



# Probability density functions for fluctuations in turbulent two-phase flames

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## Abstract

Probability density function methods for characterizing various fluctuating quantities in the reacting gas phase of turbulent spray flames are presented, for the development of which E.E. O'Brien has been a pioneer. Balance equations for two-phase flows considered as a piecewise continuous medium are first presented, with the interface conditions containing the vaporization-condensation equilibrium. From these, the equation for the joint probability density function of temperature and composition in the gas phase is derived, in which specific terms related to vaporization are identified. The terms needing modeling are then highlighted. The first one is the classical micromixing term, which has to include all the features already discussed for single phase reacting flows, but adaptation to two phase flow has to be questioned. And, mainly, several unclosed terms are dealing with vaporization, needing a specific attention. It is shown that proposals for their modeling can be obtained on the same bases as in the so-called Eulerian-Lagrangian models, which will directly induce changes of the shape of the probability density function. We finally discuss the model presented herein and comment its connection with recently published literature for this subject. Specific attention is devoted to the combustion regimes and to the presence of diffusion flamelets surrounding fuel droplets or groups of droplets. The proposals presented here go one step further compared to the recent studies, and they have now to be compared with well defined experiments, numerical experiments with direct numerical simulations or real experiments in conveniently controlled conditions.

*Keywords* : Turbulent spray flames, JPDP for chemical species, Eulerian-Lagrangian models, Modeling of droplets vaporization-condensation

## 1/ Introduction

Turbulent flames calculations have been a challenging problem since more than fifty years, when the use of high velocity flows for flames has shown its interest. The fact that species diffusion becomes turbulent was not a new difficulty, but how the turbulent fluctuations of chemical composition and temperature do modify the mean turbulent reaction rates has been much more difficult to understand. In fact, it has been very early recognized that, for a jet diffusion flame with very fast chemistry, the mean reaction rate is controlled by the mixing process and not by chemistry (H.L. Toor<sup>1</sup>, 1962, inspired by Hawthorne et al.'s<sup>2</sup> pioneering study in 1949).

Ten years later, in 1974, C. Dopazo and E.E. O'Brien<sup>3</sup> found the possibility of calculating the multidimensional joint PDF (Probability Density Function) of all interesting species and temperature, that allows to get the reaction rates in all cases. In 1976, R.W. Bilger<sup>4</sup> has shown that the knowledge of the PDF of the fluctuations of some « mixture fraction », adequately defined, allows to calculate exactly this mean reaction rate, but he proposed approximate forms of this PDF, instead of more precise calculations.

It appeared later that independantly, at the same time, some studies have been initiated in Russia also (in 1975, by V.A. Frost<sup>5</sup>, and by V. Sabelnikov<sup>6</sup>, in 1982). In the field of chemical engineering, some PDF models have been proposed, even earlier, for taking into account

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fluctuations of chemical composition in reactive flows, but they were « heuristically » proposed, see for instance R.L. Curl<sup>7</sup>, J. Villiermaux and J.C. Devillon<sup>8</sup> or R.C. Flagan and J.P. Appleton<sup>9</sup>. Since this time, the so-called « PDF methods » have been used with success for many different types of turbulent flames. The effect of « small scale mixing » has concentrated the efforts, E.E. O'Brien and coworkers<sup>10, 11, 12</sup>, again have been pioneers for that. Equally, the way to process the numerical computation of the multidimensional equations involved has needed also specific methods (see the 1985 review paper by S.B. Pope<sup>13</sup>). For the application to realistic multi-reactive flows, it has been necessary to study specifically the handling of the sufficiently complex chemical mechanisms, that can be different for each particular problem (the pioneers here are U. Maas and S.B. Pope<sup>14</sup>).

Another kind of turbulent flames, that are involved in many situations and practical devices, present even more difficult modelling problems. In « spray flames », the reacting turbulent flow is a two-phase (or sometimes multiphase) flow, the fuel being injected into the liquid phase and immediately dispersed in parcels and fine droplets (or sometimes the oxidizer alone, or both). The combustion reactions themselves take place essentially in the gas phase, using the species vaporized from the liquid parcels, and are confronted also to turbulent fluctuations (that are complicated due to the many droplets with velocities and mass density different from the surrounding gas).

The problem with modeling two-phase flows, with the turbulent dispersion of liquid parcels (even spherical), is already very difficult without any chemical reaction, but it has been studied for fifty years. Almost all models are using an Eulerian description for the turbulent gas phase and a Lagrangian model in order to follow samples of liquid droplets. The attempts for taking into account chemical reactions do assume generally that the vapor issued from very small and dispersed droplets is immediately mixed with the surrounding turbulent fluid in the calculation cell, and the reaction rates in the gas phase are then calculated simply using the purely chemical reaction rate. But this means implicitly that the gas phase fluctuations of composition and temperature are very rapidly dissipated, before chemical reactions will take place, and we know that this assumption may be very poor in turbulent flames.

The small scale structure of the spray flames has often been discussed, with the question of knowing if there is a flamelet around each droplet, or if one longer flamelet surrounds a group of droplets. Indeed, spray flames can possibly show very different small scale structures, for different conditions or for different positions within the flame brush. It is interesting to discuss this with some more details here. The drawings below (fig.1) show the expected structures in an « academic » configuration where a spray flame is established in the mixing zone between one flow carrying fuel droplets (in some inert gas) and a flow of gaseous oxidiser (taken from the french text book by R. Borghi and M. Champion<sup>15</sup>, chapter 9). There are diffusion-reaction zones (D) between fuel rich parts of the medium, around droplets or groups of droplets (on the fuel lean right part of the flame brush), and also around pockets of oxidant gases that large scale turbulent vortices have pushed towards the fuel rich left side. In the mid of the flame brush, there is necessarily a much longer « percolating » diffusion-reaction zone between fuel rich gases (on the left side) and fuel lean gases. When the chemical reactions are not very fast, the flame brush is stabilized, at some distance from the confluence of the two streams, by small premixed flamelets (PM), related with the diffusion-reaction zones by « triple flamelets ».

The lift-off distance depends on how fast the chemical reactions are, with respect to the turbulence created by the streams. It is, of course, shorter when the chemical reactions are faster. The diffusion-reaction zones are more or less wrinkled, stretched by turbulent motions, and may be locally and temporarily extinguished, depending on their intensity. When (and where) there are too many local extinctions and reignitions, the flame brush can be qualified as « distributed ». We have not shown this situation, the structure of the flame brush is simply « fuzzy ».

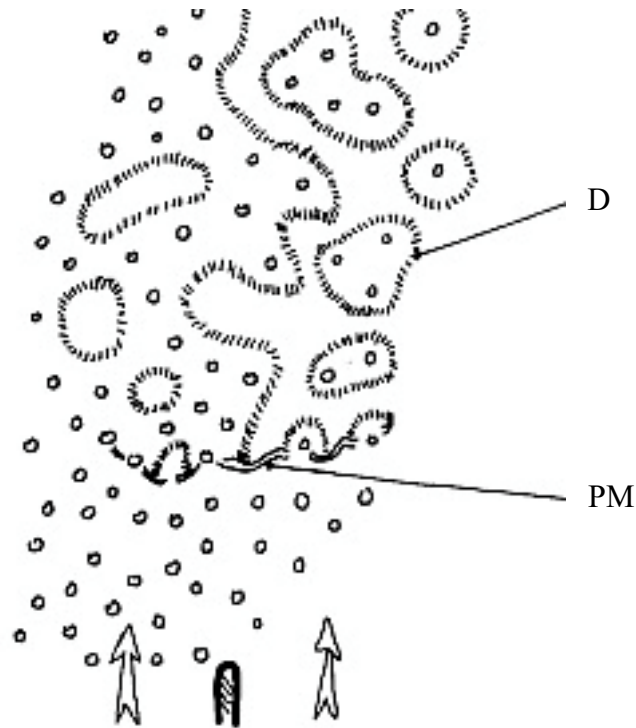


Fig.1. Spray flame between a stream feeding fuel droplets and an oxidizer gas stream.

These features can be more or less easily recognized in laboratory experiments. Figure 2 shows spray flames behind a single injector of liquid methanol assisted by a gas flow, in the surrounding air, studied by D. Stepowski et al.<sup>16</sup>. The images are taken using planar LIF OH, and the droplets are visible also, and the injector is assisted by air, N<sub>2</sub> or air + O<sub>2</sub>.

When the atomizing gas is nitrogen, only the diffusion-reaction zone is visible, enveloping the flame brush and the fuel rich central zone. Due to the atomizing air, an internal diffusion flame is seen, the global flame draws a W. In all cases, there are small premixed flamelets, with triple flamelets, allowing the stabilization of the flame brush. Their size is very amplified when the atomizing air contains more oxygen. The lift-off distance is shorter, one can see much more premixed flamelets in the center of the flame brush.

For all these possible structures, even if they show very large fluctuations, some knowledge of the fluctuations in the gas phase could be obtained with the joint PDF of temperature and species concentration, at it was in single phase turbulent flames. It is known that chemical reactions can be easily taken into account, it remains to be able to take into account the mass added by vaporization. Several attempts have been done in this direction, since 1995 to 2000, for instance by R. Borghi<sup>17</sup> and by P. Durand, M. Gorokhovski and R. Borghi<sup>18</sup>. At first sight, it seems easy to propose a stochastic Monte Carlo model using many « notional particles », to be added to the classical Lagrangian tracking of liquid droplets. This has been adopted since now in different papers (see the review paper by Jenny et al.<sup>19</sup>). But, in these models, a crucial submodel is related to the technique of injection of these notional particles, originated from the vaporized mass coming from the droplets, and there are different ways of doing so (see ref.19 just given, sec. 5.4.5). This problem can be clarified by using a joint PDF balance equation, which can be obtained with a theoretical basis as sound as that obtained by E.E. O'Brien. A first study of such an equation has been initiated by M. Zhu et al.<sup>20</sup>, but not in a « closed » form. The basic problem is to establish a secure theoretical basis for deriving this PDF with account of the feeding of the gas phase by vaporized fuel issued from the liquid phase. In fact, different one point PDF's could be of interest ; the most general is the joint PDF of the velocities, species mass fraction and temperature for both liquid and gas phases, but it is the most difficult to

handle ; the joint PDF conditioned for the gas phase only is simpler ; we will be interested in this paper simply by the joint PDF of temperature and composition in the gas. Anyway, all these equations that will not be « closed », because they consider variables in only one point of the flow, have to be completed by some assumptions to model some of the terms. Following the paper by M. Zhu et al, we have discussed both aspects of this problem at the end of our general book on multiphase turbulent flows<sup>21</sup>. We present here an extended version of this study.

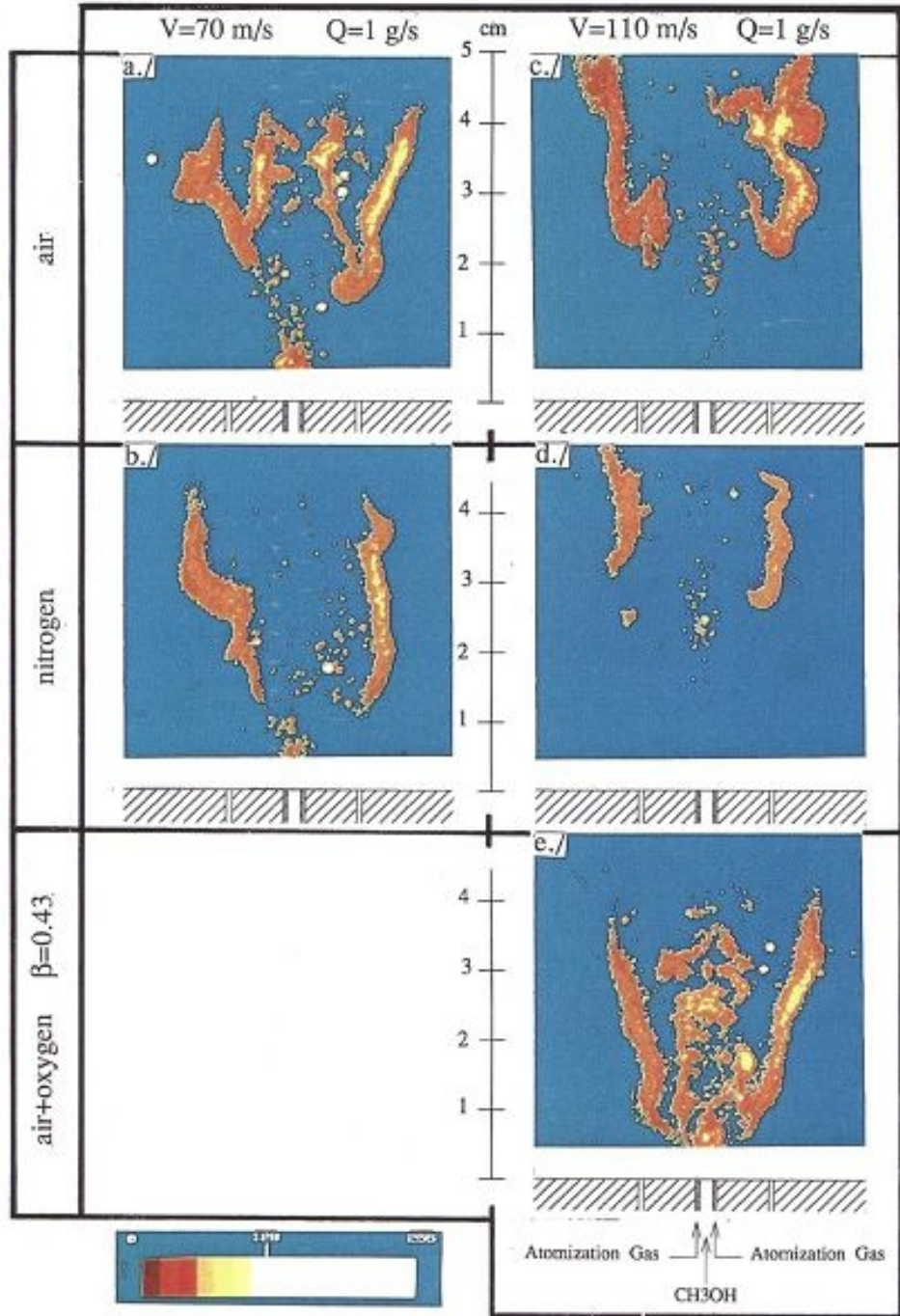


Fig. 2. Images of spray flames of liquid methanol and air (the real lift-off distance is not shown).

