor is $\bar{\tau}_k = \mu_k({}^t\nabla \overline{\mathbf{V}}_k + \nabla \overline{\mathbf{V}}_k), p_k$ is the pressure. It can again be noticed that, in this framework, jump conditions at the interface are given by the singular part of (3).

Balance of energy:

$$\frac{\partial}{\partial t}\left(\bar{\rho}_k\left(\bar{e}_k + \frac{1}{2}\overline{\mathbf{V}}_k^2\right)\right) + \nabla \cdot \left(\bar{\rho}_k\left(\bar{e}_k + \frac{1}{2}\overline{\mathbf{V}}_k^2\right)\overline{\mathbf{V}}_k + \bar{\mathbf{q}}_k - \overline{\Sigma}_k \cdot \overline{\mathbf{V}}_k\right)$$

$$= \bar{\rho}_k \mathbf{g} \cdot \overline{\mathbf{V}}_k - \left(\mathbf{q}_k + \bar{\rho}_k\left(\bar{e}_k + \frac{1}{2}\overline{\mathbf{V}}_k^2\right)(\overline{\mathbf{V}}_k - \mathbf{W}) - (-\bar{p}_k \mathbf{1} + \bar{\tau}_k) \cdot \overline{\mathbf{V}}_k\right) \cdot \mathbf{n}_k \delta_I$$

(4)

The equation satisfied by the characteristic function χ_k of the phase k is the so-called topological equation:

$$\frac{\partial \chi_k}{\partial t} + \nabla \cdot (\chi_k \overline{\mathbf{V}}_k) + (\overline{\mathbf{V}}_k - \mathbf{W}) \cdot \mathbf{n}_k \delta_I = \overline{\nabla \cdot \mathbf{V}_k}$$

(5)

2.2. Averaged equations, and averaged quantities

We can now proceed with the averaging (see Marle (1982) for example). Let us consider now, a spatial filter or a kernel, $W(\mathbf{r}, h)$, which is a mollifier, that is $W \geqslant 0$, $\int W(\mathbf{r}, h)\, d\mathbf{r} = 1$, the support of W is the sphere of radius h, and $W \underset{h \to 0}{\to} \delta$. Quantities and equations will be smoothed by convolution with this kernel. The volume fraction $\phi_k(\mathbf{r}, t)$ of the phase k is defined by:

$$[\phi_k(\mathbf{r}, t)] = \chi_k * W = \int \chi_k(\mathbf{r} - \mathbf{r}', t) W(\mathbf{r}', h)\, d\mathbf{r}' = \int \chi_k(\mathbf{r}', t) W(\mathbf{r} - \mathbf{r}', h)\, d\mathbf{r}'$$

(6)

So that the averaged value of any quantity $\bar{\varphi}_k$ is defined by:

$$\phi_k(\mathbf{r}, t)[\varphi_k(\mathbf{r}, t)] = (\chi_k \bar{\varphi}) * W = \int \chi_k(\mathbf{r}', t) \bar{\varphi}(\mathbf{r}', t) W(\mathbf{r} - \mathbf{r}', h)\, d\mathbf{r}'$$

(7)

In relations (6) and (7), we have put between brackets [ ], the newly defined macroscopic quantity φ_k. This notation will be used all along this text when new macroscopic quantities are introduced. It is useful to point out that [ ] is not considered as an averaging operator thought it is related to the averaging process.

The averaged density is then defined by:

$$\phi_k[\rho_k] = (\chi_k \bar{\rho}_k) * W$$

(8)

Two averaged velocities can be defined; the first one is a "mass averaged" velocity,

$$\phi_k \rho_k[\mathbf{V}_k] = (\chi_k \bar{\rho}_k \overline{\mathbf{V}}_k * W)$$

(9)

and the second one is a "volume averaged" velocity,

$$\phi_k[\mathbf{U}_k] = \chi_k \overline{\mathbf{V}}_k * W \tag{10}$$

As it will be useful, we can define now as usual, the total density

$$\rho = \sum_{k=1}^{2} \rho_k \phi_k \tag{11}$$

and the mass average velocity

$$\rho \mathbf{V} = \sum_{k=1}^{2} \rho_k \mathbf{V}_k \phi_k \tag{12}$$

One can now convolute the balance equations with the kernel W. As the equations are non-linear, some new "macroscopic" quantities appear. So that, this averaging procedure gives the macroscopic equations and the new macroscopic quantities:

The topological equation (5) gives:

$$\frac{\partial \phi_k}{\partial t} + \nabla \cdot (\phi_k \mathbf{U}_k) = \omega_k \tag{13}$$

with

$$[\omega_k](\mathbf{x}) = \int_I \chi_k(\mathbf{y}) \nabla \cdot \overline{\mathbf{V}}_k(\mathbf{y}) W(\mathbf{x} - \mathbf{y}) \, \mathrm{d}\mathbf{y} - \int_I (\overline{\mathbf{V}}_k(\mathbf{y}) - \mathbf{W}(\mathbf{y})) \cdot \mathbf{n}_k(\mathbf{y}) W(\mathbf{x} - \mathbf{y}) \, \mathrm{d}s(\mathbf{y}) \tag{14}$$

The balance of mass (2) becomes:

$$\frac{\partial (\phi_k \rho_k)}{\partial t} + \nabla \cdot (\phi_k \rho_k \mathbf{V}_k) = -(\bar{\rho}_k (\overline{\mathbf{V}}_k - \mathbf{W}) \cdot \mathbf{n}_k) \delta_I * W \tag{15}$$

One can notice that in the case of condensation $(\mathbf{W} - \mathbf{V}_k) \cdot \mathbf{n}_k > 0$, then the mass source term in (15) is positive which is consistent. The averaged balance of momentum is:

$$\frac{\partial}{\partial t}(\rho_k \phi_k \mathbf{V}_k) + \nabla \cdot (\rho_k \phi_k \mathbf{V}_k \otimes \mathbf{V}_k) = \rho_k \phi_k \mathbf{g} + \nabla \cdot \phi_k \boldsymbol{\sigma}_k + \mathbf{F}_{mk} + \mathbf{F}_{Ik} \tag{16}$$

In relation (16) the following definitions have been used:

$$\rho_k \phi_k \mathbf{V}_k \otimes \mathbf{V}_k - \phi_k [\boldsymbol{\sigma}_k] = (\bar{\rho}_k \overline{\mathbf{V}}_k \otimes \overline{\mathbf{V}}_k + \bar{p}_k \mathbf{1} - \bar{\boldsymbol{\tau}}_k) \delta_I * W \tag{17}$$

$$[\mathbf{F}_{mk}] = -\bar{\rho}_k \overline{\mathbf{V}}_k (\overline{\mathbf{V}}_k - \mathbf{W}) \cdot \mathbf{n}_k \delta_I * W \tag{18}$$

$$[\mathbf{F}_{Ik}] = (-\bar{p}_k \mathbf{n}_k + \bar{\boldsymbol{\tau}}_k \cdot \mathbf{n}_k) \delta_I * W \tag{19}$$

As usual, we can notice from (17) that the macroscopic stress tensor $\boldsymbol{\sigma}_k$ is not the average of the microscopic tensor $\bar{\boldsymbol{\sigma}}_k$.

The averaged energy balance reads:

$$\frac{\partial}{\partial t}\left(\phi_k \rho_k \left(e_k + \frac{1}{2} \mathbf{V}_k^2\right)\right) + \nabla \cdot \left(\phi_k \rho_k \left(e_k + \frac{1}{2} \mathbf{V}_k^2\right) \mathbf{V}_k + \mathbf{q}_k - \phi_k \boldsymbol{\sigma}_k \cdot \mathbf{V}_k\right)$$

$$= \rho_k \phi_k \mathbf{g} \cdot \mathbf{V}_k - P_k \frac{\partial \phi_k}{\partial t} + X_k + Y_k \tag{20}$$

with the following definition for the average internal energy:

$$\rho_k \phi_k \left([e_k] + \frac{1}{2} \mathbf{V}_k^2 \right) = \bar{\rho}_k \left(\bar{e}_k + \frac{1}{2} \overline{\mathbf{V}}_k^2 \right) * W \tag{21}$$

And for the energy supply X_k and Y_k due to the interface, the definitions are:

$$[X_k] - P_k \frac{\partial \phi_k}{\partial t} = (-\bar{\mathbf{q}}_k + (-\bar{p}_k \mathbf{1} + \bar{\mathbf{\tau}}_k) \cdot \overline{\mathbf{V}}_k) \cdot \mathbf{n}_k \delta_I * W \tag{22}$$

$$[Y_k] = -\bar{\rho}_k \left(\bar{e}_k + \frac{1}{2} \overline{V}_k^2 \right) (\overline{\mathbf{V}}_k - \mathbf{W}) \cdot \mathbf{n}_k \delta_I * W$$

The heat flux $\mathbf{q}_k$ can be defined by:

$$\rho_k \phi_k \left(e_k + \frac{1}{2} \mathbf{V}_k^2 \right) \mathbf{V}_k + [\mathbf{q}_k] - \phi_k \mathbf{\sigma}_k \cdot \mathbf{V}_k = \left(\bar{\rho}_k \left(\bar{e}_k + \frac{1}{2} \overline{\mathbf{V}}_k^2 \right) \overline{\mathbf{V}}_k + \bar{\mathbf{q}}_k - \bar{\mathbf{\Sigma}}_k \cdot \overline{\mathbf{V}}_\mathbf{k} \right) * \mathbf{W} \tag{23}$$

Some new terms have been defined and closure relations are needed for them. For the applications we have in mind, we will assume a ''no slip'' condition at interfaces so that the interfacial velocity $\mathbf{W}$ is equal to the fluids velocities at interfaces, and then the force $\mathbf{F}_{mk}$ and the energy transfer term Y_k are null. This implies that no phase change does occur. We will be especially concerned with the interaction force $\mathbf{F}_{Ik}$ defined by relation (19). Usually this force is decomposed in three parts:

$$\mathbf{F}_{Ik} = \mathbf{F}_{Lk} + \mathbf{F}_{vmk} + \mathbf{F}_{fk}$$

Since a great difference of density between the two fluids is assumed, we will not consider the lift force $\mathbf{F}_{Lk}$. In the same way, the added mass force $\mathbf{F}_{vmk}$, which is of the same order as the lift force, will be neglected. It remains to obtain a model for the drag force $\mathbf{F}_{fk}$. As a closure relation for interaction force between phase p and phase q, some authors (see Morel (1997) and Drew and Passman (1998) for an extensive exposition) have proposed expressions like:

$$F = -\frac{1}{8} A \rho_p \langle C_D | \mathbf{V}_p - \mathbf{V}_q | (\mathbf{V}_p - \mathbf{V}_q) \rangle_A$$

where the symbol $\langle \rangle$ stands for interfacial averaging with the probability density distribution function and A is the interfacial area concentration. The presence of interfacial area concentration A appears desirable because this quantity partly drives the interaction between phases. In order to be coherent with our treatment, we have to consider a spatial averaged interfacial area concentration and not an ensemble averaged density. In the next paragraph we will derive an equation for the interfacial area concentration.

