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Numerical analyses of laminar flames propagating in droplet mists using detailed and tabulated chemistry

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Abstract

Numerical analyses of laminar flames propagating in mono-dispersed quiescent droplet mists are addressed. Attention has been given to the evolution of combustion reactions interacting with different mists and the assessment of chemistry simplifications. In a first part, investigations are exclusively performed in a detailed description of the chemistry. Focus is concentrated on simplifications typically assumed in simulations of turbulent spray combustion. Namely, evaporative cooling and differential diffusion. These phenomena are systematically studied together with the characteristics of the observed flammability limits. In a second part, simulations conducted with the flamelet generated manifold (FGM) method are compared with detailed chemistry results. Flame propagation speed and profiles of selected quantities are used. Results demonstrate to what extent common simplifications applied to turbulent spray combustion, as well as FGM tables constructed with single-phase flamelets, are suitable to represent spray flame characteristics.

Keywords: Spray combustion, Detailed chemistry, Tabulated chemistry, Flamelet Generated Manifold

1. Introduction

The representation of combustion reactions by means of tabulated chemistry (TC) is a feasible strategy to address turbulent and complex flows. Diverse works have been applied this methodology to obtain more understanding and to proof modeling tools in flames interacting with evaporating liquid droplets (e.g. [1–7]). However, despite its popular usage, the performance of tabulated chemistry approaches is still not extensively evaluated. The growing interest in the assessment of tabulated chemistry methods using detailed chemistry (DC) computations (e.g. [6,8–10]) corroborates this statement.

Computations of general two-phase reacting flows with detailed description of the chemistry have been receiving more attention along the last years [6,8–12]. Regarding the evaluation of tabulated chemistry strategies, Olguin and Gutheil [10] and Franzelli et al. [9] exclusively analyzed laminar flames, while Vié et al. [8] and Franzelli et al. [6] also considered turbulent flows. Olguin and Gutheil [10] and Franzelli et al. [9] analyzed counter-flow configurations. The first reformulated the flamelet equations written in the mixture fraction space to include spray effects. Franzelli et al. [9] performed a-priori tests of different adiabatic tabulated chemistry strategies to reproduce spray flame characteristics in the context of multi-regime gaseous combustion. In [8] not only laminar flows, but also effects of the turbulence are investigated. Vié et al. [8] present a set of direct numerical simulations (DNS) to perform phenomenological investigation of reactive two-phase flows also in counter-flow configurations. The ability of the traditional formulation of the mixture fraction¹ to represent all phenomena is also evaluated for different droplet sizes. Franzelli et al. [6] performed laminar simulations of counter-flow flames with similar configuration as in [9] and also monodispersed-monokinetic (see [6]) Large Eddy Simulations (LES) of a turbulent swirled flame comparing the influence of the treatments given for the chemistry. There, a reduced chemistry mechanism composed by 24 species is compared with the FPI (Flamelet Prolongation of Intrinsic low dimensional manifold) approach to represent the n-dodecane combustion. Heat losses are not included in the FPI chemistry. Their effects are accounted for in a post-processing stage of the mixture temperature following the TTC² formalism. Despite being a-posteriori analyses, the combination of the TTC formalism and the

¹The traditional formulation of the mixture fraction is referenced here as the one proposed in [13].

²Tabulated Thermochemistry for Compressible flows

complexities of the counter-flow setup makes it difficult to isolate the influence of the spray on the chemistry. The same remark can be done for the complexities imposed by turbulence and the necessary modeling to account for the unresolved turbulence-flame interaction in those LES cases.

A common characteristic of the analyses of tabulated chemistry methods summarized above is the consideration of counter-flow burners for the laminar cases. Although this kind of reactor offers the possibility to isolate the turbulence, the distinctive dynamic of the two phases is clearly evident. As shown in [14], and [10], different droplets trajectories are obtained depending on the boundary conditions. In some cases, droplets show oscillatory behavior around the stagnation plane, demonstrating remarkable unsteady characteristics. Additional complications are noticed concerning the flame topology. Vié et al. [8] concluded that the resulting distribution of the droplets within the two-phase flow is able to modify the flame topology regarding the appearance of two simultaneous flame modes. In brief, the existence of flame stretch and the mixing of counter-flowing reactant interfere with the interaction of droplet mists and combustion reactions.

An alternative that allows a more isolated scenario for the analysis of chemistry effects is the configuration of a freely propagating flame in a droplets mist. It mimics a flat flame that propagate in a homogeneously distributed quiescent droplet mist within an adiabatic reactor. Once the flame freely propagates, no strain is necessary to stabilize the reaction zone. As a result, the importance of the dynamics of the flow field is reduced, in which the movement of droplets is reserved to the momentum exchange created by the thermal expansion. When compared with counter-flow burners, the absence of a counter stream of oxidizer allows better description of reactions in the oxidation zone. Particularly, the investigation of freely propagating flames aligns with the crescent interest in the usage of tables based on premixed flames to characterize turbulent spray combustion (e.g. [2,4,6,7]). The predominance of the premixed flame mode [15] and its relevance for the flame stabilization is supposed to justify the successful application of it. However, despite of this crescent usage, the performance of premixed tables is not evaluated in such an isolated context up to now.