Reproduced with permission from the paper by D. Stepowski, A. Cessou and P. Goix<sup>16</sup> (1994). Copyright 1994 by The Combustion Institute. Published by Elsevier Science Inc.

We are not dealing here with the the problem of liquid dispersion itself (in the physical space). This problem is difficult and only partially solved, even if the liquid is considered under the

form of many droplets. We will assume that we know sufficiently realistic models for that, but we will be obliged to consider how the model for the fluctuations of the gas phase can be coupled with that for the liquid phase.

The equations that we can use as a basis for this study, valid with the same conditions and scales as the Navier-Stokes equations, can be obtained representing the multiphase medium as a « piecewise continuous medium », for instance as presented by D.A. Drew<sup>22</sup>. The approach has been extended with mass exchange between phases by I. Kataoka<sup>23</sup>. The book by R.I. Nigmatulin<sup>24</sup> uses also this approach, proposed independantly with different notations. The second section of this paper will give these basic equations, emphasizing their peculiarities and particularly the « instant interface balance relations ».

We will then discuss how to use this theoretical description in order to build prediction models for two-phase flows with combustion. We consider possible DNS, models for mean values, and finally we introduce a Lagrangian-Eulerian PDF modelling.

The third section of this paper is devoted to showing how it is possible to obtain equations for the PDF's of interest for two-phase turbulent combustion. The PDF of chemical mass fractions, and temperature, in the gas phase is particularly interesting. The PDF of temperature of the liquid phase, particularly the surface temperature of droplets, is also very interesting for calculating the mean rate of mass exchange between the liquid and gas phases. The line of the method is described but not all the details. More interestingly, we will explain the physical significances and implications of the different terms in the equations.

The fourth section will discuss the question to find closure assumptions for the different terms related to the vaporization. We will not discuss generally this problem, but we will propose simply a way to couple the PDF approach with a classical Lagrangian simulation model that uses the classical local knowledge on vaporization around droplets. The method that we propose is an improved way to use this classical knowledge, taking into account the fluctuations of the gas medium in the vicinity of the droplets. The question of the influence of the droplets on the small scale dissipation of the scalar fluctuations in the gas will be also discussed.

In the fifth and last section, we will situate our work with respect to the studies performed recently for the prediction of such a PDF. We will not present calculations for a given case, but instead we will discuss what is expected from this PDF approach, pointing a few important questions that will have to be studied in more details, in order to improve the proposed modelling.

## 2/ Theoretical grounds

### 2.1 Basic instant equations for a piecewise continuous flow

The spray flames are mainly concerned by gas-liquid flows, where the liquid is dispersed in several (often very many) parcels, of different sizes and shapes. The description of the two-phase flow as a piecewise continuous medium begins with the definition of the variables<sup>2</sup>  $f_k$ , which are 1 in the phase  $k$  only, and zero elsewhere. Here there are only two  $f_k$ 's, for  $k=liq.$  and  $k=gas$ , and they are such that  $f_g=1-f_l$ , but the writing of theoretical developments is easier with the use of the general notation  $f_k$ . The surface where the  $f_k$ 's are discontinuous is the liquid-gas interface. Of course, the interface is permanently moving. Without any assumptions concerning the droplets formation and interactions, the interface area is permanently created by the stretching due to the liquid and gas flows, and destroyed when two parts of this interface are pushed sufficiently close one against the other, creating parcels or droplets, or bubbles or

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<sup>2</sup> The list of the main notations and symbols is provided as an Appendix at the end of the text.

gas pockets. In addition, a change of interface area results also from the exchange of mass between the liquid and gas phases, by vaporization and condensation.

It is interesting to emphasize that the indices  $f_k$ , within the entire interesting spatial domain, do follow a pure convection equation, with the velocity of the interface  $v_s$ . It writes :

$$\frac{\partial}{\partial t}(f_k) + v_s^a \frac{\partial}{\partial x_a}(f_k) = 0 \quad (1)$$

We use here tensorial notation, but restricted for orthonormal reference frame. Therefore, superscript  $a$  in terms such as  $v_s^a$  refers to the  $a$  component of the velocity vector  $v_s$ . The same holds for  $\frac{\partial}{\partial x_a}$ . The equations, containing non continuous quantities, are written in the framework of generalized functions. In principle, it would be necessary for (1) to extend the definition of  $v_s$  at every point of the domain (out of the interface), but  $\frac{\partial}{\partial x_a}(f_k)$  is zero outside the interface.

Without exchange of mass between the phases,  $v_s$  is always equal to both  $v_{gs}$  and  $v_{ls}$  at each point of the interface. But we will see that, when there is exchange of mass between the phases, there is a discontinuity on the interface for the normal velocities of the phases,  $v_s$  is different from  $v_{gs}$  as well as from  $v_{ls}$ , and is linked to them once the mass flow rate of exchange is known (we will see how at the end of this section).

The flows within each fluid, as well at the interface itself, have to follow balance equations. It is interesting to write these equations with in a unified form, even if the variables can be discontinuous at the interface. The equations are then written in a generalized sense (as was (1)), see refs. 21-22-19.

#### A/ The instant Eulerian balance equation for the masses

The instant Eulerian balance equation for the mass of phase  $k$  within the entire space is :

$$\frac{\partial}{\partial t}(\rho_k f_k) + \frac{\partial}{\partial x_a}(\rho_k f_k v_k^a) = m_k \sigma \quad (2)$$

The right hand side term, which represents the local mass exchange between the two adjacent phases, is non zero only on the interface ( $m_k$  is defined only on the interface). It is the local volumetric mass of phase  $k$  « created » (or destroyed) at the interface. We define  $\sigma$  as the « local instantaneous interface area by unit of volume ». It has the structure of a Dirac function located on the interface, it can be shown that :

$$-n_k^a \sigma = \frac{\partial}{\partial x_a} f_k$$

where  $n_k$  is the normal at the interface (assumed always unique at a given point), outwards oriented with respect to the phase  $k$  (then, at the same point of the interface,  $n_l = -n_g$ ).

This equation describes both the bulk of phase  $k$  and the interface. Eq. (2) can be written also :

$$f_k \left( \left( \frac{\partial}{\partial t} \rho_k \right) + \frac{\partial}{\partial x_a}(\rho_k v_k^a) \right) = - \left( \rho_k \left( \frac{\partial}{\partial t} f_k \right) + \rho_k v_k^a \frac{\partial}{\partial x_a}(f_k) \right) + m_k \sigma$$

The right hand side is non zero only at the interface, it has the structure of a Dirac peak. On the contrary, the left hand side has no Dirac peak, and concerns only the continuous bulk of phase  $k$ . They have then both to be zero. Effectively, the left hand side results in the well known continuity equation inside phase  $k$ . The right hand side, with (1) and the relation defining  $\sigma$ , gives finally :

$$m_k \sigma = -\rho_k (v_k^a - v_s^a) n_k^a \sigma = -\rho_k v_{krs}^a n_k^a \sigma \quad (3)$$

That gives a more physical meaning to  $m_k$ , and we can write :

$$\frac{\partial}{\partial t}(\rho_k f_k) + \frac{\partial}{\partial x_a}(\rho_k f_k v_k^a) = -\rho_k v_{krs}^a n_k^a \sigma \quad (2')$$

When there is no mass exchange, the velocities of both phases at the interface, and of the interface, are equal, but the exchange of mass gives non zero normal  $v_{krs}$ .

On the other hand, the sum of the equations (2), or (2'), for  $g$  and  $l$  does represent the instant balance equation for the mass of both phases (even with density discontinuity). It has not to show any source or sink term. It follows that :

$$\rho_g v_{grs}^a n_g^a \sigma + \rho_l v_{lrs}^a n_l^a \sigma = 0 \quad (4)$$

Not surprisingly, the mass flow normally entering (or exiting) the gas phase is the same as the one exiting (or entering) the liquid phase ( $m_g = -m_l$ ). This is an « interface balance relation ».

Remark that this relation is not able to prescribe  $m_g$  (nor the  $v_{krs}$ ), which have to be given by considering the phenomenon that is responsible for the mass exchange (we will see later how to calculate it).

## B/ The instant balance equations for the momentum in each phase

We can write also the instant momentum balance equation for the mass of phase  $k$ , that includes the Cauchy stress tensor  $C_k$  within the phase  $k$  (including the isotropic contribution of the pressure plus the viscous friction tensor), plus the contact force  $F_k$  experienced by  $k$  from the interface, per unit of area, plus the exchange of momentum due to the exchange of mass. If it is not negligible, gravity force is included.

$$\frac{\partial}{\partial t} (\rho_k f_k v_k^b) + \frac{\partial}{\partial x_a} (\rho_k f_k v_k^b v_k^a) = \frac{\partial}{\partial x_a} (f_k C_k^{ab}) + \rho_k f_k g^b + m_k v_k^b \sigma + F_k^b \sigma \quad (5)$$

Here, the « interface balance relations » have to take into account interfacial forces (if we do not neglect them). We can obtain these relations by the same algebra as for the mass, but now writing them between the phase  $k$  and the interface, and between the interface and the adjacent phase  $k'$ . Taking the case of gas and liquid phases, we obtain first that :

$$C_g^{ab} n_g^a \sigma = F_g^b \sigma \quad (6) \quad \text{and the same for liquid}$$

That means that the contact force per unit of area is equal and opposite to the force that the Cauchy stress in one phase exerts normally on the interface.

If we disregard surface tension and exchange of mass, we know that the sum of momentum of both phases does not display any contact force term, then

$$F_l^b + F_g^b = 0 \quad (6')$$

But with exchange of mass, we have :

$$F_g^b + m_g v_{gs}^b + F_l^b + m_l v_{ls}^b = 0 \quad \text{or} \quad C_{gs}^{ab} n_g^a + m_g v_{gs}^b + C_{ls}^{ab} n_l^a + m_l v_{ls}^b = 0 \quad (7)$$

If we consider surface tension, there is a special source of momentum on the interface, due to physical actions at the molecular level. Then the right hand side of (6'), or (7), is not nil, but equal to the surface tension force, and a special model for it has to be added (usually, the algebraic Laplace model).

## C/ The instant balance equations for the total energy of each phase

We can write also the instant balance equation for total energy of phase  $k$  (internal energy plus kinetic energy),  $e_{tk}$ , taking into account molecular conduction within this phase, power of contact forces and heat exchange between the phases. It is :

$$\begin{aligned} \frac{\partial}{\partial t} (\rho_k f_k e_{tk}) + \frac{\partial}{\partial x_a} (\rho_k f_k e_{tk} v_k^a) = & \frac{\partial}{\partial x_a} (f_k (-j_{ek}^a + C_k^{ab} v_k^b)) + \rho_k f_k v_k^b g^b \\ & + F_k^b v_k^b \sigma + q_k \sigma + m_k e_{tk} \sigma \end{aligned} \quad (8)$$

The  $j_{ek}$  is the diffusive energy flux within phase  $k$ , i.e the conduction heat flux when the phase is not a mixture of several constituents (we do not consider here radiative heat transfer). The term  $\frac{\partial}{\partial x_a} (f_k C_k^{ab} v_k^b)$  is the power due to Cauchy stresses, and the power of gravity force is written also. The energy exchange flux with the interface is composed by the heat flux coming



from the interface (normal to the surface),  $q_k\sigma$ , the power of the contact force  $F_{kv_k}\sigma$ , and the total energy brought with the exchange of mass,  $m_k e_{tk}\sigma$ .

In the case where one phase is a mixture, even with possible chemical reactions, the equation is written in a very similar form. It has just to be noticed that the energy diffusion flux is not only due to conduction, there is also a contribution due to species diffusion. There is no reaction term (even if the reactions can change the temperature), if we define the internal energy including the energies of formation of all the species involved.

On the interface, equation (8) for  $k$  gives, at each point and each time, that heat coming from the interface is equal to the normal conduction flux entering within the phase,  $q_k = -j_{eks} n_k$  ( $q_k$  is only defined on interface,  $j_{eks}$  is the value of  $j_{ek}$  at the interface). The same holds for  $k'$ .

When the surface tension is neglected, the interface balance relation for total energy for adjacent phases  $k$  and  $k'$  gives that the total energy flux leaving one phase is entering into the adjacent one :

$$C_{gs}^{ab} v_{gs}^b n_g^a + m_g e_{tgs} + q_g + C_{ls}^{ab} v_{ls}^b n_l^a + m_l e_{tls} + q_l = 0 \quad (9)$$

In this relation we can also replace  $q_g$  by  $-j_{egs} n_g$ , and for the liquid equally. Then the sum of the total energy balance equations for all phases does not display any exchange term.

With surface tension, the power of the surface tension force has to be taken into account, and it is generally preferred to define an « internal interfacial energy », by unit of area, and to write its own balance equation. The sum of the total energy of all phases, plus this interfacial energy, then does not show any additional term.