3. Interfacial area concentration

As mentioned previously, this section will be devoted to the derivation of an adapted equation for the interfacial area concentration. Let us firstly define this density as coarse-grained surface area density:

$$[A] = \int_I W(\mathbf{x} - \mathbf{y}, h) \, \mathrm{d}s(\mathbf{y}) = W * \delta_I \tag{24}$$

The symbol δ_I stands for the Dirac measure on the interface I (i.e. $\langle \delta_I, \varphi \rangle = \int_I \varphi \, dS$ for any function φ indefinitely derivable with compact support. δ_I can be considered as the fine-grained surface area density. One can verify that as a function of h the relation $A \underset{h \to 0}{\longrightarrow} \delta_I$ holds. Let us emphasize that h is the size of the support of the smoothing function and can be interpreted as the size of the fluid particle or "elementary representative volume" of the equivalent medium being constructed.

In the same way the interfacial mean velocity is defined by:

$$A[\mathbf{V}_I] = \int_I W(\mathbf{x} - \mathbf{y}, h)\mathbf{V}(\mathbf{y}, t)\, ds(\mathbf{y}) = W * (\mathbf{V}\delta_I) \tag{25}$$

Let us consider the total (or material time) derivative of A, from (24) we obtain using a transport theorem for surface integral:

$$\frac{dA(\mathbf{x}, \mathbf{t})}{dt} = \int_I \nabla W \cdot \left(\mathbf{V}_\mathbf{x} - \mathbf{V}_\mathbf{y}\right) ds(\mathbf{y}) + \int_I W(\mathbf{x} - \mathbf{y})(\nabla \cdot \mathbf{V} - \nabla \mathbf{V} : \mathbf{n} \otimes \mathbf{n})\, ds(\mathbf{y})$$

The symbol $\otimes$ represents the tensorial product and $\nabla \mathbf{V} : \mathbf{n} \otimes \mathbf{n}$ is the contracted product of the two tensors. Then we obtain:

$$\begin{aligned}\frac{dA(\mathbf{x}, \mathbf{t})}{dt} = &\mathbf{V}(\mathbf{x}, \mathbf{t}) \cdot \int_I \nabla W \, ds(\mathbf{y}) - \int_I \nabla W \cdot \mathbf{V}(\mathbf{y}, t)\, ds(\mathbf{y}) \\ &+ \int_I W(\mathbf{x} - \mathbf{y})(\nabla \cdot \mathbf{V} - \nabla \mathbf{V} : \mathbf{n} \otimes \mathbf{n})\, ds\end{aligned}$$

As for distributions, convolution and derivation can commute, the first term of the right hand side can be written $\mathbf{V}(\mathbf{x}, \mathbf{t}) \cdot \int_I \nabla W \, ds(\mathbf{y}) = \mathbf{V}(\mathbf{x}, \mathbf{t}) \cdot \nabla_\mathbf{x} A$ and likely the second term is $\nabla_\mathbf{x} \cdot (A\mathbf{V}_I)$. We obtain then

$$\frac{dA}{dt} = \nabla_\mathbf{x} A \cdot \mathbf{V}(\mathbf{x}, \mathbf{t}) - \nabla_\mathbf{x} \cdot (A\mathbf{V}_I) + \int_I W(\mathbf{x} - \mathbf{y})(\nabla \cdot \mathbf{V} - \nabla \mathbf{V} : \mathbf{n} \otimes \mathbf{n})\, ds$$

But taking into account that

$$\frac{dA}{dt} = \frac{\partial A}{\partial t} + \nabla_\mathbf{x} A \cdot \mathbf{V}(\mathbf{x}, \mathbf{t})$$

and after a simplification, the result reads:

$$\frac{\partial A}{\partial t} + \nabla \cdot (A\mathbf{V}_I) = \int_I W(\mathbf{x} - \mathbf{y})(\nabla \cdot \mathbf{V} - \nabla \mathbf{V} : \mathbf{n} \otimes \mathbf{n})\, ds \tag{26}$$

Let us point out that using space averaging circumvents potential mathematical problems (related to measure theory) and physical problem (related to ergodicity and ensemble averaging). Another advantage of using volume averaging is that the right hand side of (26) can be simplified using a "surface green formula" (cf. the Lemma 1 given in Appendix A):

$$\begin{aligned}\int_I W(\mathbf{x} - \mathbf{y})(\nabla \cdot \mathbf{V} - \nabla \mathbf{V} : \mathbf{n} \otimes \mathbf{n})\, ds = &\int_I \left(\frac{\partial W}{\partial \mathbf{n}} + CW\right)\mathbf{V} \cdot \mathbf{n}\, ds \\ &+ \int_\Gamma W\mathbf{V}_S \cdot \mathbf{n}_v \, dl - \int_I \mathbf{V} \cdot \nabla W \, ds\end{aligned} \tag{27}$$

where C is the mean curvature of the interface, $\mathbf{V}_S$ is the tangential component of the velocity $\mathbf{V}$, Γ is the line boundary of the interface I, $\mathbf{n}_v$ is the unit vector normal to Γ. Using ∇_S the gradient and the divergence operator on the surface (Aris, 1962), we can obtain:

$$\int_I W(\mathbf{x} - \mathbf{y})(\nabla \cdot \mathbf{V} - \nabla \mathbf{V} : \mathbf{n} \otimes \mathbf{n})\,\mathrm{d}s = \int_I \{CW\mathbf{V} \cdot \mathbf{n}\}\,\mathrm{d}s + \int_\Gamma W\mathbf{V}_S \cdot \mathbf{n}_v\,\mathrm{d}l$$
$$- \left(\int_I \nabla_S \cdot (W\mathbf{V}_S) - W\nabla_S \cdot (\mathbf{V}_S)\,\mathrm{d}s \right) \tag{28}$$

We can use now the Stoke's theorem on the interface I, taking into account the possible discontinuities lines Γ_i of the tangent velocity on the interface, and we obtain:

$$\int_I W(\mathbf{x} - \mathbf{y})(\nabla \cdot \mathbf{V} - \nabla \mathbf{V} : \mathbf{n} \otimes \mathbf{n})\,\mathrm{d}s = \int_I \{CW\mathbf{V} \cdot \mathbf{n}\}\,\mathrm{d}s + \int_I W\nabla_S \cdot \mathbf{V}_S\,\mathrm{d}s$$
$$+ \sum_{i=1}^n \int_{\Gamma_i \cap I} W[\![\mathbf{V}_S \cdot \mathbf{n}_v]\!]\,\mathrm{d}l \tag{29}$$

where the symbol $[\![\ ]\!]$ stands for the discontinuity of the bracketed quantity on crossing the line Γ_i. For instance, $[\![\mathbf{V}_S \cdot \mathbf{n}_v]\!] = (\mathbf{V_L} - \mathbf{V_R}) \cdot \mathbf{n}_v$ in the left case of Fig. 1 and more generally, $[\![\mathbf{V}_S \cdot \mathbf{n}_v]\!] = \mathbf{V_L} \cdot \mathbf{n}_v^L - \mathbf{V_R} \cdot \mathbf{n}_v^R$, when the unit normal, tangent to the discontinuity line, differs between the left and right side of the discontinuity. Thus the evolution equation for the interfacial area concentration is:

$$\frac{\partial A}{\partial t} + \nabla \cdot (A\mathbf{V}_I) = S_c + S_e + S_{fc} \tag{30}$$

The right hand side of (30) contains three source terms:

$$S_c = \int_I WC\mathbf{V} \cdot \mathbf{n}\,\mathrm{d}s \tag{31}$$

this term corresponds to the presence of curvature,

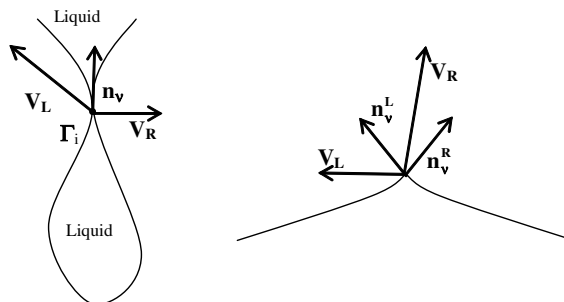$$S_s = \int_I W\nabla_S \cdot \mathbf{V}_S\,\mathrm{d}s \tag{32}$$

Fig. 1. Discontinuity of surface velocity during breakup.

this term corresponds to the stretching of the interface, and

$$S_{fc} = \sum_{i=1}^{n} \int_{\Gamma_i \cap I} W [\![\mathbf{V}_S \cdot \mathbf{n}_v]\!] \, \mathrm{d}l \tag{33}$$

which corresponds, because of the possible discontinuities of the normal (or of the tangent plane), to fragmentation and coalescence of drops. The following relation will be used in the following:

$$S = S_c + S_s + S_{fc} \tag{34}$$

Of course the final form of (30) is similar to the one proposed initially by Ishii (1975). It is fruitful to compare this result with the same kind of equation derived recently by other authors. Actually Eq. (26) can be related to (Lhuillier, 2003) formulation:

$$\frac{\partial \delta_I}{\partial t} + \mathbf{V} \cdot \nabla \delta_I = -\delta_I \mathbf{n} \mathbf{n} \cdot \nabla \mathbf{V} \tag{35}$$