The work of Neophytou and Mastorakos [11] stands out in the analysis of freely propagating flames in droplet mists. There, it was demonstrated that a great part of the phenomena associated with laminar spray combustion (e.g. [16–18]) could be recovered with a detailed description of the chemistry and the simplification of the entire flow in a one-dimensional framework. Previously to this work, the underlying mechanisms related to the interaction between droplets and flame were often interpreted exclusively in terms of multi-dimensional features (see [19,20]). For example, the flame wrinkling caused by the dispersed presence of droplets had been used to justify the increase of the flame propagation speed encountered in spray combustion. Since in the one-dimensional framework the multi-dimensionality is omitted, the interpretation of droplets-flame interaction from the viewpoint of the mixture characteristics (or the concept of an effective equivalence ratio [21]) received more attention. It worths to notice that analyses of tabulated chemistry strategies were beyond the scope of that work.

The structure of freely propagating flames in mists can be divided in five zones [19]: primary evaporation, preheat, homogeneous reaction, droplet burning and secondary evaporation. All of them are schematically exhibited in Fig. A1 for mists composed by large Sauter-Mean Diameters (SMD). In the two initial zones defined by Silverman et al. [19] the liquid fuel evaporation prepares the mixture to be burnt early in the homogeneous reaction zone. Small droplets usually do not survive the crossing of these

zones. Large droplets, which move through the reaction zone, may feature the droplet burning if enough oxidizer remains. Under this condition, the fuel reacts as soon as it evaporates. Finally, when no more oxidizer is available, the remaining drops evaporate in the secondary evaporation zone. It is important to highlight that such a structure does not always occur since it is strongly depending on the initial characteristics of the mist. In the investigations of Neophytou and Mastorakos [11] the burning of single droplets was neglected. Such simplification is supposed to be valid when droplets crossing flames are not large enough to stabilize an enveloped flame.

[Figure 1 about here.]

In the present study, simulations using a detailed description of the chemistry are used to evaluate the performance of tabulated chemistry modeling in the context of the flamelet generated manifold (FGM) method based on premixed flames [22]. For that, flames propagating in droplet mists are considered due to the suitable isolation of the influences of the flow field. The different aspects and simplifications commonly applied to the simulation of turbulent flames are systematically investigated. In a first stage, discussions are exclusively treated in the detailed chemistry context similarly as in Neophytou and Mastorakos [11]. While these authors concentrate on the influence of the liquid phase in the flame structure and the propagation speed for n-heptane and n-decane droplet mists, the initial investigations performed here with DC focus on the definition of the flammability limits, the relevance of differential diffusion transport, and heat losses. These aspects combined with the usage of ethanol and different numerical methods in the present work also complements and gives a valuable indication about the generality of the conclusions obtained in [11]. In a second stage, detailed chemistry computations are compared with simulations performed with FGM tables. Effects of heat losses are included in the chemistry of both methods, namely DC and FGM. It is not from our knowledge that, a FGM table build with single-phase flamelets with such a comprehensive description of heat losses as done here has already been used to address the spray combustion. Experimental test cases that could be used to reinforce the quality of such models are not available in literature yet. Furthermore, the conducted analyses complement and emphasize previous investigation presented in [6,9,10]. The closest setup to freely propagating flames refers to the analysis performed by Hayashi et al. [21], which is improved along the years by Nomura et al. [16,17,23]. Herein, flames propagating in enclosed vessels at high pressure were investigated. The droplet's size distributions are quite narrow and approach mono-dispersed mists. Nevertheless, the operating conditions are not precisely specified and the resulting flame is stretched, since it is spheric. Due to this lack of validation data, we rather rely on results obtained with detailed chemistry computations following the methodology applied in [11].

2. Modeling Procedure

An Eulerian-Lagrangian approach is applied to numerically describe the two-phase flow. Accordingly, the gas phase is described as a continuous medium in an Eulerian specification, whereas the liquid phase is composed by dispersed droplets tracked in a Lagrangian framework. Although both phases are treated separately, a full inter-phase two-way coupling is considered. The carrier gas phase quantities are interpolated to the droplets positions, while the influences originated by the dispersed phase are introduced through source terms in the computational cells of the Eulerian phase.

In order to isolate the diverse aspects of the spray combustion, the simulations presented in this manuscript mimic unstrained laminar flames propagating in isotropic droplet mists. Similar to Neophytou and Mastorakos [11], the following simplifications and approximations are assumed: (a) the multi-dimensional aspects of the droplets dispersion does not modify the flame surface, (b) mists are diluted, thereby no droplets interaction are accounted for, (c) no micro-mixing model is included, so that all the mass evaporated from a drop fills instantaneously the host control volume³. Furthermore, parcels are tracked instead of real drops. In this way, a parcel may represent more or even less than one real drop. The role played by them reduces to a dispersed source of fuel which follows the physical models described in sections 2.1 and 2.2. By setting a high value of parcels, it is possible to guarantee that the distribution of droplets is uniform within the domain. In this sense, transitory effects (oscillations) caused by the discontinuous distribution of fuel can be avoided, attending the statistically steady and one-dimensional representation of isotropic mists.