#### D/ The instant balance equations for the species in gas phase

When one phase is a mixture of different constituents (for spray flames, it is always the case of the gas phase), we can write instant Eulerian balance equations for each constituent. Let us define the mass fraction of species  $j$  within the phase  $g$ ,  $Y_{jg}$ . The instant balance equation, valid everywhere in the physical space, is :

$$\frac{\partial}{\partial t} (\rho_g f_g Y_{jg}) + \frac{\partial}{\partial x_a} (\rho_g f_g v_g^a Y_{jg}) = \frac{\partial}{\partial x_a} (-f_g j_{Dj}^a) - \rho_g Y_{jg} v_{rjg}^a n_g^a \sigma + \rho_g f_g w_j^{ch} \quad (10)$$

We see first on the right hand side the molecular diffusion flux of  $j$  (within the phase  $g$ ). The last term represents the chemical reaction rate from reactions within the gas phase. It is generally a sum of the reaction rates of all significant reactions. Of course, the sum of the  $Y_{jg}$  is unity, and if there are  $N$  species it is sufficient to write  $N-1$  equations like that.

More interesting here, the second term is the exchange term of the mass of  $j$  between the two phases, due to vaporization-condensation in case of spray flames, proportional to  $\sigma$ . Here  $v_{rjgs}^a$  is the velocity of species  $j$  in the gas, relative to the interface. That means that we have to take into account the molecular diffusion flux of mass of  $j$ , at the interface,  $j_{Djs}^a$ :

$$-\rho_{gs} Y_{jgs} v_{rjgs}^a n_g^a = -(\rho_{gs} Y_{jgs} (v_{gs}^a - v_s^a) + j_{Djs}^a) n_g^a = m_g Y_{jgs} - j_{Djs}^a n_g^a \quad (11)$$

When the species  $j=V$  can move from liquid phase ( $l$ ) to gas phase ( $g$ ), or vice versa, by vaporization or condensation,  $v_{rVgs}$  is not zero, and the interface balance relation for  $Y_{Vg}$  writes (knowing that  $m_g = -m_l$  and  $n_g = -n_l$ ) :

$$m_g Y_{Vgs} - j_{DVgs}^a n_g^a = m_g Y_{Vls} - j_{DVls}^a n_g^a \quad (12)$$

When the liquid phase is composed only of species  $V$ , of course  $Y_{Vl}=1$  everywhere in the liquid, and it comes :

$$m_g Y_{Vgs} - j_{DVgs}^a n_g^a = m_g \quad (12')$$

When the species  $j$  can be only present in gas phase, no exchange of mass of  $j$  is possible,  $v_{rjgs}$  is nil, and we have :

$$m_g Y_{jgs} - j_{Djs}^a n_g^a = 0 \quad (12'')$$

(of course, without any exchange of mass  $j_{Djgs}=0$ ).

When considering the vaporization-condensation processes, it is very classically considered that the rate of vaporization as well as the rate of condensation are very fast with respect to all others occurring (convection, diffusion), in such a way that there is at any time a vaporization-condensation equilibrium. Thermodynamics says that this equilibrium prescribes that  $Y_{Vgs}$  equals its saturation value, a function depending on the interface temperature :

$$Y_{Vgs} = Y_{Vsat}(T_s, p_s)$$

(in fact, the primitive formula uses the mole fraction, but mass and mole fractions are related, almost equal if the molar masses of components are close). The interface temperature (common to the gas and the liquid at the considered point) is always smaller than, or equal to, the boiling temperature, and in this case  $Y_{Vgs}$  is one.

This condition, together with the interface relation (12'), gives then the law prescribing  $m_g$  :

$$m_g = -j_{DV_s}^a n_g^a / (1 - Y_{Vsat}(T_s, p_s)) \quad (13)$$

## E/ Equations of state and irreversible laws for each phase

Completing the description to the instant two-phase medium, we have to add two additional informations.

First, « Equations of state » are needed for each phase, and, of course, they are different. But they have the same structure : the Cauchy stress tensor is  $C_k^{ab} = -p_k \delta^{ab} + \tau_k^{ab}$  and the first state equations will allow to relate  $p_k$  (the reversible part of  $C_k$  here), at each location in the medium, to the variables that are satisfying the balance equations. As well for a liquid as for a gas, the role of thermodynamics is to give us a general law relating the pressure to the density  $\rho$  and the internal energy per unit of mass :  $p_k = p_k(\rho_k, e_k)$  (which can be inverted in  $\rho_k = \rho_k(p_k, e_k)$ ). In the limit case where the liquid phase can be assumed to display a constant density, this property plays the role of equation of state, and the result is that local pressure is given by the mass balance equation. The second (thermal) equation of state allows to compute the local temperature of the medium, as a function of the energy per unit of mass and the density,  $T_k = T_k(\rho_k, e_k)$  (which can be inverted in  $e_k = e_k(\rho_k, T_k)$ ). When the gas phase is a mixture of gases, and we are concerned with that for combustion, all the variables  $Y_{jg}$  have also to appear in the state equations. In this case, usually, the well known laws for a mixture of perfect gases can be used, and the simplest realistic formulae are :  $p_g = \rho_g R(\sum Y_{jg}) T_g$ , and  $e_g = C_v T_g + \sum (e_j^0 Y_{jg})$ , the  $e_j^0$  being the energies of formation of species  $j$  at 0°K.

Second, the laws for the irreversible fluxes are needed, within each phase, and again the irreversible thermodynamics is able to fulfill this need. These concern viscosity (the irreversible part of the Cauchy stress tensor), heat conduction, species diffusion when the phase is a mixture of different chemical species, and in this case the simplest Fick laws are generally used. All these laws relate the local fluxes to the local gradients of velocities,  $T$  and all  $Y_{jg}$ . For reactive flows, the reaction rates for each reactive species have also to be given, once a reactive scheme has been chosen, as functions of  $T$  and all  $Y_{jg}$ . Chemical kinetics is able to satisfy this need.

## 2.2 Different ways for using these equations

### A/Direct Numerical Simulations

The balance equations for each phase, associated to state equations and irreversible laws, plus the  $f_g$ 's equations, give us a complete set of equations. Let us recall that the exchange fluxes between phases are not unknown, they are directly related to the fluxes (reversible and irreversible) within the phases, of course at the conditions of the interface. In particular, the mass exchange term  $m_g$  is given by (13), and it plays also on the dynamics of the interface because it is linked with its velocity by (3). At this interface, there are discontinuities for some of the variables, and the interface balance relations prescribe these. Other variables are

continuous, for instance tangential velocities, temperature when there is no interfacial internal energy, these prescriptions are usual for non rarefied fluids. The balance equations are partial differential equations, they have to be associated to initial conditions and boundary conditions in a given spatial domain, as it is the case in classical fluid dynamics.

It has to be emphasized that the interface position can be obtained from the  $f_g$  (or  $f_l$ ) equation or from the Eulerian balance equation of the mass of one phase, but with very delicate numerical processing in an unsteady framework. A method of « interface reconstruction » has to be used to obtain the interface position from the obtained numerical Eulerian field of mass (or volume) of liquid, which suffers directly from the numerical diffusivity. Another way is to use (1) for a Lagrangian track of the interface, from an initial position, with difficulties due to the very large lengthening of the interface.

With all these equations it is then possible to obtain « true DNS » of the instant two phase-flow, without any models at a scale larger than that of the Navier-Stokes equations. Such DNS's have already been performed for problems without vaporization (nor combustion), in order to study liquid droplets with a shape perturbed<sup>25</sup>, and even for studying the « primary break up » of a liquid jet<sup>26</sup>. The « Atomization regime », where the velocity of the liquid jet is very high and produces a plume of very many droplets, behind the liquid jet core, with wide distribution of sizes, has been also studied<sup>27, 28, 29</sup>. Two-phase flows with vaporization have been also studied<sup>30, 31</sup>.

Anyway, as usual, such DNS will need very very small grid size. Already, for turbulent combustion, a grid size at least ten times thinner than the thickness of a premixed laminar flame has to be used. For liquid jet break up and atomization, it has been necessary to implement a mesh refinement close to the interface (for taking into account the capillarity, it is necessary to be able to obtain the curvature of the interface), and to resolve sufficiently the droplets. With liquid vaporization, it is necessary to resolve even more the droplets, in order to be able to obtain a good approximation of the gradients normal to the interfaces, because they are necessary for computing through (13) the vaporization rates. For keeping tractable the time and cost, the simulations will have to be limited in the range of Reynolds, Damköhler and Weber numbers.

#### B/Use of mean equations, « closed » with additional models

For studying turbulent flows displaying large fluctuations for all measured quantities, in the first instance, it is useful to look at some « mean values ». This has been done in single phase flows, with different definitions of the « mean ». It follows that it is of primary interest to know equations able to calculate directly these mean values. Anyway, the DNS computations are very difficult, very expensive, and it will be impossible to simulate exactly the experiment we are looking for, because we do not know with sufficient precision the boundary and initial conditions that correspond to this experiment.

In single phase flows, turbulence appears when the velocities are too large, giving rise to instabilities that interact for giving the apparent randomness of the flow. The initial and entering conditions of the flow cannot be free of perturbations, that induce turbulence, and even, turbulence is often already present in the initial and entrance conditions. In two-phase flows, the randomness is due, in addition, to the presence of the liquid phase. In many cases, the liquid phase is already dispersed in the entrance flow, and the droplets or liquid parcels are randomly distributed, in terms of sizes and velocities. If the liquid phase entering is continuous, the friction with the gas flow is a source of instabilities and produces very shortly droplets, randomly again. When the liquid entrance velocity is large, a regime called « atomization » of the liquid is obtained, which is of greatest interest because it allows a large increase of the exchanges between the two phases.

The numerical computation of the mean values equations is much easier, only needing a much coarser grid. It is possible also to devote different equations to different kinds of mean values, statistical mean values or « filtered » mean values, more interesting for the study of large scale oscillations of the flow (these are called « LES »). From the beginning of the study of two-phase flows, the works have been developed mainly with mean values within a « representative volume », and the balance equations have been established by models with physical bases, and not with the mathematical precise approach shown here.

For getting « better sounded » equations, it is useful to take as a basis the instant basic equations we have described in the previous section. But, anyway, as it is well known for turbulent single phase flows, these equations cannot be used without some « closure assumptions ». This is shown in details in ref. (19). For the case of statistical means, it is more interesting to deal separately with gas phase and liquid phase, although it is possible to represent the two-phase flow as a global single flow.

We give as an example two of these mean equations, the first one being the mean mass balance equation for the gas phase. From (2), we have :

$$\frac{\partial}{\partial t}(\overline{\rho_g^*} \overline{f_g}) + \frac{\partial}{\partial x_a}(\overline{\rho_g^*} \overline{f_g} \widetilde{v_g^a}) = \overline{m_g \sigma} \quad (14) \quad \text{curieux encore}$$

The notation  $\overline{\rho_g^*}$  (the « intrinsic mean gas density ») means the statistical mean of  $\rho$  inside the phase  $g$  only, and  $(\overline{\rho_g^*} \overline{f_g}) = \overline{\rho_g f_g}$ . The quantity  $\overline{f_g}$ , often called the mean gas volume fraction, is more exactly the probability of presence of gas phase. Of course, since this equation is an usual partial differential equation, all quantities appearing are continuous functions, in particular the mean values of  $f_g$  and of  $m_g \sigma$ .

We can also be interested by the mean velocity of the mixture of two phases (weighed by the density). Defining  $\overline{\rho} = \overline{\rho_g f_g} + \overline{\rho_l f_l}$ ,  $\overline{\rho} \widetilde{v^a} = \overline{\rho_g f_g v_g^a} + \overline{\rho_l f_l v_l^a}$ , and  $\overline{\rho} \widetilde{f_g} = \overline{\rho_g^*} \overline{f_g}$ , it can be written :

$$\frac{\partial}{\partial t}(\overline{\rho} \widetilde{f_g}) + \frac{\partial}{\partial x_a}(\overline{\rho} \widetilde{f_g} \widetilde{v^a}) = -\frac{\partial}{\partial x_a}(\overline{\rho v^a f_g}) + \overline{m_g \sigma} \quad (14')$$

We see now appearing a « diffusion term » of phase  $g$  with respect to the mean mixture.

Of course, in both cases, the last term of exchange of mass needs a closure assumption, in the second case, a model for the « diffusion term » is necessary also. Similar equations can be written for the liquid phase.

Balance equations for phase  $k$  can be found also for mean momentum, mean total energy, and mean mass fraction of each chemical species in the phase, with again closure assumptions needed, and in particular, for turbulent diffusion terms (with respect to the mean velocity of the phase). In addition, there are mean terms of momentum and energy exchanges between the two phases, setting new problems of closure.

All these equations, together with « mean state equations », do constitute what has been called a « Mean Eulerian-Eulerian model with two fluids ». The closure assumptions of these equations have been studied mainly when the spray is dilute, or very dilute, with spherical droplets dispersed in the gas phase. They are much more difficult to study when the spray is dense, with a wide distribution of sizes, and with a randomly moving « liquid core » (see ref. 13).

When the dispersed spray is polydispersed, and eventually gives birth to a turbulent flame, the problems associated to the closure assumptions are even more difficult, because each droplet is vaporizing at its own rate, and this rate depends also strongly of the gas environment. In this case, a Lagrangian description of the « population » of these droplets is usually considered. It is possible, with some very realistic assumptions, to get informations on the droplets, concerning their temperatures, velocities, sizes, and their trajectories with models of turbulent dispersion. In order to obtain realistic approximations of the droplets rates of exchanges with the gas phase, it is necessary to know precisely enough the gas phase, and for that an Eulerian

mean description is kept. By following a sufficiently large number of samples of droplets (feeded by the liquid injector), we then get a sufficiently realistic « Lagrangian-Eulerian model ».

### C/ Lagrangian-Eulerian-PDF modeling

Lagrangian-Eulerian models are now very often used, for a larger range of applications, including flames. But these studies are possible only in the limits of the assumption of spherical droplets, which is always adopted, and provided that collision and formation of droplets can be very simply modelled (or even neglected).