Relation (35) is written in the distributional sense and then averaged over different realisations of the physical phenomena. Previously, Lhuillier et al. (2000) had derived the following relation:

$$\frac{\partial \delta_I}{\partial t} + \nabla \cdot ((\mathbf{V}_I \cdot \mathbf{n})\mathbf{n}\delta_I) = (\mathbf{V}_I \cdot \mathbf{n})\nabla \cdot \mathbf{n}\delta_I + S_V \tag{36}$$

let us remind that, as mentioned earlier, if h tends to 0, $A \overset{h\to 0}{\to} \delta_I$ and $A\mathbf{V}_I \overset{h\to 0}{\to} \mathbf{V}_I \delta_I$ then (36) is similar to (30) in the limit $h \to 0$ but does not contain the stretching term, and the source term S_V is not defined. In another older work, Morel (1997) had obtained the relation:

$$\frac{\mathrm{d}A}{\mathrm{d}t} = \frac{\mathrm{d}}{\mathrm{d}t} \int_V \delta_I = -\int_{I \cap \partial V} (\mathbf{V}_I \cdot \mathbf{n})\mathbf{n} \cdot \mathbf{N} \frac{\mathrm{sign}(\mathbf{n} \cdot \mathbf{N})}{\sqrt{1 - (\mathbf{n} \cdot \mathbf{N})^2}} \, \mathrm{d}C + \int_{I \cap \partial V} (\mathbf{V}_I \cdot \mathbf{n})\nabla \cdot \mathbf{n} \, \mathrm{d}S + \int_V S_V \, \mathrm{d}V \tag{37}$$

where $\mathbf{N}$ is a unit vector, normal and exterior to the surface ∂V, boundary of the volume V, and $\mathbf{n}$ is the unit vector normal to the interface I. Eq. (37) has been demonstrated to be equivalent to (36) by Lhuillier et al. (2000), but the source term is still not given. Previously, Candel and Poinsot (1990) derived the following equation for the evolution of the fine-grained flame surface density:

$$\phi_S = \frac{1}{\delta_I} \frac{\mathrm{d}}{\mathrm{d}t}(\delta_I) = -\mathbf{n} \otimes \mathbf{n} : \nabla \mathbf{V} + \nabla \cdot \mathbf{V} + S_L \nabla \cdot \mathbf{n}$$

relation which is equivalent to (35), but where a flame front velocity S_L has been introduced. In this work, the division by the surface distribution δ_I seems to be simply a convenient notation and is not used any more in the recent review by Veynante and Vervisch (2002).

Eqs. (30)–(33) is thus similar to (36) but in the latter, all the source terms are fully written. As far as we know, space averaging seems to be the only way to compute completely this source term. As a matter of fact, this term is related to fragmentation and coalescence events which lead to singularities of the surface (cf. Fig. 1). This effect seems to be difficult to obtain by statistical

averaging of the fine-grained density δ_I. Lastly, let us notice that (30)–(33) is completely similar to an equation obtained by Marle (1982), in a different manner, within the context of porous media.

4. Closure relations

In the preceding section, new quantities $(\mathbf{V}_I, S_e, S_{fc}, S_c)$ where introduced which can be rigorously computed if the velocity field on the interface is perfectly known. This however cannot be done for most numerical computations of two-phase flow where the size of fluid blobs can be much smaller than the size of the computing mesh. These quantities are thus, for most computations, unclosed quantities and closures relations are needed. In the following section, acceptable form of the closure relations for $\mathbf{V}_I$ and the interfacial momentum transfer will be determined. Empirical closures for the mean interfacial velocity are $\mathbf{V}_I = \mathbf{V}$ (Vallet and Borghi, 1999) and $\mathbf{V}_I = \phi_1 \mathbf{V}_2 + \phi_2 \mathbf{V}_1$ (Lhuillier, 2004) which was introduced in order to recover Ishii (1975)'s first results that interfacial velocity should be equal to the dispersed phase velocity in the case of diluted mixtures. In the present work, closures relations will be obtained by writing down the balance of entropy and then by writing that the entropy production is positive. Actually, the entropy is *constructed* in such a way to have a positive production term. This method has been often used as a tool for obtaining "equivalent medium" equations, or homogenized equations. Without being exhaustive, Marle (1982) is a good reference for porous medium. In order to cope with non-thermodynamic equilibrium situation, different kind of thermodynamics can be used mainly the EIT (extended irreversible thermodynamic) developed in Jou et al. (1996) and the RET (rational extended thermodynamic) developed in Müller et al. (1998). In these theories, the space of the state variables upon which the entropy may depend, is enlarged. Particularly dissipative fluxes can be introduced. We will consider at the end of this section a consequence of using such extended entropies. Calculations will be undertaken using EIT (which leads to simpler calculations than RET and is equivalent to RET in the linear approximation); but the entropy that is being used can also be considered, as in RET, as a mathematical entropy ensuring that the resulting hyperbolic system of conservation law is mathematically well posed.

4.1. Entropy balance

Generally the entropy balance equation is written:

$$\rho \frac{\mathrm{d}s}{\mathrm{d}t} = -\nabla \cdot \mathbf{J}_s + \sigma_s \tag{38}$$

$\mathbf{J}_s$ is the entropy flux and σ_s is the entropy production. The total entropy can be constructed from the specific entropy of each separate phase and in the case without interfacial concentration one can consider:

$$s = \sum_{k=1}^{2} c_k s_k \quad \text{where } c_k = \frac{\phi_k \rho_k}{\rho} \tag{39}$$

Functions s_k (extended partial entropies) will depend a priori upon internal energies e_k, specific volumes $1/\rho_k$ and mass fractions c_k.

There are several ways to introduce the entropy dependency with the interfacial area concentration. We will firstly suppose that the entropy is defined by:

$$s = \sum_{k=1}^{2} c_k s_k + \Phi(A) \tag{40}$$

where Φ is a convex function of A. Here, the interface is considered as a third medium different from both fluids. Its main advantage comes from the ease of introducing interfacial chemical reactions. Moreover identification of equilibrium surface tension is rather straightforward. Rigorously, one should write down all the necessary balance equations (mass, momentum and energy) for the interfacial medium, this is Marle's original point of view. Here it will be assumed that the mass of this third medium is infinitesimally small, thus all the balance equations written so far are sufficient to describe the evolution of the mixture.

In the second point of view, the interfacial area is considered as an internal variable for each phase "which means that in practice, these variables are measurable but not controllable..." (Maugin and Muschik, 1994). Then, the entropy will be written using (39). And the associated Gibbs-like relation will be written:

$$\Theta_k \, \mathrm{d}s_k = \mathrm{d}e_k + \Pi_k \mathrm{d}\left(\frac{1}{\rho_k}\right) - M_k \, \mathrm{d}c_k + \Theta_k \frac{\partial s_k}{\partial p_i} \, \mathrm{d}p_i \tag{41}$$

where $\Theta_k = \frac{\partial e_k}{\partial s_k}$, $\Pi_k = -\Theta_k \rho_k^2 \frac{\partial s_k}{\partial \rho_k}$, $M_k = -\Theta_k \frac{\partial s_k}{\partial c_k}$ and the p_i are the internal variables (here, interfacial area) not to be confounded with pressure; the variables Θ_k, Π_k and M_k are non-equilibrium variables that we will suppose equal to their equilibrium values mainly T_k for the temperature, P_k for the pressure and μ_k for chemical potential. As demonstrated in Jou et al. (1996) this corresponds to a first order expansion. Then (41) takes the following form:

$$T_k \, \mathrm{d}s_k = \mathrm{d}e_k + P_k \mathrm{d}\left(\frac{1}{\rho_k}\right) - \mu_k \, \mathrm{d}c_k + T_k \frac{\partial s_k}{\partial p_i} \, \mathrm{d}p_i \tag{42}$$

4.1.1. Entropy balance equation with internal variables

Let us derive the entropy balance equation in the general context of irreversible thermodynamics with internal variables. The derivation of (39) leads to:

$$\frac{\mathrm{d}s}{\mathrm{d}t} = \sum_{k=1}^{2} \left(\frac{\mathrm{d}^{(k)}c_k}{\mathrm{d}t} s_k + \frac{\mathrm{d}^{(k)}s_k}{\mathrm{d}t} c_k - s_k \mathbf{V}_k^{\mathrm{dr}} \cdot \nabla c_k - c_k \mathbf{V}_k^{\mathrm{dr}} \cdot \nabla s_k \right) \tag{43}$$

In (43) $\frac{\mathrm{d}^{(k)}}{\mathrm{d}t} = \frac{\partial}{\partial t} + \mathbf{V}_k \cdot \nabla$ is the derivative along the motion of phase k and $\mathbf{V}_k^{\mathrm{dr}}$ is the drift velocity, defined by:

$$\mathbf{V}_k^{\mathrm{dr}} = \mathbf{V}_k - \mathbf{V} \tag{44}$$

The relation between the two derivatives is:

$$\frac{\mathrm{d}^{(k)}}{\mathrm{d}t} = \frac{\mathrm{d}}{\mathrm{d}t} + (\mathbf{V}_k - \mathbf{V}) \cdot \nabla = \frac{\mathrm{d}}{\mathrm{d}t} + \mathbf{V}_k^{\mathrm{dr}} \cdot \nabla \tag{45}$$

In order to calculate the entropy derivative in (43) we need to compute the derivative of partial entropies along the flow of the phase k. It will be done using relation (42):

$$T_k \frac{\mathrm{d}^{(k)} s_k}{\mathrm{d}t} = \frac{\mathrm{d}^{(k)} e_k}{\mathrm{d}t} - \frac{P_k}{\rho_k^2} \frac{\mathrm{d}^{(k)} \rho_k}{\mathrm{d}t} - \mu_k \frac{\mathrm{d}^{(k)} c_k}{\mathrm{d}t} + T_k \frac{\partial s_k}{\partial p_i} \frac{\mathrm{d}^{(k)} p_i}{\mathrm{d}t} \tag{46}$$