Considering that parcels allow the statistically steady representation of the problem, the fact that real droplets are bigger than cells does not invalidate these one-dimensional simulations. As stated in Portela and Oliemans [24], the only consequences refer to that droplets experience a local flow field with scales smaller than the ones forcing the actual droplets in a general case. Since the flow is laminar and one-dimensional here, the influences of such small scales do not modify the displacement of droplets. Additional obstacles could emerge regarding the distribution of vapor, which could lead to wrong estimation of the mixture composition in the far-field. Nevertheless, this problem does not affect the chosen one-dimensional setup. As multi-dimensional aspects are not considered, the remaining discontinuities of fuel vapor along the flame are avoided by the statistically steady representation.

Two computational softwares are employed to address the investigations presented here. The computations with the detailed description of the chemistry are performed with CHEM1D [25]. During this work, this software has been extended with a Lagrangian module to include the effects of the liquid phase. Simulations conducted with tabulated chemistry employed the software FASTEST [26]. Both codes and performed computations are different in many aspects. While our computations with CHEM1D follow the steady-state formulation, FASTEST operates in the unsteady mode. The choice of two different solvers with different formulations is also motivated by verification purposes, which ensure the comparability among results.

2.1. Gas Phase

The description of the carrier phase follows a variable-density low Mach number formulation. In agreement with the strategy used in Somers [25], the set of equations used to compute steady freely propagating flat flames in CHEM1D is

$$\frac{\partial \dot{m}}{\partial s} = S_{\text{vapor}}^L, \quad (1)$$

$$\frac{\partial (\dot{m} Y_i)}{\partial s} - \frac{\partial}{\partial s} \left(\frac{\lambda}{Le_i c_p} \frac{\partial Y_i}{\partial s} \right) = \dot{\omega}_i + \delta_{ik} S_{\text{vapor}}^L, \quad \text{and} \quad (2)$$

³When a droplet crosses a cell boundary a splitting factor is used to interpolate the source of vapor between the two host control volumes.

$$\frac{\partial(\dot{m}h)}{\partial s} - \frac{\partial}{\partial s} \left(\frac{\lambda}{c_p} \frac{\partial h}{\partial s} \right) = \frac{\partial}{\partial s} \left(\frac{\lambda}{c_p} \sum_{i=1}^{N_s} \left(\frac{1}{Le_i} - 1 \right) h_i \frac{\partial Y_i}{\partial s} \right) + S_h^L. \quad (3)$$

Equation 1 ensures the mass conservation of the coupled system, where $\dot{m} = \rho u$ is the mixture mass flux, ρ the density, u the gas velocity, s the spatial coordinate, and S_{vapor}^L the source term of vapor. The conservation of chemical species is described by Eq. 2. Herein, Y_i is the mass fraction of the species $i \in [1, N_s - 1]$, λ the thermal conductivity, c_p the isobaric specific sensible heat, Le_i the Lewis number for species i , $\dot{\omega}_i$ the reaction rate for species i , and δ_{ik} the Kronecker delta. The subscript k in Eq. 2 refers to the vapor species. The last equation of this set is associated to the conservation of energy, which is expressed in terms of the absolute enthalpy h . The absolute enthalpy of each transported species is represented by h_i , while the coupling term between phases is denoted by S_h^L .

As the computations performed with FASTEST are conducted in an unsteady context using the FGM method, the following set of equations is employed

$$\frac{\partial \rho}{\partial t} + \frac{\partial \rho u}{\partial s} = S_{\text{vapor}}^L, \quad (4)$$

$$\frac{\partial \rho u}{\partial t} + \frac{\partial \rho u u}{\partial s} = -\frac{dp}{ds} + \frac{\partial}{\partial s} \left(\mu \frac{\partial u}{\partial s} \right) + S_u^L, \quad \text{and} \quad (5)$$

$$\frac{\partial \rho \psi}{\partial t} + \frac{\partial \rho \psi u}{\partial s} = \frac{\partial}{\partial s} \left(\Gamma_\psi \frac{\partial \psi}{\partial s} \right) + \dot{\omega}_\psi + S_\psi^L. \quad (6)$$

In the momentum transport equation (i.e. Eq. 5), p is the pressure, μ the dynamic viscosity, and S_u^L the coupling source term of momentum. The relevance of this last term is evaluated in Appendix A. Equation 6 represents the conservation of the control variables used to access our FGM tables by means of a general quantity ψ . They are the mixture fraction Z , a reaction progress variable Y_{pv} , and h . Γ_ψ equals to μ/Sc for Z and Y_{pv} , while μ/Pr for h . As the unitary Le hypothesis is assumed in the tabulated chemistry analysis, $Sc = Pr = 0.7$. $\dot{\omega}_\psi$ is only present in the transport of Y_{pv} and refers to a chemical source term. Since in the combination of variables used to construct Y_{pv} the liquid vapor is not present, S_ψ^L only exists for Z and h . Details about the coupling source terms are delivered in section 2.3. As focus is given to the investigations of diluted spray mists, no void fraction is necessary to be included in Eqs. 1-6 (see [11]).