As we have explained in the introduction, there is always an uncomfortable assumption in these Lagrangian-Eulerian models : the Eulerian equations say something about the mean values of the characteristics of the gas phase, but nothing concerning the fluctuations of temperature and mass fraction of species in the gas phase. It is not possible to determine how these fluctuations influence the evaporation rates, nor how they influence the chemical reaction, something that is well known for single phase flames. For Eulerian-Eulerian models, the closure assumptions concerning these points set the same problems. For single phase turbulent flames, this second problem can be addressed with PDF's. We will study in the next chapters how the instant two-phase flow balance equations of section II enable us to get balance equations for the gas phase PDF, and the problems of closure assumptions within this equation will be discussed. We will show finally that, using a Lagrangian approach for droplets with the classical assumptions, it is possible to propose an interesting closure model for all vaporization terms. We will then get a more powerful Lagrangian-eulerian PDF model.

## 3. Unclosed joint PDF equations in the gas phase

### 3.1 Recalling the departure equations

Let us first rewrite the instant balance equation for the vapor of the liquid, based on the Eq. (10) and (12'), as :

$$\frac{\partial}{\partial t}(\rho_g f_g Y_V) + \frac{\partial}{\partial x_a}(\rho_g f_g v_g^a Y_V) = \frac{\partial}{\partial x_a}(-f_g j_{DV}^a) + m_g \sigma + \rho_g f_g w_V^{ch}$$

Using the total mass balance in the gas phase (2), it gives :

$$\rho_g f_g \frac{\partial}{\partial t}(Y_V) + \rho_g f_g v_g^a \frac{\partial}{\partial x_a}(Y_V) = \frac{\partial}{\partial x_a}(-f_g j_{DV}^a) + (1 - Y_V)m_g \sigma + \rho_g f_g w_V^{ch} \quad (15)$$

and the Fick law gives :  $-j_{DV}^a = \rho_g D_V \frac{\partial}{\partial x_a}(Y_V)$ .

The instant equations for all the chemical species  $j$  that are only present in gas phase are :

$$\frac{\partial}{\partial t}(\rho_g f_g Y_j) + \frac{\partial}{\partial x_a}(\rho_g f_g v_g^a Y_j) = \frac{\partial}{\partial x_a}(-f_g j_{Dj}^a) + \rho_g f_g w_j^{ch}$$

With the total mass balance equation and (12''), we then obtain :

$$\rho_g f_g \frac{\partial}{\partial t}(Y_j) + \rho_g f_g v_g^a \frac{\partial}{\partial x_a}(Y_j) = \frac{\partial}{\partial x_a}(-f_g j_{Dj}^a) - Y_{jg} m_g \sigma + \rho_g f_g w_j^{ch} \quad (16)$$

with  $-j_{Dj}^a = \rho_g D_j \frac{\partial}{\partial x_a}(Y_j)$  .

From the instant balance equation for the total energy of gas phase (8), it is possible to derive a balance equation for the enthalpy, defined as  $h_g = e_g + p_g/\rho_g$ . Then the equations of state are written :  $T_g = T_g(\rho_g, \text{all } Y_{jg}, h_g)$ , and :  $p_g = p_g(\rho_g, \text{all } Y_{jg}, h_g)$ , in the framework of a « perfect mixture ».

This enthalpy equation can be written in an exactly similar manner, with some simplifying assumptions that are very classically used. They are :

- we neglect the surface tension and the interfacial energy, as well as the gravitational force ;
- we neglect the kinetic energy and the power of friction forces (the flame takes place in a low velocity medium), and this leads to the possibility of neglecting the pressure fluctuations with respect to the mean pressure ;
- we assume that Fick laws are valid and that all mass diffusivities and thermal diffusivity are equal (the Lewis numbers are unity, a very classical assumption, just for the sake of simplicity of writing in this section). The general problem of the influence of species diffusivities in spray flames is too wide for being considered in this paper ;

Then, the instant balance equation for enthalpy can be written as :

$$\frac{\partial}{\partial t}(\rho_g f_g h_g) + \frac{\partial}{\partial x_a}(\rho_g f_g h_g v_k^a) = \frac{\partial}{\partial x_a}(-f_g j_{eg}^a) + f_g \frac{\partial \bar{p}}{\partial t} - j_{egs}^a n_g^a \sigma + m_g h_{gs} \sigma$$

Using the total mass balance instant equation of gas phase (2'), we obtain for  $h_g$  :

$$\rho_g f_g \frac{\partial}{\partial t}(h_g) + \rho_g f_g v_g^a \frac{\partial}{\partial x_a}(h_g) = \frac{\partial}{\partial x_a}(-f_g j_{eg}^a) - j_{egs}^a n_g^a \sigma + f_g \frac{\partial \bar{p}}{\partial t} \quad (17)$$

with  $-j_{eg}^a = \rho_g D_g \frac{\partial}{\partial x_a}(h_g)$ , due to the unity Lewis numbers.

Let us recall that the diffusive enthalpy flux is not simply due to thermal conduction, it has to take into account also the diffusion fluxes of the species.

Finally, all the informations needed for describing the chemical and thermal state of the medium can be taken in a new vector of variables  $c_i = (Y_{Vg}, Y_{jg}, \dots, h_g)$ , each of them follows, in the whole space, instant balance equations like that :

$$\rho_g f_g \frac{\partial}{\partial t}(c_i) + \rho_g f_g v_g^a \frac{\partial}{\partial x_a}(c_i) = \frac{\partial}{\partial x_a}(-\rho_g f_g D_g \frac{\partial c_i}{\partial x_a}) + J_{is}^a n_g^a \sigma + \rho_g f_g W_i \quad (18)$$

The terms  $W_i$  are representing volumic source terms in these equations : it is the chemical reaction term for  $Y_{Vg}$  and  $Y_{jg}$ , and the mean pressure term for the enthalpy.

The terms  $J_{is}$  are producing the exchange of mass or heat between the gas and liquid phases.

The term for enthalpy is  $J_{hs}^a n_g^a \sigma = -j_{egs}^a n_g^a \sigma = (\rho_g D_g \frac{\partial}{\partial x_a}(h_g))_s n_g^a \sigma$ . The enthalpy interface relation, translated from (9), gives :

$$m_g h_{gs} \sigma + (\rho_g D_g \frac{\partial}{\partial x_a}(h_g))_s n_g^a \sigma + q_l \sigma - m_g h_{ls} \sigma = 0 ,$$

meaning that the enthalpy flux entering the gas (the two first terms) equates the enthalpy flux leaving the liquid (minus the two others).

In addition, we have to take into account that, in order to be able to vaporize some liquid, the diffusive enthalpy flux  $(\rho_g D_g \frac{\partial}{\partial x_a}(h_g))_s n_g^a \sigma = -j_{egs}^a n_g^a \sigma$  has to be negative (it enters the liquid) and to be used partly to vaporize the flux  $m_g \sigma$  of the liquid, and partly to heat the remaining liquid. We can then write :  $j_{egs}^a n_g^a \sigma = m_g L_{hVs} \sigma + q_l \sigma$ , where the quantity  $L_{hVs}$  is a latent heat of vaporisation, but related to the enthalpy flux and not the conduction heat flux alone, and  $q_l$  is the heat flux received by the liquid phase, locally, per unit of interface area. Of course, we have then  $m_g h_{gs} \sigma = m_g h_{ls} \sigma + m_g L_{hVs} \sigma$ , and subtracting the previous relation from this one allows to recover the interface relation above.

Concerning the terms related to  $Y_V$ , and to the others  $Y_j$ , the interface relations (12') and (12'') can simplify their expressions.

There are three types of  $J_{is}$ , and finally we can write :

$$J_{Vs}^a n_g^a \sigma = m_g \sigma (1 - Y_{Vs}) \quad (19)$$

$$J_{js}^a n_g^a \sigma = -m_g \sigma Y_{js} \quad (19')$$

$$J_{hs}^a n_g^a \sigma = -(m_g \sigma L_{hVs} + q_l \sigma) \quad (19'')$$

Concerning  $L_{hVs}$ , it is different from  $L_{Vs}$ , as  $\rho_g D_g \frac{\partial}{\partial x_a}(h_g)$  is different from  $\rho_g D_g \frac{\partial}{\partial x_a}(C_p T_g)$ . Using  $h_g = \sum Y_i (C_{pi} T_g + h_i^0)$ , it comes that  $\frac{\partial}{\partial x_a}(h_g) = \frac{\partial}{\partial x_a}(C_p T_g) + \sum h_i^0 \frac{\partial}{\partial x_a}(Y_i)$ , and using the interface relations for all the  $Y_i$ , we obtain :  $L_{hVs} = L_{Vs} + \sum (h_i^0 Y_{is} - h_V^0)$ . There is a difference due to the composition of the gas mixture at the surface from the pure  $Y_V=1$ . The number of species that are to be considered in this vector is, at maximum, equal to N-1 if the number of species present is N. But one can diminish this number by considering only the most important species for the chemical reactions. In the simplest model of a single irreversible reaction, only vaporized fuel (V) and oxidiser (O) (which can include some inert species, like nitrogen in air) are important, and the products are not ( $Y_V + Y_O + Y_P = 1$ ). The vector  $c$  is then reduced to  $Y_V, Y_O, h$ .

### 3.2 Deriving the PDF equation

By using the instant balance equations (15), we will now derive a balance equation for the joint PDF of the random variables  $\hat{Y}_i$ , corresponding to  $c_i$ , then conditioned for the gas phase only. The derivation uses the Lundgren method<sup>32</sup>, shorter than the historical method of A.S. Monin or V.M. Ievlev, using the framework of « functionals » and characteristic functions, that E.E. O'Brien and coworkers used initially.

This method defines a « fine-grained PDF », at each point  $x$  of the considered spatial domain, and at each instant  $t$ , which is the PDF that would be associated to a single « realization » of the very many ones that are needed for the true PDF. Because, in this single realization, the value of each random variable is equal to a single (unknown) value of the corresponding physical variable, this fine-grained PDF is nothing but a Dirac peak in the multidimensional probability space, i.e. the product  $\prod_i \delta(\hat{Y}_i - c_i(x, t))$ . In the greatest generality, the physical variables  $c_i$  are those able to describe entirely the problem addressed. In our case, they would comprise the variables of the gas phase, that of the liquid phase, and that defining the interface. Then, the joint PDF that we are looking for is simply the statistical mean of the fine-grained PDF for a very large number of realizations. But now we are interested in the gas phase only, then we will consider only the mean value conditioned for the gas phase. It is defined by :

$$\bar{f}_g P_g(\hat{Y}; x, t) = \overline{f_g \prod_i \delta(\hat{Y}_i - c_i(x, t))}. \quad \text{To simplify writing, it will be noted in the following}$$

$$\prod_i \delta(\hat{Y}_i - c_i(x, t)) = \Pi(\hat{Y}) \quad (\text{not using the index } i \text{ says that all } i \text{ have to appear}).$$

It is, therefore, possible to find an equation for the PDF from the previous equations (18).

First we remark that :

$$\frac{\partial}{\partial t}(\Pi(\hat{Y})) = \sum_i \frac{\partial}{\partial \hat{Y}_i}(\Pi(\hat{Y})) \left( -\frac{\partial c_i}{\partial t} \right)$$

and similarly for the derivatives with respect to spatial coordinates.

Therefore, from (18) it is found :

$$\begin{aligned} f_g \frac{\partial}{\partial t}(\Pi(\hat{Y})) + f_g v_g^a \frac{\partial}{\partial x_a}(\Pi(\hat{Y})) = \\ - \sum_i \frac{\partial}{\partial \hat{Y}_i}(\Pi(\hat{Y})) \left( \frac{1}{\rho_g} \frac{\partial}{\partial x_a}(\rho_g f_g D_V \frac{\partial c_i}{\partial x_a}) + \frac{1}{\rho_g} J_{is}^a n_g^a \sigma + f_g W_i \right) \end{aligned}$$

After multiplying by the gas density, and using the continuity equation (2) for the gas phase, the equation becomes :

$$\begin{aligned} \frac{\partial}{\partial t}(\rho_g f_g \Pi(\hat{Y})) + \frac{\partial}{\partial x_a}(\rho_g f_g v_g^a \Pi(\hat{Y})) - m_g \sigma \Pi(\hat{Y}) = \\ - \sum_i \frac{\partial}{\partial \hat{Y}_i}(\Pi(\hat{Y})) \left( \frac{\partial}{\partial x_a}(\rho_g f_g D_V \frac{\partial c_i}{\partial x_a}) + J_{is}^a n_g^a \sigma + \rho_g f_g W_i \right) \end{aligned}$$

The average of this relationship must now be taken.