After introducing the mass fraction one obtains:

$$c_k \frac{\mathrm{d}^{(k)} s_k}{\mathrm{d}t} = \frac{\phi_k \rho_k}{T_k \rho} \frac{\mathrm{d}^{(k)} e_k}{\mathrm{d}t} - \frac{\phi_k \rho_k}{T_k \rho} \frac{P_k}{\rho_k^2} \frac{\mathrm{d}^{(k)} \rho_k}{\mathrm{d}t} - \frac{\phi_k \rho_k}{T_k \rho} \mu_k \frac{\mathrm{d}^{(k)} c_k}{\mathrm{d}t} + \frac{\phi_k \rho_k}{\rho} \frac{\partial s_k}{\partial p_i} \frac{\mathrm{d}^{(k)} p_i}{\mathrm{d}t} \tag{47}$$

In (47) the following derivatives are needed: $\frac{\mathrm{d}^{(k)} e_k}{\mathrm{d}t}, \frac{\mathrm{d}^{(k)} \rho_k}{\mathrm{d}t}, \frac{\mathrm{d}^{(k)} c_k}{\mathrm{d}t}$. The variation of internal energy is obtained substituting kinetic energy obtained from balance of momentum (16) in the energy balance (20), after some algebra one obtains:

$$\phi_k \rho_k \frac{\mathrm{d}^{(k)} e_k}{\mathrm{d}t} + \nabla \cdot \mathbf{q}_k = -P_k \frac{\partial \phi_k}{\partial t} + X_k + \phi_k \boldsymbol{\sigma}_k : \nabla \mathbf{V}_k - \mathbf{F}_{Ik} \cdot \mathbf{V}_k \tag{48}$$

The variation of density is:

$$\frac{1}{\rho_k} \frac{\mathrm{d}^{(k)} \rho_k}{\mathrm{d}t} = -\frac{1}{\phi_k} (\omega_k + \nabla \cdot \phi_k (\mathbf{V}_k - \mathbf{U}_k)) \tag{49}$$

And the variation of mass fraction is given by:

$$\frac{\mathrm{d}^{(k)} c_k}{\mathrm{d}t} = -\frac{1}{\rho} \nabla \cdot \left(\phi_k \rho_k \mathbf{V}_k^{\mathrm{dr}} \right) + \mathbf{V}_k^{\mathrm{dr}} \cdot \nabla c_k \tag{50}$$

The variation of the parameters p_i will be discussed in a special section. Once (48)–(50) are substituted in (47), one obtains after calculation:

$$\begin{aligned}
\rho c_k \frac{\mathrm{d}^{(k)} s_k}{\mathrm{d}t} = {} & -\frac{1}{T_k} \nabla \cdot \mathbf{q}_k - \frac{1}{T_k} P_k \frac{\partial \phi_k}{\partial t} + \frac{1}{T_k} X_k + \frac{1}{T_k} \phi_k \boldsymbol{\sigma}_k : \nabla \mathbf{V}_k - \frac{1}{T_k} \mathbf{F}_{Ik} \cdot \mathbf{V}_k \\
& + \frac{P_k}{T_k} (\omega_k + \nabla \cdot \phi_k (\mathbf{V}_k - \mathbf{U}_k)) + \frac{\mu_k}{T_k} \nabla \cdot \left(c_k \phi_k \rho_k \mathbf{V}_k^{\mathrm{dr}} \right) \\
& - 2 \frac{1}{T_k} \mu_k \phi_k \rho_k \mathbf{V}_k^{\mathrm{dr}} \cdot \nabla c_k + \rho c_k \frac{\partial s_k}{\partial p_i} \frac{\mathrm{d}^{(k)} p_i}{\mathrm{d}t}
\end{aligned} \tag{51}$$

For convenience and since the final form of entropy variation (38) is desired, relation (51) will be put in the following form:

$$\begin{aligned}
\rho c_k \frac{\mathrm{d}^{(k)} s_k}{\mathrm{d}t} & = -\nabla \cdot \mathbf{J}_s^{(k)} + \sigma_s^{(k)} + \rho c_k \frac{\partial s_k}{\partial p_i} \frac{\mathrm{d}^{(k)} p_i}{\mathrm{d}t} \\
& = -\nabla \cdot \mathbf{J}_s^{(k)} + \sigma_s^{0,(k)} + \sigma_s^{1,(k)} + \sigma_s^{2,(k)} + \rho c_k \frac{\partial s_k}{\partial p_i} \frac{\mathrm{d}^{(k)} p_i}{\mathrm{d}t}
\end{aligned} \tag{52}$$

In relation (52), the following definitions have been used:

$$\mathbf{J}_s^{(k)} = \frac{1}{T_k} \left(\mathbf{q}_k + \phi_k P_k \mathbf{V}_k - P_k \phi_k (\mathbf{V}_k - \mathbf{U}_k) - c_k \mu_k \phi_k \rho_k \mathbf{V}_k^{\mathrm{dr}} \right) \tag{53}$$

$$\sigma_s^{0,(k)} = -\frac{P_k}{T_k} \frac{\partial \phi_k}{\partial t} + \frac{X_k}{T_k} + \frac{P_k \omega_k}{T_k} \tag{54}$$

$$\begin{aligned}
\sigma_s^{1,(k)} = {} & \left(\mathbf{q}_k + P_k \phi_k \mathbf{U}_k - \phi_k \rho_k c_k \mu_k \mathbf{V}_k^{\mathrm{dr}} \right) \cdot \nabla \frac{1}{T_k} \\
& - \frac{1}{T_k} \mathbf{V}_k \cdot (\mathbf{F}_{Ik} - \nabla \phi_k P_k) - \frac{1}{T_k} \phi_k (\mathbf{V}_k - \mathbf{U}_k) \cdot \nabla P_k \\
& - \frac{1}{T_k} \phi_k \rho_k c_k \mathbf{V}_k^{\mathrm{dr}} \cdot \nabla \mu_k - 2 \frac{1}{T_k} \mu_k \phi_k \rho_k \mathbf{V}_k^{\mathrm{dr}} \cdot \nabla c_k
\end{aligned} \tag{55}$$

$$\sigma_s^{2,(k)} = \phi_k \frac{\overset{\circ}{\boldsymbol{\sigma}}_k : \overset{\circ}{\widehat{\nabla \mathbf{V}_k}}}{T_k} \tag{56}$$

where the stress and strain rate tensors have been split in spherical and deviatoric part: (De Groot and Mazur, 1962)

$$\nabla \mathbf{V}_k = \frac{1}{3} \nabla \cdot (\mathbf{V}_k) \mathbf{1} + \overset{\circ}{\widehat{\nabla \mathbf{V}}}_k$$

$$\boldsymbol{\sigma}_k = -P_k \mathbf{1} + \overset{\circ}{\boldsymbol{\sigma}}_k$$

The contracted product of these two tensors now reads:

$$\boldsymbol{\sigma}_k : \nabla \mathbf{V}_k = -P_k \nabla \cdot (\mathbf{V}_k) + \overset{\circ}{\boldsymbol{\sigma}}_k : \overset{\circ}{\widehat{\nabla \mathbf{V}}}_k \tag{57}$$

One can notice that the first upper index of the entropy source terms in (53)–(56) indicates the tensorial order of the quantities taken into account. They sum in the following way:

$$\sigma_s^{(k)} = \sigma_s^{0,(k)} + \sigma_s^{1,(k)} + \sigma_s^{2,(k)} \tag{58}$$

There are different possibilities in the definition of the entropy flux and source terms. Actually, it is either possible to write $\phi_k P_k \nabla \cdot (\mathbf{V}_k)$ or $\nabla \cdot (\phi_k P_k \mathbf{V}_k) - \mathbf{V}_k \cdot \nabla(\phi_k P_k)$. It is somewhat a matter of choice and the latter case has been chosen here. The consequences of this choice will be analyzed further in this paper.

Substituting relations (52) in relation (43), using notations (53)–(56) one obtains:

$$\rho \frac{\mathrm{d}s}{\mathrm{d}t} = \sum_{k=1}^{2} (-\nabla \cdot (\mathbf{J}_s^{(k)} - \rho c_k s_k \mathbf{V}_k^{\mathrm{dr}}) + \sigma_s^{(k)}) + \sum_{k=1}^{2} \rho c_k \frac{\partial s_k}{\partial p_i} \frac{\mathrm{d}^{(k)} p_i}{\mathrm{d}t} \tag{59}$$

Relation (59) will be put in the form (38), that is:

$$\rho \frac{\mathrm{d}s}{\mathrm{d}t} = -\nabla \cdot \mathbf{J}_s + \sigma_s + \sum_{k=1}^{2} \rho c_k \frac{\partial s_k}{\partial p_i} \frac{\mathrm{d}^{(k)} p_i}{\mathrm{d}t} \tag{60}$$

4.2. Entropy source and flux due to the interfacial area concentration variation

4.2.1. Interfacial area as a property of a third medium

Here, entropy is defined by (40) and p_i is set equal to A in (42). Therefore:

$$\rho \frac{\mathrm{d}s}{\mathrm{d}t} = \rho \sum_{k=1}^{2} \frac{\mathrm{d}c_k s_k}{\mathrm{d}t} + \rho \frac{\partial \Phi}{\partial A} \frac{\mathrm{d}A}{\mathrm{d}t} \tag{61}$$

Let us define T_i the temperature of the interface by $\rho \frac{\partial \Phi}{\partial A} = \frac{\gamma}{T_i}$ where γ can be considered as a surface tension between both fluids. Among the different ways of writing sources and fluxes of entropy, one can choose:

$$\mathbf{J}_s^I = \mathbf{0} \tag{62}$$

$$\boldsymbol{\sigma}_s^I = \frac{\gamma}{T_i} (S - A\nabla \cdot \mathbf{V}_I + (\mathbf{V} - \mathbf{V}_I) \cdot \nabla A) \tag{63}$$