2.2. Liquid Phase

As long as the *Point-particle method* (PPM), also denoted as *Particle-Source-In-Cell* (PSIC) method, [27,28] is adopted, the description of the dispersed phase reduces to the modeling of the phenomena acting on the droplet-particles. Considering our focus on diluted sprays, some simplifications can be assumed. They can be listed as: (1) no secondary break-up exists and droplets are spherical, non-deformable liquid structures; (2) effects of the inner flow to droplets are not considered; (3) no additional modeling is included to represent the burning of single droplets; (4) the surrounding gas is always

treated as a mixture of fuel-vapor and air at the same temperature and pressure as the reacting fluid; and (5) droplets are single-component composed by liquid ethanol.

In the one-dimensional Lagrangian framework, the tracking of a parcel is essentially described by two ordinary differential equations (ODE) (e.g. Sirignano [29])

$$\frac{dx_p}{dt} = u_p, \quad \text{and} \quad (7)$$

$$\frac{du_p}{dt} = \frac{3}{4} \frac{C_D}{d_p} \frac{\rho}{\rho_p} |u - u_p|(u - u_p) \quad (8)$$

for the parcel position and acceleration, respectively.

In the present context, the drag is the unique force acting on a droplet. This can be observed in Eq. 8 which features the balance of forces. In both Eq. (7) and (8), quantities labeled with the subscript p are associated with droplet parcels. Specifically, x_p corresponds to the droplet position, and d_p is the diameter. To compute the drag coefficient C_D the model proposed by Yuen and Chen [30] is adopted.

Heat and mass exchanges are computed following the formulation of [31]. Both phenomena are described by

$$\frac{dT_p}{dt} = \frac{f_2 Nu}{3Pr} \left(\frac{\theta_1}{\tau_p} \right) (T - T_p) + \left(\frac{L_V}{c_l} \right) \frac{\dot{m}_p}{m_p}, \quad (9)$$

$$\frac{dm_p}{dt} = -\frac{Sh}{3Sc} \left(\frac{m_p}{\tau_p} \right) H_M, \quad (10)$$

with T the temperature, Nu , the Nusselt number, f_2 a correction factor due to evaporation, Pr the Prandtl number, and L_V the heat of vaporization. θ_1 is the ratio of the gaseous and liquid specific sensible heat (c_p/c_l), $\tau_p = \rho_p d_p^2 / 18\mu$ expresses the particle relaxation time, and $\dot{m}_p = dm_p/dt$. In Eq. (10), Sh is the Sherwood number, Sc the gas Schmidt number⁴, and H_M represents the specific driving potential for mass transfer according to [31]. Observe that the heat transfer by radiation is not considered.

The structure of the heat and mass exchange equations proposed by Miller et al. [31] is quite general and allows for the computation of the evaporation process with many modeling approaches. During the development of this work, different approaches have been tested. As the infinite liquid conductivity version of the non-equilibrium evaporation model of Miller et al. [31] properly addresses the evaporation phenomena in a broader range of operating conditions than the other tested models, it is applied in our investigations. Accordingly, the correction factor due to evaporation (f_2) and the specific driving potential for mass transfer (H_M) are computed following the formulation defined in [31].

⁴The non-dimensional numbers Pr and Sc are explicitly computed at each time integration of the evaporation equations based on material properties of the mixture composition in the film surrounding each tracked droplet. They do not follow the unitary Le assumption.

2.3. Phase coupling

The interpretation of the tracking of parcels in the unsteady formulation is straightforward, however a special treatment is required for the computation of freely propagating flames in the steady-state regime in CHEM1D. Basically, the overall structure of the Lagrangian routines of CHEM1D does not strongly differ from a typical Lagrangian solver. The main modifications are associated with the appropriate coupling of the intrinsically unsteady solution of the Lagrangian ODEs with the steady-state treatment of the carrier phase.

Concerning the above mentioned coupling, the time integration of the Lagrangian ODEs must fulfill two requisites. The first corresponds to the definition of a suitable time step to represent the steady solution of the dispersed phase properly. The second refers to the proper definition of the amount of mass, heat, and momentum to be added or extracted from the carrier phase. With respect to the first point, one may realize that as soon as the overall solution converges the unsteadiness of the Lagrangian tracking disappears. According to this observation, the distinction between both regime formalisms disappears as long as the two requisites are properly addressed.

The integration's time step can be computed as follows

$$\Delta t_{\text{lag}} = C_{\text{lag}} \frac{\Delta x}{u_d}, \quad (11)$$

in which C_{lag} is a user defined Courant number, and Δx the host cell width. In this sense, the time step varies along the domain to keep a sufficient number of time integrations per cell. For the present analyses C_{lag} is equal to 0.1, which allows 10 time steps per grid cell. When the solutions converges, u_d and Δx do not vary anymore which result in constant values for Δt_{lag} in each cell.