As defined above, the density is a function of the variables  $c_i$  (and of  $\bar{p}$ , non random parameter). The average of the product  $\rho_g(c)f_g \Pi(\hat{Y})$  is nothing other than  $\rho_g(\hat{Y})\bar{f}_g P_g(\hat{Y})$ . Likewise, the average of  $\rho_g f_g W_i(c) \Pi(\hat{Y})$  is  $\rho_g(\hat{Y})W_i(\hat{Y})\bar{f}_g P_g(\hat{Y})$ . In addition, defining the velocity fluctuation as  $v_g^a = v_g^a - \bar{v}_g^a$ , the following is obtained for the average of the above equation :

$$\begin{aligned} \frac{\partial}{\partial t}(\rho_g(\hat{Y})\bar{f}_g P_g(\hat{Y})) + \frac{\partial}{\partial x_a}(\rho_g(\hat{Y})\bar{f}_g \bar{v}_g^a P_g(\hat{Y})) = \\ \frac{\partial}{\partial x_a}(-\overline{\rho_g f_g v_g^a \Pi(\hat{Y})} + \overline{m_g \sigma \Pi(\hat{Y})}) \\ - \sum_i \frac{\partial}{\partial \hat{Y}_i} \left\{ \overline{\frac{\partial}{\partial x_a}(\rho_g f_g D_V \frac{\partial c_i}{\partial x_a}) \Pi(\hat{Y})} + \rho_g(\hat{Y})\bar{f}_g W_i(\hat{Y}) P_g(\hat{Y}) + \overline{J_{is}^a n_g^a \sigma \Pi(\hat{Y})} \right\} \end{aligned}$$

The first term under the summation gives mainly the so-called « micro-mixing » term (when the molecular diffusivity is low), which destroys the fluctuations. It can be broken down, and after some algebra, it is found :

$$\begin{aligned} - \sum_i \frac{\partial}{\partial \hat{Y}_i} \left\{ \overline{\frac{\partial}{\partial x_a}(\rho_g f_g D_V \frac{\partial c_i}{\partial x_a}) \Pi(\hat{Y})} \right\} = \\ \frac{\partial}{\partial x_a}(\overline{\rho_g D_V \frac{\partial}{\partial x_a}(\bar{f}_g P_g)} + \overline{\rho_g D_V n_g^a \sigma \Pi(\hat{Y})}) - \sum_i \sum_j \frac{\partial}{\partial \hat{Y}_i} \frac{\partial}{\partial \hat{Y}_j} \overline{(\rho_g D_V \frac{\partial c_i}{\partial x_a} \frac{\partial c_j}{\partial x_a} f_g \Pi(\hat{Y}))} \end{aligned}$$

At the end of these calculations, we have neglected fluctuations in coefficient  $\rho_g D_V$ , or rather its correlation with fluctuations of factor  $\frac{\partial}{\partial x_a}(f_g \Pi(\hat{Y}))$ .

Finally, the PDF equation is :

$$\begin{aligned} \frac{\partial}{\partial t}(\rho_g(\hat{Y})\bar{f}_g P_g(\hat{Y})) + \frac{\partial}{\partial x_a}(\rho_g(\hat{Y})\bar{f}_g \bar{v}_g^a P_g(\hat{Y})) = \\ \frac{\partial}{\partial x_a}(\overline{\rho_g D_V \bar{f}_g} \frac{\partial}{\partial x_a}(P_g) - \overline{\rho_g f_g v_g^a \Pi(\hat{Y})} + \overline{\rho_g D_V}(P_g \frac{\partial \bar{f}_g}{\partial x_a} + \overline{n_g^a \sigma \Pi(\hat{Y})})) \\ - \sum_i \sum_j \frac{\partial}{\partial \hat{Y}_i} \frac{\partial}{\partial \hat{Y}_j} \overline{(\rho_g D_V \frac{\partial c_i}{\partial x_a} \frac{\partial c_j}{\partial x_a} f_g \Pi(\hat{Y}))} - \sum_i \frac{\partial}{\partial \hat{Y}_i}(\rho_g(\hat{Y})W_i(\hat{Y})\bar{f}_g P_g(\hat{Y})) \\ - \sum_i \frac{\partial}{\partial \hat{Y}_i}(\overline{J_{is}^a n_g^a \sigma \Pi(\hat{Y})} + \overline{m_g \sigma \Pi(\hat{Y})}) \quad (20) \end{aligned}$$

### 3.3 The « closure » problems

This equation is not usable as it is, because it involves in the right hand side several mean values that depend on other variables than those involved in the PDF studied. The equation is « unclosed », and we need now to discuss some « closure assumptions » for these terms. Nevertheless, as it is the case for single phase reacting flows, the term representing the mean reaction rate appears here in exact form, because it does not depend on fluctuating physical variables, but only on all the random variables that we have considered. Before discussing the possibilities for closures, we have to give the physical meaning of each of these terms, successively.

The first term on the right hand side of (20) is a « diffusion term of the PDF » in the physical space, and there are three contributions : one is due to molecular diffusion, the second is due to turbulence (with the velocity fluctuations), and a new contribution due to the presence of



interfaces appears. The molecular diffusion term is in closed form, but it is very often negligible with respect to the turbulence contribution, when the Reynolds number is sufficiently high. The turbulent dispersion flux is not closed, it would be such if the Joint PDF involving a random variable corresponding to the gas velocity was used. However, this is a very well known problem. The third contribution is new, being justified by the possibility of presence of interface at the studied point for some realizations. It is difficult to interpret the term  $\overline{n_g^a \sigma \Pi(c)}$ . This has to be studied further. Its contribution being proportional to the molecular diffusivity, it is probably of the same order of magnitude as the molecular diffusion term.

The other terms on the right hand side, which display partial derivatives with respect to all random variables, represent a transport of probability in the random variables space. It is a production term for the fluctuations (positive or negative), thickening or narrowing the PDF shape. There are three parts :

- the first part is the classical term called « micromixing », narrowing the PDF towards a Dirac peak that would occur when all the fluctuations are dissipated. It is already well known for PDF's in single phase flows. In single phase gas flames, the study of such closure assumptions has needed a large amount of work. We will examine in a next section if, and how, the assumptions have to be reexamined for two phase flames.

- the second part is the contribution of the chemical reactions, and does not need any closure assumption. For the enthalpy, the factor  $\frac{\partial}{\partial t}(\bar{p})$  is very often negligible, and anyway is not a fluctuating quantity.

- the last part is due to mass exchange between gas and liquid, and is again splitted in three types of terms, that are coming from (19), (19'), and (19'') :  $\overline{j_{ls}^a n_g^a \sigma \Pi(\hat{Y})}$ . These three types of exchange terms involve  $m_g \sigma$ , and need closure assumptions. Indeed  $m_g$  is given by the relation (13), namely  $m_g = -j_{DV}^a n_g^a / (1 - Y_{Vsat}(T_s, p_s))$ , where  $-j_{DV}^a = \rho_g D_V \frac{\partial}{\partial x_a}(Y_V)$ , and it is seen that the normal gradient of  $Y_V$  on the interface is needed. However, the PDF considered is a one point PDF, it says nothing on this gradient, we have to build a formula involving an approximation of this gradient. In addition,  $\sigma$  and the liquid phase quantities  $T_s, p_s$  are not known because we have not yet considered the liquid phase, we have to do that in a simplified manner.

Finally, there is also a last term due to the exchange of total mass between gas and liquid phases :  $\overline{m_g \sigma \Pi(\hat{Y})}$ , equally to be studied and closed. It has to be remarked that the equation (20) does not concern the multidimensional PDF  $P_g(\hat{Y})$  itself, whose integration on  $\hat{Y}$  space is always unity, but concerns the product  $\bar{f}_g P_g(\hat{Y})$ , which, if integrated on this space, gives the « gas probability of presence » itself,  $\bar{f}_g$ , called also the « mean gas volume fraction ». The last term is due to that.

As expected, the integration of (20) on the random variables space has to give a balance equation for  $\bar{f}_g$ . We can verify it, recalling that  $\int \rho_g(\hat{Y}) \bar{f}_g P_g(\hat{Y}) d\hat{Y} = \bar{\rho}_g^* \bar{f}_g$ , and that the

integrals of all terms of the type of  $\sum_i \frac{\partial}{\partial \hat{Y}_i}(\dots)$  will vanish, because they will be finally

proportional to the values of the PDF on each boundary of the integration domain in the random space, and the PDF is always zero on the boundaries. We obtain :

$$\begin{aligned} \frac{\partial}{\partial t}(\bar{\rho}_g^* \bar{f}_g) + \frac{\partial}{\partial x_a}(\bar{\rho}_g^* \bar{f}_g \bar{v}_g^a) &= \frac{\partial}{\partial x_a} \left( - \int \rho_g f_g v_g'^a \Pi(\hat{Y}) d\hat{Y} + \bar{\rho}_g D_V \left( \frac{\partial \bar{f}_g}{\partial x_a} + \int \bar{n}_g^a \sigma \Pi(\hat{Y}) d\hat{Y} \right) \right) \\ &\quad + \int \overline{m_g \sigma \Pi(\hat{Y})} d\hat{Y} \end{aligned}$$

The integrals here are just the full mean values of the terms considered, with all the fluctuating variables involved, not only the  $c_i$ 's, but also,  $v_g^a, n_g^a, m_g, \sigma$ . Then, we can write :

$$\int \overline{\rho_g f_g v_g'^a \Pi(\hat{Y})} d\hat{Y} = \overline{\rho_g f_g v_g'^a} = 0, \text{ and } \int \overline{n_g^a \sigma \Pi(\hat{Y})} d\hat{Y} = \overline{n_g^a \sigma} = -\frac{\partial \overline{f_g}}{\partial x_a}, \text{ and finally :}$$

$\int \overline{m_g \sigma \Pi(\hat{Y})} d\hat{Y} = \overline{m_g \sigma}$ . The classical equation (14) is recovered. The mean intrinsic gas density can be calculated knowing the PDF :  $\overline{\rho_g^*} = \int \rho_g(\hat{Y}) P_g(\hat{Y}) d\hat{Y}$ .

#### 4/ Proposals for closure models

##### 4.1 Turbulent dispersion and micromixing modeling

###### A/ Turbulent dispersion modeling

The turbulent contribution in the « diffusion term of the PDF » which appears on the right hand side of (20) is generally represented by a law containing a turbulent diffusion coefficient (which can be calculated with the turbulent kinetic energy  $k$  and its dissipation rate  $\varepsilon$ , these quantities being relative to the gas phase only,  $D_t \approx k^2/\varepsilon$ ) by simple analogy with the turbulent diffusion of any mean quantity. Therefore, the diffusion term is first expressed in terms of the conditional average of the velocity fluctuation  $v_g'^a$  :

$$\overline{-\rho_g f_g v_g'^a \Pi(\hat{Y})} = -\overline{\rho_g} \overline{f_g} \langle v_g'^a | Y(x,t) = \hat{Y} \rangle P_g(\hat{Y})$$

And the conditional velocity is then modeled using turbulent diffusion,

$$\langle v_g'^a | \hat{Y} \rangle P_g(\hat{Y}) = -D_t \frac{\partial P_g}{\partial x_a}$$

which finally gives

$$\overline{-\rho_g f_g v_g'^a \Pi(\hat{Y})} = \overline{\rho_g} \overline{f_g} D_t \frac{\partial P_g(\hat{Y})}{\partial x_a}$$

Modeling of turbulent dispersion can be avoided if an interest is taken in the joint PDF for composition and velocities, as was done by S.B.Pope<sup>13</sup> for reactive single phase medium, but the challenge is much more complicated since this approach then implies the use of Monte-Carlo simulations.

###### B/ Micromixing modeling

Micromixing modeling has been devoted a lot of attention over the years, from the pioneering works of Curl<sup>7</sup>, Villermaux and Devillon<sup>8</sup> and Dopazo and O'Brien<sup>3</sup> to more refined models developed recently (see the review paper by Meyer and Jenny<sup>33</sup> for instance). These three models are usually referred to as, respectively, the CD (Coalescence and Dispersion), IEM (Interaction by Exchange with Mean) and LMSE (Linear Mean-Square Estimation) models. The CD was originally developed to describe mixing of colliding droplets with different concentrations in a two-liquid suspension. Consequently, Curl's model can be easily integrated into a stochastic particle framework as used for PDF simulations. In each time step, particle pairs are randomly selected and their individual concentrations set to their mean value. The two other models are based on the fact that molecular mixing reduces scalar fluctuations and draws instantaneous scalar values towards their local mean. While the IEM model correctly predicts that the variance of an inert scalar decays in homogeneous turbulence, it is not able to describe the evolution of the scalar PDF appropriately, i.e. the shape of the PDF is preserved and it does

not approach a Gaussian distribution. To overcome this difficulty, the mapping closure model was proposed later on by Chen et al.<sup>34</sup>, 1989 (see also Pope<sup>35</sup>, 1991). Interaction by exchange with the mean is the simplest mixing model. In stationary homogeneous turbulence, the scalar values of the particles relax at a constant rate towards the local scalar mean as :

$$\frac{dc_i}{dt} = -\frac{C}{2\tau}(c_i - \tilde{c}_i)$$

where  $C$  is a parameter which is often considered as a constant and  $\tau$  is a characteristic time scale for the turbulence that creates mixing. So that the micromixing term then writes

$$-\sum_i \sum_j \frac{\partial}{\partial \hat{Y}_i} \frac{\partial}{\partial \hat{Y}_j} (\rho_g D_V \frac{\partial c_i}{\partial x_a} \frac{\partial c_j}{\partial x_a} f_g \Pi(\hat{Y})) = + \sum_i \frac{\partial}{\partial \hat{Y}_i} (\bar{\rho}_g(\hat{Y}) \bar{f}_g \frac{C}{\tau} (Y_i - \hat{Y}_i) P_g(\hat{Y})) \quad (21)$$

However, despite its simple formalism, quite a few questions are embedded within such simple mixing models, which can be summarized as : i) what is the value of  $C$ , and is this value a constant or does it depend on the particular situation under concern (Reynolds number value, combustion regime, and so on) and, ii) what are the physical phenomena which are embedded within the time scale  $\tau$  (or its equivalent in terms of the frequency  $\omega=1/\tau$ ) ? It has been first assumed that  $\tau$  is just equal to (or directly related to)  $k/\varepsilon$ , but there is sufficient proof showing that this is far from being always a sound approximation. In particular since, for any scalar quantity such as  $c_i$ , a specific time scale defined by  $\tau_{c_i} = \frac{\overline{c_i'^2}}{\chi_{c_i}}$  (where  $\chi_{c_i}$ , the equivalent of  $\varepsilon$  for

$k$ , is the dissipation rate of  $c_i'^2$ ) is associated to its turbulent mixing properties. How to relate  $\tau$  to both  $k/\varepsilon$  and  $\tau_{c_i}$  is still an open question (see Stöllinger and Heinz<sup>36</sup>, 2008, for instance). This may in particular depend on the combustion regime, on the presence of flamelets in the case of premixed combustion, .... However, as we will see in section 5.1, the basic model using  $\tau=k/\varepsilon$  performs quite well for the simulation of a turbulent spray flame carried out by H-W. Ge and E. Gutheil<sup>37</sup>.