Let us consider that each fluid is incompressible, that the heat flux is null, the interfacial coefficient is constant and that there is no chemical reactions, so that chemical potentials are constant. Moreover, as it is often assumed, let us assume that there is only one pressure and one temperature so that $T_i = T$. The fluxes and entropy sources simplify in:

$$\mathbf{J}_s = \sum_{k=1}^{2} \frac{\phi_k P}{T} \mathbf{V}_k + \left(s_k - \frac{c_k}{T} \mu_k \right) \phi_k \rho_k \mathbf{V}_k^{\mathrm{dr}} \tag{64}$$

$$\sigma_s^0 = \sum_{k=1}^{2} \left(-\frac{P}{T} \frac{\partial \phi_k}{\partial t} + \frac{X_k}{T} \right) + \frac{\gamma}{T} (S - A\nabla \cdot \mathbf{V}_I), \tag{65}$$

$$\sigma_s^1 = \sum_{k=1}^{2} \left\{ -\frac{1}{T} \mathbf{V}_k \cdot (\mathbf{F}_{Ik} - \nabla \phi_k P) - 2 \frac{1}{T} \mu_k \phi_k \rho_k \mathbf{V}_k^{\mathrm{dr}} . \nabla c_k \right\} + \frac{\gamma}{T} (\mathbf{V} - \mathbf{V}_I) . \nabla A, \tag{66}$$

$$\sigma_s^2 = \sum_{k=1}^{2} \frac{\phi_k \overset{\circ}{\boldsymbol{\sigma}}_k : \overset{\frown}{\overset{\circ}{\nabla \mathbf{V}_k}}}{T} \tag{67}$$

Considering the first order tensorial terms (66), writing all the coupling in linear approximation (thermodynamic forces are supposed to be proportional to fluxes), one gets.

$$\mathbf{F}_k = \mathbf{F}_{Ik} - \nabla(\phi_k P) = \sum_{p=1}^{2} \left(-\mathbf{L}_{\mathbf{F}_k \mathbf{V}_p} \mathbf{V}_p - \mathbf{L}_{\mathbf{F}_k \nabla c_p} 2\mu_p \phi_p \rho_p \nabla c_p \right) + \mathbf{L}_{\mathbf{F}_k \nabla A} \frac{\gamma}{T} \nabla A \tag{68}$$

$$\mathbf{V}_k^{\mathrm{dr}} = \sum_{p=1}^{2} \left(-\mathbf{L}_{\mathbf{V}_k^{\mathrm{dr}} \mathbf{V}_p} \mathbf{V}_p - \mathbf{L}_{\mathbf{V}_k^{\mathrm{dr}} \nabla c_p} 2\mu_p \phi_p \rho_p \nabla c_p \right) + \frac{\gamma}{T} \mathbf{L}_{\mathbf{V}_k^{\mathrm{dr}} \nabla A} \nabla A \tag{69}$$

$$\mathbf{V}_I^{\mathrm{dr}} = \sum_{p=1}^{2} \left(\mathbf{L}_{\mathbf{V}_I^{\mathrm{dr}} \mathbf{V}_p} \mathbf{V}_p + \mathbf{L}_{\mathbf{V}_I^{\mathrm{dr}} \nabla c_p} 2\mu_p \phi_p \rho_p \nabla c_p \right) + \frac{\gamma}{T} \mathbf{L}_{\mathbf{V}_I^{\mathrm{dr}} \nabla A} \nabla A \tag{70}$$

One can notice that the contribution $\nabla(\phi_k P)$ has been subtracted to the interaction force. The appearance of this term is related to the choice of the entropy flux and source terms previously mentioned. However, this is a standard contribution which can also be obtained for instance, through ensemble averaging of microscopic two-phase flow equation (Chen et al., 1996, thought it is a two pressures model).

The most general linear closure relations for this problem can be summarized in matrix notation by:

$$
\begin{pmatrix}
\mathbf{F}_1 \\
\mathbf{F}_2 \\
\mathbf{V}_1^{dr} \\
\mathbf{V}_2^{dr} \\
\mathbf{V}_I^{dr}
\end{pmatrix}
=
\begin{pmatrix}
-\mathbf{L}_{\mathbf{F}_1\mathbf{V}_1} & -\mathbf{L}_{\mathbf{F}_1\mathbf{V}_2} & -\mathbf{L}_{\mathbf{F}_1\nabla c_1} & -\mathbf{L}_{\mathbf{F}_1\nabla c_2} & \mathbf{L}_{\mathbf{F}_1\nabla A} \\
-\mathbf{L}_{\mathbf{F}_2\mathbf{V}_1} & -\mathbf{L}_{\mathbf{F}_2\mathbf{V}_2} & -\mathbf{L}_{\mathbf{F}_2\nabla c_1} & -\mathbf{L}_{\mathbf{F}_2\nabla c_2} & \mathbf{L}_{\mathbf{F}_2\nabla A} \\
-\mathbf{L}_{\mathbf{V}_1^{dr}\mathbf{V}_1} & -\mathbf{L}_{\mathbf{V}_1^{dr}\mathbf{V}_2} & -\mathbf{L}_{\mathbf{V}_1^{dr}\nabla c_1} & -\mathbf{L}_{\mathbf{V}_1^{dr}\nabla c_2} & \mathbf{L}_{\mathbf{V}_1^{dr}\nabla A} \\
-\mathbf{L}_{\mathbf{V}_2^{dr}\mathbf{V}_1} & -\mathbf{L}_{\mathbf{V}_2^{dr}\mathbf{V}_2} & -\mathbf{L}_{\mathbf{V}_2^{dr}\nabla c_1} & -\mathbf{L}_{\mathbf{V}_2^{dr}\nabla c_2} & \mathbf{L}_{\mathbf{V}_2^{dr}\nabla A} \\
\mathbf{L}_{\mathbf{V}_I^{dr}\mathbf{V}_1} & \mathbf{L}_{\mathbf{V}_I^{dr}\mathbf{V}_2} & \mathbf{L}_{\mathbf{V}_I^{dr}\nabla c_1} & \mathbf{L}_{\mathbf{V}_I^{dr}\nabla c_2} & -\mathbf{L}_{\mathbf{V}_I^{dr}\nabla A}
\end{pmatrix}
\cdot
\begin{pmatrix}
\mathbf{V}_1 \\
\mathbf{V}_2 \\
2\mu_1\rho_1\phi_1\nabla c_1 \\
2\mu_2\rho_2\phi_2\nabla c_2 \\
\frac{\gamma}{T}\nabla A
\end{pmatrix}
\tag{71}
$$

In order to ensure a positive entropy production term the matrix in (71) has to be definite positive and the Onsager reciprocity relations imply that this matrix is symmetric, that is:

$$\mathbf{L}_{\mathbf{F}_p\mathbf{V}_q} = {}^t\mathbf{L}_{\mathbf{F}_q\mathbf{V}_p} \tag{72}$$

$$\mathbf{L}_{\mathbf{V}_p^{dr}\mathbf{V}_k} = {}^t\mathbf{L}_{\mathbf{F}_k\nabla c_p} \tag{73}$$

$$\mathbf{L}_{\mathbf{V}_p^{dr}\nabla c_q} = {}^t\mathbf{L}_{\mathbf{V}_q^{dr}\nabla c_p} \tag{74}$$

$$\mathbf{L}_{\mathbf{V}_I^{dr}\mathbf{V}_p} = {}^t\mathbf{L}_{\mathbf{F}_p\nabla\rho} \tag{75}$$

$$\mathbf{L}_{\mathbf{V}_I^{dr}\nabla c_p} = {}^t\mathbf{L}_{\mathbf{V}_p^{dr}\nabla A} \tag{76}$$

Moreover, if one assumes that the interaction force between phases is Galilean invariant, the following relations must hold:

$$\mathbf{L}_{\mathbf{F}_q\mathbf{V}_q} = -\mathbf{L}_{\mathbf{F}_q\mathbf{V}_p}, \quad p \neq q \tag{77}$$

The interfacial velocity $\mathbf{V}_I$ is thus related to the velocity of the different phases and to the gradient of the interfacial area concentration in the following (yet undetermined) way:

$$\mathbf{V}_I^{dr} = \mathbf{L}_{\mathbf{V}_I^{dr}\mathbf{V}_1}\mathbf{V}_1 + \mathbf{L}_{\mathbf{V}_I^{dr}\mathbf{V}_2}\mathbf{V}_2 + 2\mu_1\rho_1\phi_1\mathbf{L}_{\mathbf{V}_I^{dr}\nabla c_1}\nabla c_1 + 2\mu_2\rho_2\phi_2\mathbf{L}_{\mathbf{V}_I^{dr}\nabla c_2}\nabla c_2 - \mathbf{L}_{\mathbf{V}_I^{dr}\nabla A}\frac{\gamma}{T}\nabla A \tag{78}$$

Indeed some of the effects in the preceding relations may not really be coupled, and suppressing some of the coupling can lead to the following closures:

$$
\begin{pmatrix}
\mathbf{F}_1 \\
\mathbf{F}_2 \\
\mathbf{V}_1^{dr} \\
\mathbf{V}_2^{dr} \\
\mathbf{V}_I^{dr}
\end{pmatrix}
=
\begin{pmatrix}
-\mathbf{L}_{\mathbf{F}_1\mathbf{V}_1} & -\mathbf{L}_{\mathbf{F}_1\mathbf{V}_2} & 0 & 0 & 0 \\
-\mathbf{L}_{\mathbf{F}_2\mathbf{V}_1} & -\mathbf{L}_{\mathbf{F}_2\mathbf{V}_2} & 0 & 0 & 0 \\
0 & 0 & -\mathbf{L}_{\mathbf{V}_1^{dr}\nabla c_1} & 0 & 0 \\
0 & 0 & 0 & -\mathbf{L}_{\mathbf{V}_2^{dr}\nabla c_2} & 0 \\
0 & 0 & 0 & 0 & -\mathbf{L}_{\mathbf{V}_I^{dr}\nabla A}
\end{pmatrix}
\begin{pmatrix}
\mathbf{V}_1 \\
\mathbf{V}_2 \\
2\mu_1\rho_1\phi_1\nabla c_1 \\
2\mu_2\rho_2\phi_2\nabla c_1 \\
\frac{\gamma}{T}\nabla A
\end{pmatrix}
\tag{79}
$$

We then obtain for the interfacial velocity the following relation:

$$\mathbf{V}_I - \mathbf{V} = -\frac{\gamma}{T}\mathbf{L}_{\mathbf{V}_I^{dr}\nabla A}\nabla A \tag{80}$$

The interfacial velocity is thus equal to the mass averaged velocity plus an interfacial area diffusion term. This term should disappear when the mixture is translating uniformly. Actually, considering $\frac{\gamma}{T}\mathbf{L}_{\mathbf{V}_I^{dr}\nabla A}$ as a turbulent diffusivity, (80) reduces to the closure of Vallet and Borghi (1999).