To better address the second requisite, the structure of the coupling source terms is reformulated. For unsteady solvers, these terms are typically written as [27]

$$S_{\text{vapor},i} = \frac{\theta_i N_p}{V_i \Delta t} \Delta m_{p,i}, \quad \text{and} \quad (12)$$

$$S_{h,i} = \frac{\theta_i N_p}{V_i \Delta t} \left(m_{p,i}^{\text{old}} \int_{T_{\text{ref}}}^{T_{\text{old}}} c_l dT - m_{p,i}^{\text{new}} \int_{T_{\text{ref}}}^{T_{\text{new}}} c_l dT - \Delta m_{p,i} (h_f - h_{\text{vap}}) \right), \quad (13)$$

where Δt is the coupling time step. Specifically, this is the procedure adopted in the code FASTEST. Considering the time step definition described in Eq. 11, this same formulation could be used in CHEM1D. However, during the development of this work, it could be noticed that the iterative numerical solution procedure leads to oscillations in the definition of N_p . Such oscillations make the solver convergence difficult. In view of this, another strategy has been employed to overcome this issue. The ratio $N_p/\Delta t$ in Eqs. 12 and 13 is substituted by the rate of real drops per parcel $\dot{N}_p$

$$\dot{N}_p = \frac{\dot{m}_l^0}{m_p^0}, \quad (14)$$

where $\dot{m}_l$ is the liquid mass flux, and m_p is the mass of a real drop. The superscript ⁰ indicates injection values. This solution leads to more stable values for the coupling

source terms improving the numerical solver.

In the above equations, θ_i is the splitting factor for the cell i , $\Delta m_{p,i}$ the evaporated mass of the parcel p within i , the superscripts *old* and *new* indicate properties evaluated before and after the time integration, and h_{vap} is the enthalpy of vaporization.

Concerning the simplifications and methods adopted here, one may realize that the tracking of one single parcel is sufficient to represent the dispersed phase. In this sense, the Lagrangian computations are reduced to the tracking of a single parcel along the computational domain at each coupling step. To accomplish this task, the tracking is performed up to the disappearance of this parcel. More specifically, its complete evaporation or departure from the computational domain.

As the numerical structure available in the version of the code FASTEST employed here follows an unsteady formulation, further implementations have been done to turn its computations comparable with CHEM1D. In this code, the solution procedure is based on a steady specification and the flame is kept in a pre-defined position. It is pointed out in section 2.5 that, the distance from the droplets injection location to the flame front becomes a new variable in this numerical setup. Such feature is not directly reproducible in an unsteady flow without the addition of an external controller for the boundary conditions. In view of this, a PID (Proportional-Integral-Derivative) controller [32] is implemented in FASTEST to attain this requirement.

2.4. Table construction

The computations performed with tabulated chemistry have been conducted with non-adiabatic FGM tables. Following the strategy proposed by van Oijen and de Goey [33], the construction of this kind of table starts with the calculation of adiabatic flamelets for a desired span of mixture compositions with the fresh gases temperature of 300 K. As Le is assumed to be unity, the absolute enthalpy remains constant within these flames. Hence, in a first step, the different enthalpy levels are obtained by varying the reactants' temperature. The maximum value of the fresh gases temperature (T_{max}) determines the upper enthalpy value for a fixed mixture composition, while its minimum (T_{low}) defines the lowest temperature allowed in the computational domain.

In view of a more universal description of the heat losses, two further steps are necessary to complete the non-adiabatic manifold. The first one consists of obtaining lower enthalpy values via burner-stabilized flamelets, which mimic premixed flames burning attached to a porous medium with controlled temperature. By keeping the temperature of this medium constant and adjusting the inflow velocity, heat losses can be introduced in the domain resulting in different enthalpy levels for the flame. In this particular step, the temperature is set to the same value as for the unburned gas of the coldest adiabatic flamelet. When the lowest enthalpy levels with the burner stabilized flamelets is reached, as seen in Fig. A2 (a), the burnt gas is still far away from T_{low} . To cover the remaining range of physical states, step-wise extrapolations of the thermo-chemical data are done to the lowest enthalpy level. This is computed by assuming the equilibrium mixture composition at T_{low} . As observed in Fig. A2 (b), this last step does not influence the reaction rates. Finally, once a manifold is generated, a look-up table is constructed over the controlling variables Z , Y_{pv} , and h . Following the strategy of Ketelheun et al. [34], an additional table is created to facilitate the establishment of the boundary conditions in terms of temperature.