In addition, the way these models may have to be adapted for a medium with liquid parcels or droplets must also be analyzed since droplets are likely to both modify the way turbulence is produced or dissipated and impact velocity spectra. Turbulence modulation by droplets may depend on the particle number and size, their volumic concentration, their density, or their response time (or associated Stokes number). To analyze turbulence modulation, direct numerical simulations of turbulent flows transporting solid particles are now available, at least for rather small Reynolds numbers and for canonical situations. Among others, Boivin et al.<sup>38</sup> (1998) have investigated the modulation of isotropic turbulence by particles, for dilute flows in which particle volume fractions and inter-particle collisions are negligible. Their results show that particles increasingly dissipate fluid kinetic energy with increased loading, with the reduction in kinetic energy being relatively independent of the particle relaxation time. Viscous dissipation in the fluid decreases with increased loading and is larger for particles with smaller relaxation times (the particle Stokes number was varied between 1 and about 10). Fluid energy spectra show that there is a non-uniform distortion of the turbulence with a relative increase in small-scale energy, with this increase of the high-wave number portion of the fluid energy spectrum being attributed to transfer of the fluid-particle covariance by the fluid turbulence. More recently, Richter<sup>39</sup> (2015) and Wang and Richter<sup>40</sup> (2019) have studied in a more detailed way the two-way coupling between the turbulent flow and particles (with Stokes numbers varying between 1 and about 100) through the analysis of their influence of the regeneration cycle in the near-wall region of a planar Couette flow. When the Reynolds number is sufficiently large for turbulence to be completely established, turbulent kinetic energy ( $k$ )

budgets show that two simultaneous effects of particles occur. On the one hand, the mere presence of particles causes a reduction of  $k$  production across nearly the entire wavenumber range, where the particle Stokes number only determines the magnitude of this reduction, while, on the other hand, the direct energy exchange term between the carrier and dispersed phases is relatively small in magnitude compared to changes in production. However, its location in wave number space is highly dependent on the Stokes number. In addition, the individual components of  $k$  (namely, the streamwise and wall-normal velocity fluctuations) are affected differently by the presence of inertial particles. The effect of particles was also analysed on the laminar-turbulent transition, for a Reynolds number just above its critical value, which allowed to investigate their influence on the large-scale vortices which generate wall turbulence. Two limiting behaviours are found with increasing particle inertia: low-inertia particles trigger the laminar-to-turbulent instability whereas high-inertia particles tend to stabilize turbulence due to the extra dissipation induced by particle–fluid coupling. Furthermore, it is found that the streamwise coupling between phases is the dominant factor in damping of the turbulence and is highly related to the spatial distribution of the particles. Also, the presence of inertial particles does not alter the periodic nature of the near-wall cycle of turbulence generation nor the relative length of each of its sub-steps. Instead, high-inertia particles greatly weaken the large-scale vortices as well as the streamwise vorticity stretching and lift-up effects, thereby suppressing the fluctuating amplitude of the large-scale streaks. The primary influence of low-inertia particles, however, is to strengthen the large-scale vortices, which fosters the near-wall turbulence generating cycle and ultimately reduces the critical Reynolds number. These findings are in good agreement with the global idea stating that « small particles » respond to the mean and fluctuation properties of the gas phase, while « large particles » only respond to the mean properties of the gas phase. But how the two-way coupling modifies the known large-scale and small-scale properties for jet or wake flows remains an open question since, as far as we are aware, such direct numerical simulations were only performed so far for channel flows or for isotropic turbulence within a cubic domain.

Anyway, the use of a  $k$ - $\varepsilon$  model is possible, with an adaptation for the momentum exchange between gas and liquid droplets. This is classically done at present time, details can be found in ref. 21.

It is also of interest to reexamine the relation between the micromixing time  $\tau$  and  $k/\varepsilon$ , taking into account the influence of vaporization of droplets. DNS studies involving the presence of liquid parcels or droplets are even scarcer, and in particular when combustion induces vaporization. Such a situation has been studied by J. Reveillon and L. Vervisch<sup>41</sup> (2000), which will be discussed later in connection with the vaporization model, but no detailed analysis of the turbulence modulation is presented. However, results reported previously by J. Reveillon et al.<sup>42</sup> (1998) deal with the same DNS but with vaporization only. The small scale mixing term itself is strongly modified by vaporization, but, finally, the mean turbulent mixing time is found again proportional to  $k/\varepsilon$ , with about 20% difference with the case without vaporization.

## 4.2 Modeling the terms due to exchanges between phases

### A/ Useful known results on droplets vaporization

We are faced with four types of terms, all connected with the quantity  $m_g \sigma$ , and all are conditioned mean values. The calculation of these mean values cannot be done from (13), because here the normal gradient appears, that the one point PDF is not able to consider. In addition, the liquid surface temperature plays, related to the fluctuations of all the liquid phase

variables, and the knowledge of  $\sigma$  requires the one of the fluctuations of the interface. How can we propose closure assumptions for them ?

In classical Lagrangian-Eulerian models, a closure assumption is adopted saying that the liquid parcels are spherical droplets of different sizes, and the Lagrangian procedure is able to find their radius, surface temperature, and number density at any « macroscopic point » (given by the numerical grid). In order to do that, it is considered that each droplet is sufficiently far from the others in such a way that studies of a single droplet in a given environment can give interesting formulas. In addition, because it was wanted to treat situations where the droplets are not all fully ignited, the vaporization (or condensation) only is considered, not each droplet surrounded by its own flamelet. We will use here the same framework for obtaining closures. There are several critics restraining the interest of this, we will discuss more this point later.

The study of the vaporization and combustion of liquid fuel droplets has been well developed since the theory for a single droplet at rest in the simultaneous pioneering studies by D.B Spalding<sup>43</sup> (1953) and G.A.E. Godsave<sup>44</sup> (1953). All of this knowledge is now used for Lagrangian-Eulerian mean models (see W.A. Sirignano<sup>45</sup> (2010)). The main results are :

The vaporized mass flow from one droplet (at rest) of radius  $r$  is :

$$\dot{M}_d = 4\pi\rho_g D_V r \text{Log}(1 + (Y_{Vs} - Y_{Vinf})/(1 - Y_{Vs})) \quad \text{with} \quad Y_{Vs} = Y_{sat}(T_s) \quad (22)$$

where  $T_{inf}$  and  $Y_{Vinf}$  are related to the bulk of gas phase (more detailed comments are provided in the paragraph just after). As  $Y_{Vs}$  has to be always smaller than 1,  $T_s$  is always smaller than  $T_b$ . In addition, the theory gives that :

$$(Y_{Vs} - Y_{Vinf})/(1 - Y_{Vs}) = C_p(T_{inf} - T_s)/L_{ve} \quad (23)$$

where  $L_{ve}$  is defined as  $L_{ve} = L_{vs} + \dot{Q}_l/\dot{M}_d$ .  $L_{vs}$  is the latent heat of vaporization and  $\dot{Q}_l$  is the heat flux received by the liquid droplet from the gaseous surrounding. This allows to calculate  $\dot{Q}_l$ , and that is necessary, in the Lagrangian part, for computing the evolution of the global temperature of the droplets, and consequently of their surface temperatures. When the heat conductivity of the liquid is high, the surface temperature is very near this global temperature. When the surface temperature of the droplet goes closer to  $T_b$ ,  $\dot{Q}_l$  decreases to zero, then  $L_{ve}$  become very close to  $L_{vs}$ . The book by W.A. Sirignano<sup>45</sup> gives also different improvements of these formulas.

We have to remark that this formula does not involve only quantities related to the interface, but also quantities related to the bulk of gas phase, here  $T_{inf}$  and  $Y_{Vinf}$ . These quantities do represent in fact  $Y_{Vg}$  and  $T_g$  far from the droplets (far with respect to its radius), at maximum half the mean distance between two droplets. It can be easily understood that the vaporization rate has to depend of the properties of the gaseous environment, and the difference of  $Y_V$  between the droplet surface and the neighbouring gas represents globally the normal gradient at the interface. This dependance will lead the vaporization rate to fluctuate when the droplet travels in a fluctuating turbulent medium, it is an essential ingredient of the model.

In using this formula in the Lagrangian-Eulerian model, we have to adapt it in such a way that it will include only variables that the model can calculate, and we have to « interpret » what are the quantities  $Y_{Vinf}$  and  $T_{inf}$ . The assumption usually done is simply that these quantities can be replaced by the mean values of  $Y_{Vg}$  and  $T_g$ , noted  $\bar{Y}_V$  and  $\bar{T}$ , that are obtained by using a mean Eulerian balance equation.

With this assumption, the influence of the fluctuations of the environment are cancelled. When the droplet radius is small with respect to the scale of the macroscopic spray,  $Y_{Vinf}$  and  $T_{inf}$  are realistically approximated by the values in the bulk of the gas phase. If we don't know the fluctuations, using their mean values is necessary, but if we can know these fluctuations through  $P_g(\bar{Y}_V, \bar{Y}_O, \bar{T})$ , we can afford the influence of the fluctuations.

Concerning the values of  $T_s$  and  $r$ , which fluctuate in one point because several droplets can be passing through, they can be given directly by the Lagrangian tracking. In fact, we can obtain

a distribution function of the number of droplets,  $N(r, T_s)$ , at each position, i.e. within each small volume in the numerical grid.

The formula (22) given above is very often modified by an additional factor taking into account the fact that the droplet moves with respect to the surrounding fluid. This factor (often called the Sherwood number) is written in terms of a « droplet Reynolds number ». One question that we have to face with is what kind of velocity we have to use for this Reynolds number in a turbulent flow. Probably the simplest approximation is using the mean gas velocity minus the mean droplet velocity. We know the mean gas velocity from the Eulerian mean balance equation, and the droplet velocity from the Lagrangian tracking, including the random dispersion by gas turbulence. But if the mean liquid velocity is quite smaller than the rms of the droplets velocities, the formula has to be adapted. Probably the most powerful way to date for handling the effect of droplets velocity fluctuations on their vaporization rate is the model of Bini and Jones<sup>46</sup> (2009).

#### B/ Needed closures for vaporization terms

The result of the above discussion gives closures for all the interesting terms, in agreement with the previous modeling attempts in a Lagrangian-Eulerian framework (that assumes usually dilute or very dilute sprays).

Let us do that first for the total mean volumetric mass flux :  $\overline{m_g \sigma \Pi(\hat{Y})}$ .

With the assumptions  $Y_{Vinf} = \widehat{Y}_V$ , we obtain :

$$\overline{m_g \sigma \Pi(\hat{Y})} = (\iint N(r, T_s) M_d(r, T_s, \widehat{Y}_V) dr dT_s) P_g(all \hat{Y})$$

The distribution  $N$  contains the possibility to calculate  $\sigma = 4\pi r^2 N$ , but  $4\pi r^2$  is contained in  $M_d$  and consequently  $\sigma$  does not appear. We could calculate also  $\overline{\sigma}$ , but it is without interest here.

Considering that the values of  $\widehat{Y}_V$  in different realizations at the considered point can represent the possible values of  $Y_{Vinf}$ , it is very simple to propose an improved closure for this term :

$$\overline{m_g \sigma \Pi(\hat{Y})} = (\iint N(r, T_s) M_d(r, T_s, \widehat{Y}_V) dr dT_s) P_g(\hat{Y}) \quad (24)$$

For the production of  $Y_V$  fluctuations due to vaporization, we have :

$$\overline{m_g (1 - Y_{Vs}) \sigma \Pi(\hat{Y})} = (\iint N(r, T_s) M_d(r, T_s, \widehat{Y}_V) (1 - Y_{Vs}) dr dT_s) P_g(\hat{Y}) \quad (25)$$

For the species that are not coming from the liquid (and cannot be adsorbed in it), the main one is the oxydizer, it is found :

$$\overline{-m_g Y_{Os} \sigma \Pi(\hat{Y})} = -(\iint N(r, T_s) M_d(r, T_s, \widehat{Y}_V) Y_{Os} dr dT_s) P_g(\hat{Y}) \quad (26)$$

For the term due to the heat transfer between phases, the situation is similar. We have seen from (19'') :  $\overline{J_{hs}^a n_g^a \sigma \Pi(\hat{Y})} = -\overline{m_g \sigma (L_{vs} + q_l/m_g) \Pi(\hat{Y})}$ .

If the flux heating the liquid is negligible, this term gives simply :

$$\overline{-m_g L_{vs} \sigma \Pi(\hat{Y})} = (\iint N(r, T_s) M_d(r, T_s, \widehat{Y}_V) L_{vs} dr dT_s) P_g(\hat{Y})$$

In the general case, writting  $L_{vs} + q_l/m_g = L_{ve}$ , one gets from (23) :

$$\overline{-m_g L_{ve} \sigma \Pi(\hat{Y})} = (\iint N(r, T_s) M_d(r, T_s, \widehat{Y}_V) L_{ve} (Y_{Vs}(T_s), \widehat{Y}_V, T_g(\widehat{Y}_V, \hat{h})) dr dT_s) P_g(\hat{Y}) \quad (27)$$

The relation  $T_g(\widehat{Y}_V, \hat{h})$  comes from the thermal equation of state for the gas phase.

The vaporization modifies the shape of  $P_g(\widehat{Y}_V, \widehat{Y}_O, \hat{h})$ , due to the fact that the source of vapor is located in a particular sub-domain of the total domain possible for the random variables. For

instance, in the idealized case where all the droplets do have the same temperature, the vaporization source term would be localised where  $\widehat{Y}_V = Y_V(T_s)$ ,  $\widehat{h} = h(T_s)$ ,  $\widehat{Y}_O$  being very small (this is justified by the combustion around the droplet). Then the PDF will remain always localized below this value, because the micro-mixing term push the PDF towards the mean value, which will always be smaller, due to combustion. If we look at (22), we see that the vaporization term for  $\widehat{Y}_V$  will display as a factor  $\text{Log}(1 + (Y_{Vs} - \widehat{Y}_V)/(1 - Y_{Vs}))$ , and that will produce a transfer of probability towards  $Y_{Vs}$ . That will produce a thickening of the PDF around the mean value  $\widehat{Y}_V$ , in opposition to the narrowing effect of the micro-mixing term.