One can also recover the Lhuillier's (2004) closure $\mathbf{V}_I = \phi_1\mathbf{V}_2 + \phi_2\mathbf{V}_1$ from relation (78), setting:

$$\mathbf{L}_{\mathbf{V}_I^{dr}\mathbf{V}_1} = \frac{\rho_2\phi_2^2 - \rho_1\phi_1^2}{\rho_1\phi_1 + \rho_2\phi_2}\mathbf{1} \quad \text{and} \quad \mathbf{L}_{\mathbf{V}_I^{dr}\mathbf{V}_2} = \frac{\rho_1\phi_1^2 - \rho_2\phi_2^2}{\rho_1\phi_1 + \rho_2\phi_2}\mathbf{1} \tag{81}$$

From (81), it can be noticed that $\mathbf{L}_{\mathbf{V}_I^{dr}\mathbf{V}_1} + \mathbf{L}_{\mathbf{V}_I^{dr}\mathbf{V}_2} = \mathbf{0}$; so that using (71) and Onsager symmetry relations, Newton third law $\mathbf{F}_1 + \mathbf{F}_2 = \mathbf{0}$ is enforced.

One can thus conclude that the main empirical closures are compatible with the present thermodynamic framework.

As for the coupling coefficients $\mathbf{L}_{\mathbf{F}_q\mathbf{V}_q}$, they can be considered as a function of the interfacial area concentration A, which is null if $A = 0$, then $\mathbf{L}_{\mathbf{F}_q\mathbf{V}_q} = A\mathbf{C}_{\mathbf{F}_q\mathbf{V}_q}(T_q, \rho_q, A, \ldots)$, so that:

$$\mathbf{F}_1 = -A\mathbf{C}_{\mathbf{F}_1\mathbf{V}_1}(\mathbf{V}_1 - \mathbf{V}_2) \tag{82}$$
$$\mathbf{F}_2 = -A\mathbf{C}_{\mathbf{F}_2\mathbf{V}_1}(\mathbf{V}_2 - \mathbf{V}_1)$$

4.2.2. Interfacial area as an inner variable

At thermodynamic equilibrium the interfacial coefficient (interfacial free energy) is defined by:

$$\gamma_k = \rho_k T_k \frac{\partial s_k}{\partial A} \tag{83}$$

As we have done for temperature, pressure and chemical potential, we will suppose that the preceding relation holds outside equilibrium. The derivative of the interfacial area concentration along the motion of the kth phase is:

$$\frac{\mathrm{d}^{(k)}A}{\mathrm{d}t} = \mathbf{V}_k \cdot \nabla A - \nabla \cdot (A\mathbf{V}_I) + S \tag{84}$$

then the missing term, giving the contribution of interfacial area concentration in (60) can be written:

$$\sum_{k=1}^{2}\frac{\phi_k\gamma_k}{T_k}\frac{\mathrm{d}^{(k)}A}{\mathrm{d}t} = \left(\sum_{k=1}^{2}\frac{\phi_k\gamma_k}{T_k}\mathbf{V}_k\right)\cdot\nabla A + \left(\sum_{k=1}^{2}\frac{\phi_k\gamma_k}{T_k}\right)(S - \nabla\cdot(A\mathbf{V}_I)) \tag{85}$$

From relation (85), one can identify the interface entropy flux. The decomposition of the right hand side of the entropy variation (38) in divergence of a flux and a source term is not unique, and is again, in some way, a matter of choice. One can choose:

$$\mathbf{J}_s^I = \mathbf{0} \tag{86}$$

and

$$\sigma_s^I = \left(\sum_{k=1}^{2} \frac{\phi_k \gamma_k}{T_k} \mathbf{V}_k\right) \cdot \nabla A - \left(\sum_{k=1}^{2} \frac{\phi_k \gamma_k}{T_k} \mathbf{V}_I\right) \cdot \nabla A + S\left(\sum_{k=1}^{2} \frac{\phi_k \gamma_k}{T_k}\right) + A\left(\sum_{k=1}^{2} \frac{\phi_k \gamma_k}{T_k}\right) \nabla \cdot \mathbf{V}_I$$

(87)

Relation (86) introduces the interfacial free energy (with respect to the vacuum) γ_1 and γ_2 of each fluid separately. It is convenient to introduce their relative interfacial tension. This can be done using Girifalco and Good (1957) model of interfacial tension:

$$\gamma = \gamma_1 + \gamma_2 - 2\phi\sqrt{\gamma_1\gamma_2}$$

(88)

where ϕ is a (new) parameter of order unity introduced to take into account the nature of the intermolecular interactions (inside and between each phase). Another interesting choice, which introduces two parameters, is Fowkes (1962) model of interfacial tension:

$$\gamma = \gamma_1 + \gamma_2 - 2\sqrt{\gamma_1^d \gamma_2^d}$$

(89)

where γ_1^d and γ_2^d are the part of the interfacial free energies γ_1 and γ_2 which come from Debye's dipolar interactions. Using either model, one can define:

$$\alpha = \frac{\sum_{k=1}^{2} \gamma_k \phi_k}{\gamma}$$

(90)

$$\mathbf{U} = \frac{\sum_{k=1}^{2} \gamma_k \phi_k \mathbf{V}_k}{\sum_{k=1}^{2} \gamma_k \phi_k}$$

(91)

Once these relations are substituted in (86) and considering a one temperature model, we obtain:

$$\sigma_s^I = \frac{\alpha\gamma}{T}(S + \nabla \cdot \mathbf{V}_I) + \frac{\gamma\alpha}{T}(\mathbf{U} - \mathbf{V}_I) \cdot \nabla A$$

(92)

which leads to:

$$\mathbf{J}_s = \sum_{k=1}^{2} \frac{\phi_k P}{T} \mathbf{V}_k + \left(s_k - \frac{c_k}{T}\mu_k\right)\phi_k \rho_k \mathbf{V}_k^{\mathrm{dr}}$$

(93)

$$\sigma_s^0 = \sum_{k=1}^{2} \left(-\frac{P}{T}\frac{\partial \phi_k}{\partial t} + \frac{X_k}{T}\right) + \frac{\alpha\gamma}{T}(S + \nabla \cdot \mathbf{V}_I)$$

(94)

$$\sigma_s^1 = \sum_{k=1}^{2} \left\{-\frac{1}{T}\mathbf{V}_k \cdot (\mathbf{F}_{Ik} - \nabla\phi_k P) - 2\frac{1}{T}\mu_k \phi_k \rho_k \mathbf{V}_k^{\mathrm{dr}} \cdot \nabla c_k\right\} + \frac{\gamma\alpha}{T}(\mathbf{U} - \mathbf{V}_I) \cdot \nabla A$$

(95)

$$\sigma_s^2 = \sum_{k=1}^{2} \frac{\phi_k \overset{\circ}{\boldsymbol{\sigma}}_k : \overset{\circ}{\overbrace{\nabla\mathbf{V}}}_k}{T}$$

(96)

Now let us again concentrate on entropy sources terms due to first order tensors. Considering that thermodynamic forces are proportional to fluxes, one can write:

$$\mathbf{F}_k = \mathbf{F}_{Ik} - \nabla(\phi_k P) = \sum_{p=1}^{2} -\mathbf{L}_{\mathbf{F}_k \mathbf{V}_p} \mathbf{V}_p - \mathbf{L}_{\mathbf{F}_k \nabla c_p} 2\mu_p \phi_p \rho_p \nabla c_p + \mathbf{L}_{\mathbf{F}_k \nabla A} \frac{\gamma}{T} \nabla A \tag{97}$$

$$\mathbf{V}_k^{dr} = \sum_{p=1}^{2} -\mathbf{L}_{\mathbf{V}_k^{dr} \mathbf{V}_p} \mathbf{V}_p - \mathbf{L}_{\mathbf{V}_k^{dr} \nabla c_p} 2\mu_p \phi_p \rho_p \nabla c_p + \frac{\gamma}{T} \mathbf{L}_{\mathbf{V}_k^{dr} \nabla A} \nabla A \tag{98}$$

$$\mathbf{U} - \mathbf{V}_I = \sum_{p=1}^{2} \mathbf{L}_{\mathbf{V}_I \mathbf{V}_p} \mathbf{V}_p + \mathbf{L}_{\mathbf{V}_I \nabla c_p} 2\mu_p \phi_p \rho_p \nabla c_p + \alpha \frac{\gamma}{T} \mathbf{L}_{\mathbf{V}_I \nabla A} \nabla A \tag{99}$$