To address the combustion reactions in the FGM and detailed chemistry context as well, the chemical mechanism proposed by Marinov [35] is employed. It represents

the oxidation of ethanol in air by means of 57 species and 379 intermediate reactions. Two tables have been used in our computations. Both differ by the definition of T_{low} , while T_{max} is kept constant at 900K. In the table labeled as FGM^s, T_{low} is defined in order to cover all possible ranges of absolute enthalpy reached for an adiabatic spray flame considering a chosen thermodynamic state for each phase in the fresh mixture and unitary Le . In all of our computations the pressure is defined as 101,325 Pa, the inlet temperature of the mixture (pure air) T^0 is set with the same value given to the injection temperature of droplets T_d^0 , i.e. 300K. To define the whole enthalpy range covered by FGM^s, the temperature obtained from the adiabatic saturation process (see [36]) of liquid ethanol in pure air is computed considering the chosen thermodynamic state for each phase at the process inlet. Accordingly, it is obtained that $T = 271K$ for $Y_{fuel} = 0.032$. Since the flamelets are computed for $Z \in [0.050, 0.194]$ and interpolations are done⁵ between these limiting values up to respectively pure air and fuel at 300K, T_{low} is set to 250K. As a result, the interpolation zone can cover the adiabatic saturation condition, which consequently allows the table FGM^s to describe the entire range of mixture compositions existing in our two-phase flow cases. In the table labeled simply as FGM (denoted as the reference table throughout the text), T_{low} is defined as an insufficient value to cover the entire range of enthalpies reached in our spray combustion setups, i.e. 280K. This strategy has been adopted to analyze the influence of the proper representation of heat losses in a FGM table. In summary, mass and heat transfer between the two phases are not included in the flamelets used to generate the table. But the strategy used to construct our tables makes them able to represent heat and mass transfer processes between both phases.

[Figure 2 about here.]

In both manifolds the following combination is used to define the reaction progress variable

$$Y_{pv} = \frac{1}{M_{CO_2}}Y_{CO_2} + \frac{1}{2.5M_{H_2O}}Y_{H_2O} + \frac{1}{1.5M_{CO}}Y_{CO}, \quad (15)$$

where M_i is the molar mass of species i .

2.5. Boundary conditions

The solution strategy applied to the two-phase flow computations did not change the formalism available in CHEM1D. Herein, the flame is fixed at the coordinate 0.0 of the computational domain. Specifically, the position for which the gas temperature reaches 200 K above its inlet value is used as a reference to define the origin of the coordinate system in both computational codes. According to this procedure the distance of the injection position of droplets to the flame front (l_{inj} - see Fig. A1) becomes a new variable to characterize the simulation. It plays an essential role for the definition of the mixture arriving at the flame, as it indirectly determines the residence time of the droplets in the pre-vaporization zone [11]. This parameter is varied here along with the initial diameter of droplets and the overall mixture compositions to a broad range of values. Along this paper their respective values are pointed out. Invariably to all computed cases, the injection position is kept constant during the simulation.

Once that the simulations performed here represent quiescent atmospheres within

⁵Interpolations are done following the procedure suggested in [37].

a flame propagates, the velocity of the parcel is set as the same as for the unburned gases. However, as the flow velocity is controlled to keep the flame positioning, u_d may vary along the iterative process.

Despite the variation of u_d , the ratio $\dot{m}_l/\dot{m}$ must be kept constant along the simulation. Otherwise, the overall mixture composition undesirably changes. The liquid mass flux can be directly obtained with the overall mixture composition in the cases presented in this work. By adjusting $\dot{N}_p$ in CHEM1D and N_p in FASTEST for each phase coupling, the ratio $\dot{m}_l/\dot{m}$ is kept constant.

3. Spray combustion in the detailed chemistry context

As previously mentioned, the implementations of the Lagrangian module did not modify the main structure of the solver employed for the carrier phase in CHEM1D. According to the solution of freely propagating flames with this software, no momentum equation is used. As discussed in Poinot and Veynante [38], the momentum transport is redundant for the corresponding kind of reactor for single phase flows. Since the velocity of the flow field is indispensable to perform the tracking of dispersed droplets (see section 2.2), it is computed here as

$$u_k = \frac{\dot{m}_k}{\rho_k}, \quad (16)$$

where $\dot{m}_k$ is the mass flux in $[\text{kg m}^{-2} \text{ s}^{-1}]$ in the control volume k .

The absence of the momentum transport naturally omits its corresponding phase exchange effects. Despite being expected to affect the entire flow field, the analysis presented in Appendix A demonstrates that the two-way coupling of momentum is not relevant in the present context.

[Figure 3 about here.]

The present section addresses the underlying phenomena to droplet-flame interactions in three parts. Initially, an overall description of the influences of the presence of droplets into the reaction zone is addressed. Following it, the effects of heat losses and diffusive transport are singly investigated.

A common characteristic of the set of simulations presented in the present section corresponds to the treatment given to the diffusion's transport. To account for differential diffusion transport the *Mixture Averaged*⁶ approach is applied. As summarized in Fig. A3, the participating species in the combustion of ethanol present a broad variety of Le . Effects of differential diffusion are specifically discussed in section 3.3.

3.1. Influence of the liquid phase on the reaction progress

[Figure 4 about here.]