From the resulting PDF equation, it is possible to find the balance equations for the mean values of the physical variables associated to the random variables. For instance, for  $Y_V$ , it suffices to multiply the PDF equation by  $\widehat{Y}_V$  and to integrate over the whole domain of the random variables. The usual equation used in the Eulerian part of the Lagrangian–Eulerian model is obtained. But calculating :  $\overline{Y}_V = \int \widehat{Y}_V P_g(\widehat{Y}) d\widehat{Y}$ , or  $\overline{\rho_g^* Y_V} = \int \rho_g(\widehat{Y}) \widehat{Y}_V P_g(\widehat{Y}) d\widehat{Y}$ , is even simpler.

## 5. Some implications and perspectives

### 5.1 Some connected other recent PDF studies

It is interesting to compare the study presented here to the recent paper interested in fluctuations in spray flames.

The study by Zhu et al.<sup>20</sup> (2000) is the first considering generally the basic joint PDF of all interesting random variables, i. e. for the gas phase as well as for the liquid phase. The basic equation for this PDF is found, which can be restricted to the gas phase PDF. It takes correctly into account the probability of presence of gas. The study does not uses explicitly the interface balance relations, and particularly that for the vaporizing species. An additional random variable  $\beta$  is introduced, that we dont need here because the velocity of the interface is directly found with the interface relation of the vaporizing species. The last term, with no derivatives, related to the exchange of mass between gas and liquid, appears only if the PDF is conditioned for the gas phase, as it is in our case. The study describes also several different PDFs, obtained from the basic one, but no attempt for closure is discussed.

The PDF of a « mixture fraction » (defined from the carbon atom in all the species involved) for the gas phase in a spray flame has also been studied by H-W. Ge and E. Gutheil<sup>37</sup> (2008), and the joint PDF of mixture fraction and enthalpy is equally considered. E. Gutheil and coworkers have been studying sprays flames since several years (see, e.g. C. Hollman and E. Gutheil<sup>47</sup>, 1998), using counterflow flamelets in a mist of tiny droplets, and the PDF's of mixture fraction and scalar dissipation are needed, jointly with a library of precalculated counterflow laminar flamelets for computing the mean values of any chemical species. Better than using simply a  $\beta$ -function shape for the PDF of mixture fraction, this PDF is calculated, and even a joint PDF of mixture fraction and enthalpy is found useful for improve the predictions. The PDF of scalar dissipation is classically given as Lognormal. The balance equation of the joint PDF is given, but without taking into account the probability of presence of gas, the spray studied is very dilute, and also the last term due to mass feeded in the gas phase by vaporization is lacking. It is solved classically with « notional particles », and uses the small scale mixing model IEM (equivalent to the LMSE model of C. Dopazo and E.E. O'Brien<sup>3</sup>). But the expressions of the terms due to vaporization in the PDF equation are not described, and nothing is said about an influence of the fluctuations of mixture fraction and enthalpy in the gas phase, as we propose here. The liquid phase is described classically with the

Lagrangian tracking of the droplets, giving source terms for the Eulerian equations for mean quantities describing the gas phase.

In the same period, R.W. Bilger and coworkers have also considered the PDF of mixture fraction in gas phase, in order to take it as a basis for a version of their « Conditional Moments Closure ». M. Mortensen and R.W. Bilger<sup>48</sup> (2009) have also written this PDF, using the paper by Zhu et al.<sup>20</sup> (2000). They present the possibilities of closure, based on the interface relations, as it is here, and propose to use for that the classical knowledge of vaporizing or burning droplets. It is noted that the presence of droplets will give in the close gas large gradient of fuel mass fraction, and hence of mixture fraction, and that will modify the mean scalar dissipation. Their approach is clearly in the same direction as ours, but no explicit formulas for closure are given. This study is further developed in the paper by G. Borghesi et al.<sup>49</sup> (2011), that gives results of calculations of a spray flame already experimentally analysed. However, they use a presumed form of the PDF, as Hollman and Gutheil<sup>47</sup> do, but with a modified  $\beta$ -function that is discussed in connection with droplets vaporization. The full model uses Eulerian CMC model, with mean and variance of mixture fraction equations, and Lagrangian tracing of droplets. Vaporisation terms in the Lagrangian equations as well as in the Eulerian equations for mean mixture fraction and variance are closed as it is classical in the Lagrangian-Eulerian models, neglecting the fluctuations of the mass fractions, mixture fraction and temperature in the gas phase. It is necessary then to neglect some correlation terms in the equations for CMC and mixture fraction variance.

J. Reveillon and L. Vervisch<sup>41</sup> (2000) study the PDF of mixture fraction in the gas phase, using a DNS of a gas phase in which many discrete droplets are vaporizing in a decaying isotropic homogeneous turbulent medium (in fact, the droplets were grouped in 9 clusters of 11,000 droplets, statistically distributed in the flow field). Of course, this is not a pure DNS, because the dispersion and the vaporization of the droplets are based on models, which are simpler versions of the classical models used in Lagrangian-Eulerian approaches. The results of calculations are processed giving the vaporization terms in the mean mixture fraction equation, and equation for the variance of its fluctuations. The dissipation rate of this variance is also obtained. As it is expected, there is noticeable increase of the variance due to vaporisation, but also of the dissipation rate, clearly dependant on the vaporization time. When the time evolves, the variance increases first due to vaporization, and then obviously decreases, when the droplets disappear and vaporization diminishes. Their purpose is to study the mean vaporization rate conditioned by the value of mixture fraction, and they propose an approximate formula found from the simulations. It is possible to avoid this additional assumption with our approach using the joint PDF of species mass fractions and temperature. We can remark that the vaporization rate used in the simulation is strongly simplified. The formula (22) is used but with  $B_Y = (Y_{Vs} - Y_{Vinf})/(1 - Y_{Vs})$  given constant, for each simulated case. Then  $Y_{Vinf}$  cannot vary with time, and there is no coupling with its possible fluctuations. It would be interesting to pursue the study with realistic formulas for  $B_Y$ .

The more recent work by J.C.K. Tang et al.<sup>50</sup> presents a comparison of a Monte Carlo simulation of the joint PDF of temperature and mixture fraction in a non-premixed temporally evolving plane spray jet flame with a DNS (in the same sense as in the Reveillon et al.'s study). The PDF is obtained from a usual Monte Carlo simulation of « notional particles » (like in Ge and Gutheil<sup>37</sup>'s paper cited previously), with a mesh size larger than the DNS mesh (about 30 times). The DNS uses a classical Lagrangian-Eulerian simulation, not resolving the droplets, but the Eulerian mesh is very fine (only three times the initial droplets diameters). This very interesting work, with two different values of the Damkohler number, compares two different micro-mixing models, and the IEM model looks not really satisfactory. But its main interest is that the manner used for introducing the mass vaporized from the droplets in the « notional particles » is also studied. The vaporized mass is here given exactly by the DNS. It is shown



that three different ways of doing give noticeable differences in the PDF shape. But one seems better considering this shape, and another seems better considering conditioned temperature profiles.

The present study shows that using a classical stochastic Monte Carlo model for spray flame simulation is not as simple as it appears. The way to introduce the vaporized species has to be studied further, and a JPDP equation can help to better know what is the real phenomenon.

## 5.2 Discussions and improvements

### A/ On the influence of gas phase fluctuations on the vaporization

It is clear that the fuel vaporized from the droplets is released in the gas phase at the concentration corresponding to the saturation at the surface temperature of the droplets. In principle, the different droplets can have different sizes and then different surface temperature, but at the same location in the flame their temperatures are certainly not strongly dispersed. In consequence, we can expect that the vaporization influences the shape of the PDF, producing one maximum close to the saturation value corresponding to the mean temperature of the droplets. It seems also clear that if the vaporization is much slower than the small scale mixing, this mixing will arrange fastly this maximum, but when the vaporization rate is sufficiently high (for a sufficiently large number of droplets by unity of volume, and a sufficiently high individual rate of vaporization  $1/t_{vap} = 3M_d/(4\pi\rho_l r^3)$ ), this maximum is expected to appear. With this in mind, it appears difficult that expressions of  $M_d$  cancelling the dependance on  $\widehat{Y}_V$  could be able to recognize that the fuel vapor is released preferentially at certain values of  $\widehat{Y}_V$ . The assumptions that  $Y_{Vinf} = \widetilde{Y}_V$ , or, more evidently, that  $B_Y = cst$ , seem to be unsuitable for showing this effect.

We can remark that  $M_d$ , given by the expression (22), becomes zero when  $Y_{Vinf} = Y_{Vs}$ , and this implies that the related term for production of fluctuations of  $\widehat{Y}_V$ , given by (25), has the same property. Even when  $Y_{Vs} - \widehat{Y}_V$  is small, the result of the logarithm is proportional to this difference. On the other hand, the micromixing term, which act to push any  $\widehat{Y}_V$  to the mean value  $\widetilde{Y}_V$ , has been in the first period simply modelled as proportional to  $\widehat{Y}_V - \widetilde{Y}_V$ . This seems to indicate that the term (25) will give effectively that the vaporisation pushes the probability towards  $Y_{Vs}$ . Of course, the two terms of micromixing and vaporization will play at the same time, and with variable strengths :  $1/t_{vap}$  and  $1/t_{tur}$ . The shape of the PDF will depend on their ratio.

We could think that there is a certain realism in assuming  $Y_{Vinf} = \widetilde{Y}_V$ , because  $Y_{Vinf}$  may be seen as an average value of  $Y_V$  on a certain spherical surface at some distance of the droplet, but this average must depend on the droplet radius, compared to the integral length scale of the  $Y_V$  fluctuations. For the smallest droplets, we can expect that this partial average do show again fluctuates, and for largest droplets it is closer to the statistical mean value. This picture leads to imagine a certain filtering formula for  $Y_{Vinf}$ , but the picture is quite fuzzy, and the point has to be studied further.

It can be added finally that when the fluctuations of  $Y_{Vinf}$  are taken into account for the PDF calculation, it is consistent to take them into account, inversely, for the calculation of the vaporization rates of the droplets in the Lagrangian part of the model. Then, as the PDF of these fluctuations is known, the vaporization rate for each given droplet can be calculated with a random value of  $Y_{Vinf}$ , chosen in agreement with the calculated PDF.

## B/ Droplet vaporization or droplet combustion ?

We have said that the droplets in this model have been considered not having a spherical flamelet around them. This is justified when thinking at the regime called « group combustion », and the simulation of Reveillon and Vervisch do explicitly consider groups of 11 000 droplets. But in a real spray flame, it is not known a priori if one droplet is surrounded by a full flamelet (or more precisely a reaction sheet), or several droplets, or a group of many. It is also possible in regions of the brush on the rich side of the flame brush (or close to the centerline for fig.1d), that a quasi spherical reaction sheet can enclose a pocket of oxidant gases. The pictures of spray flames of figures 1 seems to indicate that single burning droplets are present only in the periphery of the spray flame, and only for the biggest droplets. For obtaining a quite general model, it would be interesting to consider also this case, for which the standart theory gives different formulas from (23) and (24), see W.A. Sirignano<sup>45</sup> (2010). This does not appears very complicated, with the general classical knowledge related to droplet combustion. We have seen that the example of a single chemical reaction can be described with only the three variables  $Y_V$ ,  $Y_O$  and the enthalpy  $h$ . The enthalpy being defined including the enthalpy of formation of these three species (and the products can be assumed having no enthalpy of formation, at the chosen temperature of reference), it can be used for droplets vaporizing or droplets burning (with the simplifying assumptions we have listed above, the main being the unity of Lewis numbers). Similarly, the « conserved scalar »  $Z = Y_V - Y_O/\nu_s$ , where  $\nu_s$  is the stoichiometric massic coefficient of the reaction considered, is described by equations without reaction term. This is a kind of « mixture fraction », but varying between  $-1/\nu_s$  and 1. Then, the classical theory of vaporizing/burning droplet can be rewritten using  $h$  and  $Z$ , and giving two formulas for replacing (23) and (24), that can translated at the end in terms of variables  $Y_V$ ,  $Y_O$  and  $h$ . These formulas can be used for droplet vaporization or droplet combustion, but we need to prescribe the value of oxidiser at the surface of the droplet,  $Y_{Os}$ , in a different manner. If the droplet is vaporizing, without any spherical flame around, the oxidizer and the products (existing in the far field of the droplet considered due to far reaction sheets), will represent  $1 - Y_{Vs}$ , with the same proportions as in the far gas phase. If the droplet is surrounded by a spherical reaction sheet, we can expect that  $Y_{Os}$  is very small, even if the reaction sheet is not very thin the oxidiser can be fully consumed when it diffuses to the surface of the droplet. In order to close the model, it seems very realistic to complete the two equations with an approximate formula for  $Y_{Os}$  in function of the gas temperature far from the droplet, or some mean reaction rate in the gas phase around. This can allow to choose one of the two limiting cases described above, and also some intermediate situation where a reaction sheet is close to the droplet but not completely around it.

It has to be remarked that, even when the chemistry can be assumed very fast, a classical diffusion flamelet not necessarily surrounds each droplet. More precisely, the flamelet that can be established around the droplet is not necessarily strong enough for giving a sheet of maximum temperature corresponding to combustion at equivalence ratio one. When the oxidant content around the droplet is small, because of the presence of almost unburned fuel or of combustion products from other neighbouring flamelets, the flamelet cannot increase greatly the temperature because it can burn only a very limited amount of oxidant. For moderately fast chemical reactions, the flamelet cannot be really ignited around each droplet, or all around each droplet, when the oxidant content is too small. That is the reason why only droplets on the fuel lean side of the spray flame brush can display an usual flamelet around. And for the other droplets, the vaporization without combustion around or with combustion around will give very similar results.