Therefore:

$$
\begin{pmatrix} \mathbf{F}_1 \\ \mathbf{F}_2 \\ \mathbf{V}_1^{dr} \\ \mathbf{V}_2^{dr} \\ \mathbf{U} - \mathbf{V}_I \end{pmatrix}
=
\begin{pmatrix}
-\mathbf{L}_{\mathbf{F}_1 \mathbf{V}_1} & -\mathbf{L}_{\mathbf{F}_1 \mathbf{V}_2} & -\mathbf{L}_{\mathbf{F}_1 \nabla c_1} & -\mathbf{L}_{\mathbf{F}_1 \nabla c_2} & \mathbf{L}_{\mathbf{F}_1 \nabla A} \\
-\mathbf{L}_{\mathbf{F}_2 \mathbf{V}_1} & -\mathbf{L}_{\mathbf{F}_2 \mathbf{V}_2} & -\mathbf{L}_{\mathbf{F}_2 \nabla c_1} & -\mathbf{L}_{\mathbf{F}_2 \nabla c_2} & \mathbf{L}_{\mathbf{F}_2 \nabla A} \\
-\mathbf{L}_{\mathbf{V}_1^{dr} \mathbf{V}_1} & -\mathbf{L}_{\mathbf{V}_1^{dr} \mathbf{V}_2} & -\mathbf{L}_{\mathbf{V}_1^{dr} \nabla c_1} & -\mathbf{L}_{\mathbf{V}_1^{dr} \nabla c_2} & \mathbf{L}_{\mathbf{V}_1^{dr} \nabla A} \\
-\mathbf{L}_{\mathbf{V}_2^{dr} \mathbf{V}_1} & -\mathbf{L}_{\mathbf{V}_2^{dr} \mathbf{V}_2} & -\mathbf{L}_{\mathbf{V}_2^{dr} \nabla c_1} & -\mathbf{L}_{\mathbf{V}_2^{dr} \nabla c_2} & \mathbf{L}_{\mathbf{V}_2^{dr} \nabla A} \\
\mathbf{L}_{\mathbf{V}_I \mathbf{V}_1} & \mathbf{L}_{\mathbf{V}_I \mathbf{V}_2} & \mathbf{L}_{\mathbf{V}_I \nabla c_1} & \mathbf{L}_{\mathbf{V}_I \nabla c_2} & \mathbf{L}_{\mathbf{V}_I \nabla A}
\end{pmatrix}
\cdot
\begin{pmatrix} \mathbf{V}_1 \\ \mathbf{V}_2 \\ 2\mu_1 \rho_1 \phi_1 \nabla c_1 \\ 2\mu_2 \rho_2 \phi_2 \nabla c_2 \\ \alpha \frac{\gamma}{T} \nabla A \end{pmatrix}
\tag{100}
$$

Then, again, the entropy production term is non-negative if the matrix in (100) is definite positive and the Onsager reciprocity relations imply that it shall be symmetric. Moreover, if one assumes that the interaction force between phases is Galilean invariant, the following relations must also hold:

$$\mathbf{L}_{\mathbf{F}_q \mathbf{V}_q} = -\mathbf{L}_{\mathbf{F}_q \mathbf{V}_p}, \quad p \neq q \tag{101}$$

The interfacial velocity $\mathbf{V}_I$ is thus related to the velocity of the different phases and to the gradient of the interfacial area concentration in the following way:

$$\mathbf{U} - \mathbf{V}_I = \mathbf{L}_{\mathbf{V}_I \mathbf{V}_1} \mathbf{V}_1 + \mathbf{L}_{\mathbf{V}_I \mathbf{V}_2} \mathbf{V}_2 + 2\mu_1 \rho_1 \phi_1 \mathbf{L}_{\mathbf{V}_I \nabla c_1} \nabla c_1 + 2\mu_2 \rho_2 \phi_2 \mathbf{L}_{\mathbf{V}_I \nabla c_2} \nabla c_2 + \mathbf{L}_{\mathbf{V}_I \nabla A} \frac{\gamma}{T} \alpha \nabla A \tag{102}$$

After suppressing some of the coupling, one gets:

$$
\begin{pmatrix} \mathbf{F}_1 \\ \mathbf{F}_2 \\ \mathbf{V}_1^{dr} \\ \mathbf{V}_2^{dr} \\ \mathbf{U} - \mathbf{V}_I \end{pmatrix}
=
\begin{pmatrix}
-\mathbf{L}_{\mathbf{F}_1 \mathbf{V}_1} & -\mathbf{L}_{\mathbf{F}_1 \mathbf{V}_2} & 0 & 0 & 0 \\
-\mathbf{L}_{\mathbf{F}_2 \mathbf{V}_1} & -\mathbf{L}_{\mathbf{F}_2 \mathbf{V}_2} & 0 & 0 & 0 \\
0 & 0 & -\mathbf{L}_{\mathbf{V}_1^{dr} \nabla c_1} & 0 & 0 \\
0 & 0 & 0 & -\mathbf{L}_{\mathbf{V}_2^{drs} \nabla c_2} & 0 \\
0 & 0 & 0 & 0 & \mathbf{L}_{\mathbf{V}_I \nabla A}
\end{pmatrix}
\cdot
\begin{pmatrix} \mathbf{V}_1 \\ \mathbf{V}_2 \\ 2\mu_1 \rho_1 \phi_1 \nabla c_1 \\ 2\mu_2 \rho_2 \phi_2 \nabla c_1 \\ \alpha \frac{\gamma}{T} \nabla A \end{pmatrix}
\tag{103}
$$

The following relation for the interfacial velocity is obtained:

$$\mathbf{V}_I = \mathbf{U} - \frac{\gamma}{T} \alpha \mathbf{L}_{\mathbf{V}_I \nabla A} \nabla A \tag{104}$$

the involved velocity $\mathbf{U}$ is an average of the phases velocities weighted by the interfacial free energies instead of the densities. The two closures (80) and (104), although they are similar, provide different results for a mixing of fluids with different density ratio and surface energy ratio. For instance, using data from Girifalco and Good (1957), the ratio of the density of mercury and water is 13.6 whereas the ratio of surface energies is 6.6 (for hexane and water the ratios are 0.66 and 0.25).

As in Section 4.2.1, one can recover Lhuillier's (2004) closure. A simple computation shows that the following form for the different coefficients is needed:

$$\mathbf{L_{V_IV_1}} = \left(\frac{\gamma_1\phi_1}{\gamma_1\phi_1 + \gamma_2\phi_2} - \phi_2 \right)\mathbf{1} \text{ and } \mathbf{L_{V_IV_2}} = \left(\frac{\gamma_2\phi_2}{\gamma_1\phi_1 + \gamma_2\phi_2} - \phi_1 \right)\mathbf{1} \tag{105}$$

Moreover, Newton third law is also verified and relations similar to (82) are obtained.

4.3. Non-linear and extended thermodynamic closures

As mentioned earlier, extended irreversible thermodynamic consists in introducing the dissipative fluxes in the entropy. We will consider, in this section, consequences of this assumption and extend the preceding closure (82). We will, for sake of simplicity, apply this formalism only on the interfacial forces. Surprisingly the dissipative fluxes are here the forces and the following derivation will mimic the one of Jou et al. (1996) for viscoelastic rheological law, and the ones of Séro-Guillaume et al. (2002), for porous media. Let us consider that the internal parameter is the interaction force so that:

$$\rho_k\phi_k\frac{\partial s_k}{\partial p_i}\frac{\mathrm{d}^{(k)}p_i}{\mathrm{d}t} = \rho_k\phi_k\frac{\partial s_k}{\partial \mathbf{F}_i}\frac{\mathrm{d}^{(k)}\mathbf{F}_i}{\mathrm{d}t} = \frac{\rho_k\phi_k}{T}\mathbf{V}_{si}\frac{\mathrm{d}^{(k)}\mathbf{F}_i}{\mathrm{d}t} \tag{106}$$

In (106) we have defined the "thermodynamic conjugate" velocity V_{si} by $\mathbf{V}_{si} = T\frac{\partial s}{\partial \mathbf{F}_i}$. This velocity can be considered as a functional of the real velocity $\mathbf{V}_i$, and it is null if $\mathbf{V}_i \equiv 0$. Then we can set $\mathbf{V}_{si} = \tau(A, T, \ldots)\mathbf{V}_i$, τ being a priori a second order tensor. With this definition the entropy production due to first order tensors is:

$$\sigma_s^1 = \sum_{k=1}^{2} \left\{ -\frac{1}{T}\mathbf{V}_k \cdot \left(\tau \cdot \frac{\mathrm{d}^{(k)}\mathbf{F}_k}{\mathrm{d}t} + \mathbf{F}_k \right) - 2\frac{1}{T}\mu_k\phi_k\rho_k\mathbf{V}_k^{\mathrm{dr}}.\nabla c_k \right\} + \frac{\gamma\alpha}{T}(\mathbf{U} - \mathbf{V}_I) \cdot \nabla A \tag{107}$$

If we consider the coupling only with velocities, in a linear hypothesis, using the same arguments than previously, the closure relation for the force is:

$$\tau \cdot \frac{\mathrm{d}^{(k)}\mathbf{F}_k}{\mathrm{d}t} + \mathbf{F}_k = -A\mathbf{L}_{\mathbf{FV}}^{-1} \cdot (\mathbf{V}_k - \mathbf{V}_{p\neq k}) \tag{108}$$

In fact the factor $\tau \cdot \frac{\mathrm{d}^{(k)}\mathbf{F}_k}{\mathrm{d}t} + \mathbf{F}_k$ depends as a functional of the drift velocity and (108) represents a first order expansion of this functional, at second order we could write:

$$\tau \cdot \frac{\mathrm{d}^{(k)}\mathbf{F}_k}{\mathrm{d}t} + \mathbf{F}_k = -A\rho C(Re)|\mathbf{V}_k - \mathbf{V}_{p\neq k}|(\mathbf{V}_k - \mathbf{V}_{p\neq k}) \tag{109}$$

where Re is a Reynolds number which could be related to the slip velocity and the inverse of interfacial area concentration. τ is a tensor which introduces a characteristic time for the vector $\mathbf{F}_k$.

This is a simple way to take into account memory effects akin to the Basset force for instance. However τ remains to be determined.

5. Conclusion

In this paper we have studied a modelling of two-phase flow with interfacial area concentration considered either as a third parameter or as an internal parameter. An exact transport equation for the interfacial area concentration has been obtained. The space averaging method has been used, but let us notice that we could have used averaging in space and time, the results would be the same (Estrada et al., 1980). All the closures of equations have been obtained by a thermodynamic argument. This led to the closure relations (108) and (109) for the interaction force between phases, which include a memory effect and an interfacial area concentration dependence. Moreover, two points of view on the dependency of entropy upon surface density A have led to two different closures for the mean interfacial velocity. Considering the interface as a third medium, we have obtained the relation (78) between the interfacial velocity, the densities averaged velocity, and the gradient of interfacial area concentration. Considering the interface as an inner variable, we obtained the relation (102) similar to (78) but where the velocity average is weighted by the surface energies. With relation (78) we can recover the closure of Vallet and Borghi, whereas both (78) and (102) can provide the Lhuillier's closure. Of course the determination of the coefficients in all closure relations for interfacial velocity should be determined by real experiments or more probably by numerical experiments. Closures for the source term in interfacial equation, mainly the term defined in (31)–(33) have not been addressed in this paper. The closure for zeroth order tensorial terms and second order tensorial terms has not been considered either. Especially for strain–stress relation, it is beyond the scope of this paper, as many efforts have been done to obtain rheological law for two-phase fluid (Brinkman, 1952; Roscoe, 1952; Bataille et al., 1977; Lhuillier, 2003). It can be noticed that extended irreversible thermodynamic could provide a guideline for obtaining viscoelastic law (Jou et al., 1996).