The influences of the presence of droplets in the reaction zone can be easily perceived by the plots of the flame propagation speed displayed in Fig. A4. Herein, the profile of the flame speed obtained for single phase adiabatic simulations with the unburned gas temperature $T_u = 300\text{K}$ is added as a reference. Each curve labeled with DPM

⁶Hirschfelder-Curtiss approximation (see [22])

(Discrete Phase Modeling) corresponds to the results obtained with mono-dispersed mists composed by droplets with different initial diameters. In all of these simulations, droplets are injected at 3.0 cm upstream the flame front in thermal equilibrium with the incoming mixture, i.e. at 300K. The computations are performed for a span of ϕ_{over} ⁷ ranging from ϕ_{low} to an upper limit ϕ_{up} . An explanation about these limits is addressed in the sequel.

The influence of the droplet size is quite clear in Fig. A4. Besides the extension of the flammability limits, as the diameter increases the flame speed reduces for a same ϕ_{over} . This behavior is already highlighted in Aggarwal [39] and over Neophytou and Mastorakos [11] as well. The dependence of the flame speed on d_d can be clearly interpreted with the help of the theoretical equation proposed by Ballal and Lefèbvre [18] and improved by Sacomano Filho et al. [2], which is written as

$$s_{l,s} = \left[\alpha \left(\xi \frac{(1-\gamma)\rho_l c_{p,f} d_d^2}{\lambda_f \log(1+B)} + \frac{\alpha^2}{s_l^2} \right)^{-1} \right]^{0.5}. \quad (17)$$

where α is the thermal diffusivity of the surrounding gas, ξ a proportionality parameter (see [2]), γ the proportion of fuel vapor by its total amount arriving at the flame (i.e. $\dot{m}_{vap}/\dot{m}_{fuel}$), and s_l the laminar flame speed for a gas flame at ϕ_{over} . Subscripts labeled with f refers to properties evaluated in the droplet film.

Equation 17 can also help on the understanding of the effects of l_{inj} . As the distance of the injection's point to the flame increases, the residence time becomes longer. Consequently, the amount of liquid facing the reaction zone reduces, while γ and $s_{l,s}$ increase. This behavior can be visualized in Fig. A5 for two initial droplet diameters: 40 μm and 75 μm .

[Figure 5 about here.]

Issues concerning the flammability limits

During the development of this work attention has been given to the flammability limits of simulated flames propagating in mists. As noticed in Fig. A4, the lower limit resemble the single-phase lower flammability limit which is close to experimental measurements. Since the numerical burner is adiabatic, its solutions generally cover a broader span of mixture compositions than experimental measurements which are not perfectly adiabatic. Hence, ϕ_{low} is lower in numerical cases than in experimental tests. The limiting condition identified by ϕ_{up} clearly differs from upper flammability limits of single-phase flames. While ϕ_{up} is close to the region where $s_{l,s}$ is close to its maximum, such a limit is found for slow flames in single-phase cases.

In the most pragmatic interpretation, both limits are associated with the most extreme overall mixture compositions for which the numerical solver reaches the convergence. However, the difficulty to obtain a converged solution does not presume a disconnection with the underlying physics. Contrary to the hypothesis supposed by [11], spray flames are not always expected to reach relatively low values for $s_{l,s}$ close to ϕ_{up} when d_d^0 is kept constant. Observing Fig. A5, one can notice a dependence of ϕ_{up} with l_{inj} . As this last parameter decreases, ϕ_{up} is found closer to the mixture composition where $s_{l,s}$ is maximum. This behavior reinforces the departing of spray from gas flames, which is more pronounced when l_{inj} is low. An explanation for the

⁷ ϕ_{over} is the overall equivalence ratio. It is achieved when all the liquid is completely pre-vaporized.

rich quenching mechanism is presented in the following.

The evaporation process allow a gradual release of fuel vapor throughout the flame which can be more or less intense according to the droplet diameter among other spray properties. Because of this gradual fuel release, the flame burns in a preferential mixture composition which is not far from the stoichiometry. Differential diffusion effects may improve the burning rates and flames are able to react even stronger than gas flames [21] even including the evaporative cooling⁸. In Fig. A4 this is apparent for the diameters 25 μm and 35 μm . But for mixtures slightly richer than the ϕ_{over} at which $s_{l,s}$ is maximum, the flame quenches due to the high temperature gradients at the rear part of the flame. This mechanism resembles the one explained by Williams [40] for gas flames exposed to heat losses. The increase of ϕ_{over} by keeping d_d constant can be seen as the augmentation of the number density of droplets in a mist. At the pre-vaporization zone, such an increase does not significantly modify the mixture composition since the evaporation rate is low. As long as the reaction starts, droplets face high temperature zones with low concentration of fuel. This condition allows virtually unlimited evaporation rates. As a result, strong heat sinks are introduced within the reaction zone which may grow to a limit indicated by flame quenching. Particularly, the intensity of the heat sinks increases as d_d decreases as shown in Fig. A6. For small diameters, the evaporation rates are even higher causing the flame quenching in relatively low values of ϕ_{over} . Nonetheless, it must be highlighted that such quenching mechanism is a particular property of this kind of numerical configuration. Real spray flames may present additional phenomena which may lead to other quenching features. The different aspects discussed above can be visualized in Fig. A6, where profiles of gas temperature and $\dot{S}_{vapor}$ are presented for spray flames with different diameters at their respective ϕ_{up} .