Anyway, when the PDF of the conserved scalar can be known, it is possible to directly obtain the field of any gaseous component when one assumes that the chemistry is very fast. This has

been shown for single phase turbulent diffusion flames, it holds also in the gas phase of spray flames. Using a model of vaporization for droplets without combustion is possible in this case, that will simply give results with a less convenient approximation (except for the prediction of lift off distance).

## 6. Conclusions

The study and prediction of turbulent flames has progressed significantly over the last fifty years, and the use of Joint PDF's has played a big role for that. The study of spray turbulent flames has not progressed to the same extent, and remains a big challenge for the combustion and flames community. Progresses have been obtained mostly for the description of the dispersion of droplets and their vaporization, and the level of knowledge and ability for predictions is poorer for gas phase combustion around or in between the droplets. The fluctuations of temperature and species concentrations that do exist in gas phase have been simply modelled with a Monte Carlo simulation, but without true connection with a JPDF transport equation.

We have shown how to obtain the balance equation for the Joint PDF of the chemical variables in the gas phase only, with the influence of the vaporization of the liquid droplets or parcels, based on primitive equations as sound as the Navier-Stokes equations are for gas flames. This equation, as expected, shows «unclosed» terms, and we have proposed closure approximations, in particular for the terms related to the droplets vaporization. The small scale approximations adopted for that are exactly the same as those used for the classical Lagrangian-Eulerian approach, consequently with the same domain of validity, limited to very dilute sprays of spherical droplets. These proposals go one step further compared to the recent studies on this subject. The modeling of mass added in the gas phase due to vaporization, involved in classical Monte Carlo models, sets again questions that can be studied without other approximations with a JPDF equation. The influence of gas phase fluctuations on the mean vaporization of droplets for Lagrangian simulation of their life is not really taken into account in engineering numerical codes. If we want to handle situations with dense spray flames anchored in the atomization region, the JPDF equation obtained here is valid, and can be used provided that small scale laws for droplets are replaced by appropriate laws for some « liquid elements ». Of course, these small scale laws will have to be studied with well defined experiments, or numerical experiments with DNS where the liquid parcels are resolved.

We are very happy with this paper to recognize and thank the pionnierring works of Ted O'Brien, that have shown the route in this subject.

## Appendix - List of the main notations and symbols

$C_k^{ab}$  : Cauchy stress tensor in phase  $k$

$e_{tk}$  : total energy of phase  $k$ , per unit of mass of  $k$

$f_k$  : phase index for phase  $k$

$g^a$  : gravity acceleration

$m_k$  : mass flux at the interface entering into the gas phase, per unit area

$n_k^a$  : normal at the interface (positively oriented towards the outside of  $k$ )

$P_g(Y, T, x)$  : JPDF of concentration and temperature, within the gas only

$q_l$  : energy flux entering into the liquid phase through the interface, per unit area

$q_g$  : the same for the gas

$v_s^a$  : velocity of the interface (component along the direction  $a$ )

$v_{rjgs}$  : relative velocity of species  $j$  within the gas, with respect to the interface, on the interface

$v_{krs}^a$  : relative velocity of phase  $k$  with respect to the interface, defined only on the interface  
 $Y_{jgs}$ : mass fraction of species  $j$  within the gas, on the interface  
 $\sigma$  : interface area per unit volume

## Data Availability Statement

Data sharing not applicable – no new data generated.

## References

- <sup>1</sup> H. L. Toor, « Mass transfer in dilute turbulent and non-turbulent systems with rapid irreversible reactions and equal diffusivities », *A. I. Che. J.*, **8**, 70-78 (1962)
- <sup>2</sup> W.R. Hawthorne, D.S. Weddell, H.C. Hottel, « Mixing and combustion in turbulent gas jets », *Proc. 3rd Symposium on Combustion and Flame, and Explosion Phenomena*, 266-288 (1949)
- <sup>3</sup> C. Dopazo and E. E. O'Brien, « A probabilistic approach to the autoignition of reactive turbulent mixtures », *Acta astronautica* **1**, 1239-1266 (1974)
- <sup>4</sup> R. W. Bilger, « Turbulent jet diffusion flames », *Prog. Energy. Comb. Sc.* **1**, 87-109 (1976)
- <sup>5</sup> V. A. Frost, « Model of a Turbulent, Diffusion-Controlled Flame Jet », *Fluid. Mech. Sov. Res.*, **4**(2), 124-133 (1975)
- <sup>6</sup> V.A. Sabelnikov, « Model equation for velocity and concentration PDF in turbulent mixing and diffusion combustion of gases », *Fisika Goreniya y Vzriva*, **19**(2), 37-46 (1982)
- <sup>7</sup> R.L. Curl, « Dispersed phase mixing : I theory and effects in simple reactors », *A. I. Che. J.*, **9**, 175-181 (1963)
- <sup>8</sup> J. Villiermaux and J.C. Devillon, « Représentation de la coalescence et de la redispersion des domaines de ségrégation dans un fluide par un modèle d'interaction phénoménologique », in « *Proceedings of the 2<sup>nd</sup> International Symposium on Chemical Reaction Engineering* », Elsevier, 1-13 (1972)
- <sup>9</sup> R.C. Flagan and J.P. Appleton, « A stochastic model of turbulent mixing with chemical reaction : Nitric oxide formation in a Plug-Flow reactor », *Comb. Flame* **23**, 249-267 (1974)
- <sup>10</sup> E.E. O'Brien, « The probability density function (pdf) approach to reacting turbulent flows », in « *Turbulent Reacting Flows* », 185-218, Ed. P.A. Libby and F.A. Williams, Springer-Verlag (1980)
- <sup>11</sup> F. Gao and E.E. O'Brien, « A mapping closure for multispecies Fickian diffusion », *Phys. Fluids A* **3** (5), 956-959 (1991)
- <sup>12</sup> V. Eswaran and E.E. O'Brien, « Simulations of scalar mixing in grid turbulence using an eddy-damped closure model », *Phys. Fluids A* **1** (3), 537-548 (1989)
- <sup>13</sup> S.B. Pope, « PDF methods for turbulent reactive flows », *Prog. Energy Comb. Sci.*, **11**, 119-192 (1985)
- <sup>14</sup> U.A. Maas and S.B. Pope, « Simplifying chemical kinetics: Intrinsic low-dimensional manifolds in composition space », *Comb. Flame*, **88**, 239-264 (1992)
- <sup>15</sup> R. Borghi and M. Champion, « Modélisation et théorie des flammes », Ed. Technip, Paris (2000)
- <sup>16</sup> D. Stepowski, A. Cessou, P. Goix, « Flame stabilization and OH fluorescence mapping of the combustion structures in the near field of a spray jet », *Comb. Flame*, **99**, 516-522 (1994)
- <sup>17</sup> R. Borghi, « The links between turbulent combustion and spray », in « *Transport phenomena in combustion* », Ed. S. Chan, Taylor and Francis, Vol.1, 1-18 (1995)
- <sup>18</sup> P. Durand, M. Gorokhovski, R. Borghi, « An application of the probability density function model to Diesel engine combustion », *Comb. Sci. Tech.*, **144**, 47-78 (1999)
- <sup>19</sup> P. Jenny, D. Roekaerts, N. Beishuizen, « Modeling of turbulent dilute spray combustion », *Prog. Energy Comb. Sci.*, **38**, 846-887 (2012)

- <sup>20</sup> M. Zhu, K.N.C. Bray, O. Rumberg, B. Roog, «PDF transport equations for two phase reactive flows and sprays », *Comb. Flame*, **122** 327-338 (2000)
- <sup>21</sup> R. Borghi and F. Anselmet, « Turbulent multiphase flows with heat and mass transfer » Wiley, ISTE (2014)
- <sup>22</sup> D.A. Drew, «Mathematical modeling of two-phase flow», *Ann. Rev. Fluid Mech.*, **15**, 261-291 (1983)
- <sup>23</sup> I. Kataoka, «Local instant formulation of two-phase flow», *Int. J. Multiphase Flows*, **12**(5), 745-758 (1986)
- <sup>24</sup> R.I. Nigmatulin, « Dynamics of multiphase media », Vol.1-2, Hemisphere, N.Y. (1991)
- <sup>25</sup> S. Tanguy and A. Berlemont, « Application of a level set method for simulation of droplet collisions », *Int. J. Multiphase Flows*, **31** (9), 1015-1035 (2005)
- <sup>26</sup> T. Ménard, S. Tanguy, A. Berlemont, « Coupling Level set/VOF/Ghost fluid methods : Validation and application to 3D simulation of the primary break up of a liquid jet », *Int. J. Multiphase Flows*, **5**, 510-524 (2007)
- <sup>27</sup> D. Fuster, A. Bagué, T. Boeck, L. Le Moyne, A. Leboisetier, S. Popinet, P. Ray, R. Scardovelli, and S. Zaleski, « Simulation of primary atomization with an octree adaptive mesh refinement and VOF method », *Int. J. Multiphase Flows*, **35**, 550-565 (2009)
- <sup>28</sup> X. Yang and A. Turan, « Simulation of liquid jet atomization coupled with forced perturbation », *Phys. Fluids*, **29**, 022103 (2017)
- <sup>29</sup> G. Vaudor, T. Ménard, W. Aniszewski, M. Doring, A. Berlemont, « A consistent mass and momentum flux computation method for two-phase flows. Application to atomization process », *Computers and Fluids*, **152**, 204-216 (2017)
- <sup>30</sup> S. Tanguy, T. Ménard, A. Berlemont, « A Level set method for vaporizing two-phase flows », *J. Comp. Physics*, **221** (2), 837-853 (2007)
- <sup>31</sup> B. Duret, R. Canu, J. Reveillon, F.X. Demoulin, « A pressure based method for vaporizing compressible two-phase flows with interface capturing approach », *Int. J. Multiphase Flows*, **108**, 42-50 (2018)
- <sup>32</sup> T.S. Lundgren, « Distribution functions in the statistical theory of turbulence », *Phys. Fluids*, **10**, 969-975 (1967)
- <sup>33</sup> D.W. Meyer and P. Jenny, « Micromixing models for turbulent flows », *J. Comp. Physics*, **228**, 1275-1293 (2009)
- <sup>34</sup> H. Chen, S. Chen, R.H. Kraichnan, « Probability distribution of a stochastically advected scalar field », *Phys. Rev. Lett.*, **63**, 2657-2661 (1989)
- <sup>35</sup> S.B. Pope, « Mapping closures for turbulent mixing and reaction », *Theoret. Comput. Fluid Dynamics*, **2**, 255-270 (1991)
- <sup>36</sup> M. Stöllinger and S. Heinz, « PDF modeling and simulation of premixed turbulent combustion », *Monte Carlo Methods Appl.*, **14**, 343-377 (2008)
- <sup>37</sup> H.-W. Ge and E. Gutheil, « Simulation of a turbulent spray flame using coupled PDF gas phase and spray flamelet modeling », *Comb. Flame*, **153**, 173-185 (2008)
- <sup>38</sup> M. Boivin, O. Simonin, K.D. Squires, « Direct numerical simulation of turbulence modulation by particles in isotropic turbulence », *J. Fluid Mech.*, **375**, 235-263 (1998)
- <sup>39</sup> D.H. Richter, « Turbulence modification by inertial particles and its influence on the spectral energy budget in planar Couette flow », *Phys. Fluids*, **27**, 063304 (2015)
- <sup>40</sup> G. Wang and D.H. Richter, « Modulation of the turbulence regeneration cycle by inertial particles in planar Couette flow », *J. Fluid Mech.*, **861**, 901-929 (2019)
- <sup>41</sup> J. Reveillon and L. Vervisch, « Spray vaporization in nonpremixed turbulent combustion modeling: A single droplet model », *Comb. Flame*, **121**, 75-90 (2000)
- <sup>42</sup> J. Reveillon, K.N.C. Bray, L. Vervisch, « DNS study of spray vaporization and turbulent micro-mixing », In AIAA98-1028, 36th Aerospace Sciences Meeting and Exhibit, Reno NV (1998).

- <sup>43</sup> D.B Spalding, « The combustion of liquid fuels », Int. Symp. Comb., **4**, 847–864 (1953)
- <sup>44</sup> G.A.E. Godsave, « Studies of the combustion of drops in a fuel spray - The burning of single drops of fuel », Int. Symp. Comb., **4**, 818–830 (1953)
- <sup>45</sup> W.A. Sirignano, « Fluid dynamics and transport of droplets », 2<sup>nd</sup> Edition, Cambridge University Press (2010)
- <sup>46</sup> M. Bini and W.P. Jones, « Large Eddy Simulation of an evaporating acetone spray », Int. J. of Heat and Fluid Flow, **30**, 471–480 (2009)
- <sup>47</sup> C. Hollman and E. Gutheil, « Flamelet-modeling of turbulent spray diffusion flames based on a laminar spray flame library », Comb. Sci. Tech., **135**, 175-192 (1998)
- <sup>48</sup> M. Mortensen and R.W. Bilger, « Derivation of the conditional moment closure equations for spray combustion », Comb. Flame, **156**, 62-72 (2009)
- <sup>49</sup> G. Borghesi, E. Mastorakos, C.B. Devaud, R.W. Bilger, « Modeling evaporation effects in conditional moment closure for spray autoignition », Comb. Theor. Model., **15**, 725-752 (2011)
- <sup>50</sup> J.C.K. Tang, H. Wang, M. Bolla, A. Wehrfritz, E.R. Hawkes, « A DNS evaluation of mixing and evaporation models for TPDF modelling of nonpremixed spray flames », Proc. Comb. Inst ; **37**, 3363–3372 (2019)