Acknowledgments

The authors would also like to thank the anonymous referees for their constructive criticisms and the relevance of their remarks.

Appendix A

Lemma 1. *Let $\partial\Omega$ be a C^2 surface and f a $C^1(\mathbb{R}^3, \mathbb{R})$ function. Let τ be a smooth vector field whose gradient will be written $\nabla\tau$, then the following integration formula hold:*

$$\int_{\partial\Omega}\{\boldsymbol{\tau}\cdot\nabla f+f(\mathrm{div}\,\boldsymbol{\tau}-\nabla\boldsymbol{\tau}:\ \mathbf{n}\otimes\mathbf{n})\}\,\mathrm{d}s=\int_{\partial\Omega}\left\{\left(\frac{\partial f}{\partial\mathbf{n}}+Cf\right)\boldsymbol{\tau}\cdot\mathbf{n}\right\}\mathrm{d}s+\int_{\Gamma}f\boldsymbol{\tau}_s\cdot\mathbf{n}_v\,\mathrm{d}l$$

where C is the mean curvature, $\boldsymbol{\tau}$ is the projection of $\boldsymbol{\tau}$ on the tangent plane to $\partial\Omega$, and $\mathbf{n}_v$ is the unitary vector normal to the edge Γ of $\partial\Omega$ (Fig. A.1).

Demonstration:

Let Δ be an open neighbourhood of $\mathbb{R}^2$ such that $\partial\Omega$ admits the following local coordinate on Δ:

$$x_3=\psi_\Delta(x_1,x_2)\iff\Psi_\Delta(x_1,x_2,x_3)=0$$

Let us define the mapping:

$$\hat{\psi}_\Delta:\boldsymbol{\Delta}\subset\mathbb{R}^2\to\mathbb{R}^3$$
$$(x_1,x_2)\mapsto(x_1,x_2,\psi_\Delta(x_1,x_2))$$

then a surface integral can be written using local coordinate neighbourhoods Δ and local mappings $\hat{\psi}_\Delta$:

$$\int_{\partial\Omega}f\,\mathrm{d}s=\sum_\Delta\int_\Delta f\circ\hat{\psi}_\Delta|\nabla\Psi_\Delta|\,\mathrm{d}x_1\,\mathrm{d}x_2$$

where:

$$|\nabla\Psi_\Delta|=\sqrt{1+\psi_{\Delta,1}^2+\psi_{\Delta,2}^2}$$

In the following, just one local coordinate neighbourhood Δ will be taken into account. Firstly, let us notice that if $f\in C^1(\mathbb{R}^2,\mathbb{R})$, then

$$\frac{\partial f}{\partial x_j}\circ\hat{\psi}=\frac{\partial}{\partial x_j}(f\circ\hat{\psi})-\frac{\partial}{\partial x_3}(f\circ\hat{\psi})\frac{\partial\psi}{\partial x_j},\quad j=1,2\ \text{and}$$

$$\frac{\partial f}{\partial x_3}\circ\hat{\psi}=\frac{\partial}{\partial x_3}(f\circ\hat{\psi})$$

The same kind of relation holds for a smooth vector field $\boldsymbol{\tau}$:

$$(\nabla\cdot\boldsymbol{\tau}-\langle\nabla\boldsymbol{\tau}:\ n\otimes\mathbf{n}\rangle)\circ\hat{\psi}=(\tau_{i,j}\delta_{ij}-\tau_{i,j}n_in_j)\circ\hat{\psi}=(\delta_{ij}-(n_i\circ\hat{\psi})(n_j\circ\hat{\psi}))(\tau_i\circ\hat{\psi})_{,j}+R_i(\tau_i\circ\hat{\psi})_{,3}$$

$$R_i=-\sum_{j=1}^2(\delta_{ij}-(n_i\circ\hat{\psi})(n_j\circ\hat{\psi}))\frac{\partial\psi}{\partial x_j}+\delta_{i3}-(n_i\circ\hat{\psi})(n_3\circ\hat{\psi})$$
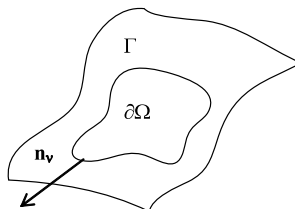

Fig. A.1. Definition of $\mathbf{n}_v$, the unitary vector normal to the edge Γ of $\partial\Omega$.

As

$$\mathbf{n} = \frac{1}{\sqrt{1 + \psi_{,1}^2 + \psi_{,2}^2}} \begin{vmatrix} \psi_{,1} \\ \psi_{,2} \\ -1 \end{vmatrix}$$

this leads to the fact that $R_i = 0$ for $i = 1, 2, 3$.

So that the following integral can be decomposed as follows:

$$I = \int_\Delta f \circ \hat{\psi}_\Delta (\nabla \cdot \boldsymbol{\tau} - \nabla \boldsymbol{\tau} : \mathbf{n} \otimes \mathbf{n}) \circ \hat{\psi}_\Delta |\nabla \Psi_\Delta| \mathrm{d}x_1 \, \mathrm{d}x_2$$

$$= \sum_{i=1}^3 \sum_{j=1}^2 \int_\Delta f \circ \hat{\psi}_\Delta (\delta_{ij} - n_i n_j)(\tau_i \circ \hat{\psi}_\Delta)|\nabla \Psi_\Delta|_{,j} \mathrm{d}x_1 \, \mathrm{d}x_2$$

Using Green formula this leads to the following

$$I = I_0 + I_1 + I_2$$

$$I_0 = \sum_{i=1}^3 \sum_{j=1}^2 \int_\Delta ((f \circ \hat{\psi}_\Delta)(\delta_{ij} - n_i n_j)(\tau_i \circ \hat{\psi}_\Delta)|\nabla \Psi_\Delta|)_{,j} \mathrm{d}x_1 \, \mathrm{d}x_2$$

$$I_1 = -\sum_{i=1}^3 \sum_{j=1}^2 \int_\Delta (f \circ \hat{\psi}_\Delta)_{,j}(\delta_{ij} - n_i n_j)(\tau_i \circ \hat{\psi}_\Delta)|\nabla \Psi_\Delta| \mathrm{d}x_1 \, \mathrm{d}x_2$$

$$I_2 = -\sum_{i=1}^3 \sum_{j=1}^2 \int_\Delta (f \circ \hat{\psi}_\Delta)(\tau_i \circ \hat{\psi}_\Delta)((\delta_{ij} - n_i n_j)|\nabla \Psi_\Delta|)_{,j} \mathrm{d}x_1 \, \mathrm{d}x_2$$

Using the divergence theorem on I_0, and summing on all local mappings, the following term appears:

$$\int_\Gamma f \boldsymbol{\tau}_s . \mathbf{n}_\nu \, \mathrm{d}l$$

As for I_1, one gets:

$$\sum_{j=1}^2 (f \circ \hat{\psi}_\Delta)_{,j}(\delta_{ij} - n_i n_j) = \sum_{j=1}^2 (f_{,j} \circ \hat{\psi}_\Delta)(\delta_{ij} - n_i n_j) + f_{,3} \circ \hat{\psi}_\Delta \sum_{j=1}^2 \hat{\psi}_{\Delta,j}(\delta_{ij} - n_i n_j)$$

As $R_i = 0$ the sum in the second term on the right hand side can be replaced by:

$$\delta_{i3} - (n_i \circ \hat{\psi})(n_3 \circ \hat{\psi})$$

so that

$$\sum_{j=1}^2 (f \circ \hat{\psi}_\Delta)_{,j}(\delta_{ij} - n_i n_j) = \sum_{j=1}^3 (f_{,j} \circ \hat{\psi}_\Delta)(\delta_{ij} - n_i n_j) = (f_{,i} - n_i f_{,j} n_j) \circ \hat{\psi}_\Delta$$

For I_2 one gets:

$$\sum_{j=1}^{2}((\delta_{ij} - n_i n_j)|\nabla \Psi_A|)_{,j} = -n_i C |\nabla \Psi_A|$$

with:

$$C = \frac{\frac{\partial^2 \psi}{\partial x_1^2}\left(1 + \left(\frac{\partial \psi}{\partial x_1}\right)^2\right) + \frac{\partial^2 \psi}{\partial x_1^2}\left(1 + \left(\frac{\partial \psi}{\partial x_1}\right)^2\right) - 2\frac{\partial^2 \psi}{\partial x_1 \partial x_2}\frac{\partial \psi}{\partial x_2}\frac{\partial \psi}{\partial x_1}}{\left(1 + \left(\frac{\partial \psi}{\partial x_1}\right)^2 + \left(\frac{\partial \psi}{\partial x_2}\right)^2\right)^{3/2}}$$

which is the mean curvature (Aris, 1962).

It follows that:

$$I_1 + I_2 = -\sum_{i=1}^{3} \int_A (\tau_i \circ \hat{\psi}_A)(f_{,i} - n_i f_{,j} n_j - n_i C f) \circ \hat{\psi}_A |\nabla \Psi_A| \, dx_1 \, dx_2$$

Summing on all the local coordinate neighbourhoods leads to

$$I_1 + I_2 = -\int_{\partial \Omega} \nabla f \cdot \boldsymbol{\tau} \, dS + \int_{\partial \Omega} \left(\frac{\partial f}{\partial n} + C f\right) \boldsymbol{\tau} \cdot \mathbf{n} \, dS$$

So that the lemma is proved. We can notice that in the preceding demonstration $\delta_{ij} - n_i n_j$ is the projection operator onto the surface. Indeed let us define

$$\mathbf{V}_s = \mathbf{V} - (\mathbf{V} \cdot \mathbf{n})\mathbf{n}$$

then

$$V_{sj} = V_j - (V_i n_i)n_j = (\delta_{ij} - n_i n_j)V_i$$

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