[Figure 6 about here.]

Thermo-chemical properties

The analysis of flame propagation speed is quite helpful to distinguish the combustion in single and two-phase flows. The same is valid to assess the sensitivity of the input variables d_d , l_{inj} , and ϕ_{over} . But to get a better understanding about the effects of the liquid phase onto the reaction progress, it is indispensable to investigate the evolution of thermo-chemical properties.

The subsequent analyses are presented for two overall mixture qualities $\phi_{over} = 1.00$ and $\phi_{over} = 2.75$. As indicated in Fig. A4, for a mixture with $\phi_{over} = 1.00$ the increase of the droplet diameter follows the theoretical estimation given by Eq. 17. However, when $\phi_{over} = 2.75$, flames propagating in mists reach higher velocities than single-phase combustion.

[Figure 7 about here.]

Figure A7 exhibits the progress of the mass fraction of fuel Y_{fuel} and the temperature for both overall mixture compositions through the coordinate s . The results of three mists characterized by different diameters (25 μm , 50 μm , and 75 μm for $\phi_{over} = 1.00$ and 35 μm , 50 μm , and 75 μm for $\phi_{over} = 2.75$) are used to represent the droplets-flame interactions. Considering the stoichiometric mixture, a gradual departure from the single-phase solution can be noticed for the temperature as the droplet size increases. The effects of the heat losses are not so intense in this scenario due to the low amount

⁸The single-phase cases used here as reference are adiabatic flames with reactants at 300 K.

of fuel. Hence, the gas temperatures exiting the reactor outlet do not significantly differ from the gas flame. The concentration of vapor arriving the flame front evidences the higher evaporation rates of small droplets when compared with larger ones. The peak of the Y_{fuel} for $d_d^0 = 25 \mu\text{m}$ at the pre-heating zone reinforces this statement. Such a behavior is not different for $\phi_{\text{over}} = 2.75$. But in this case, the evolution of both plotted quantities completely distinguishes from the other scenario. Weak connections are identified between single and two-phase cases. The monotonicity of the profiles noticed for gas flames disappears with the presence of droplets. After the consumption of the pre-vaporized mass of fuel a second peak of Y_{fuel} appears at the droplet burning zone⁹. Its distance from the reaction zone increases with the diameter. After reaching a maximum value, temperature profiles of spray flames decay to an equilibrium value and clearly differ from the reference solution. When compared with the stoichiometric scenario, the effects of heat losses are more pronounced for rich cases as the injected amount of mass increases. Also noteworthy is that despite the increased flame speed for lower diameters in this rich scenario, the penetration into the oxidation zone is more susceptible to large drops. Such feature is indirectly observed by the higher fuel concentrations for deeper regions of the flame as d_d increases.

[Figure 8 about here.]

Combining the temperature and fuel mass fraction profiles of Fig. A7 with the heat release rate (HRR) exhibited in Fig. A8, high reaction rate regions can be identified between the coordinates 0.0 mm and 2.0 mm for the mists with $\phi_{\text{over}} = 2.75$. The proximity of spray HRR profiles to the reference solution indicate the preferential burning point of rich mists. The main reaction zone looks like a stoichiometric gas flame (see T profile). Only an amount of fuel close to the stoichiometry is vaporized and burnt. The released energy heats the gas to the stoichiometric flame temperature. After the main reaction zone, the heat is transferred to the droplets, which then vaporize. Finally, the temperature approaches the equilibrium value for a rich mixture. The bi-modality of the mass fraction of vapor and the temperature reduction at the rear part the flame demonstrates that evaporation is occurring downstream the region where HRR is maximum. Bearing in mind the definition of ϕ_{over} ¹⁰, one can notice that this quantity reaches its maximum value after the entire mass of vapor is released by droplets. In the present case it occurs downstream the maximum value of HRR be reached. The resulting preferential burning point justifies the higher speeds of spray flames when compared with single-phase ones for a same ϕ_{over} . Also noteworthy is that, despite having a lower maximum value than the gas flame with $\phi_{\text{over}} = 1.00$ the HRR profile for $d_d^0 = 35 \mu\text{m}$ and $\phi_{\text{over}} = 2.75$ extends for a larger region along the coordinate s . Such a behavior demonstrates that the reaction zone is thickened with the presence of droplets. In addition, the low intensities of HRR at the deeper regions into the oxidation zone explains the presence of fuel or intermediate species within it.

The extension of the HRR profiles throughout the flame is also observed for the scenario with $\phi_{\text{over}} = 1.00$. Again, a smooth departure from the reference solution is observed here as d_d increases. However, it shall be kept in mind that the spray flames have lower absolute enthalpy than the reference gas flame which inhibits the heat release by exothermic reactions.

⁹Flame zones are indicated in Fig. A1

¹⁰Observe that the equivalence ratio is based on the fresh mixture. Since this quantity gives a clear indication of reach ($\phi > 1.0$), lean ($\phi < 1.0$), and stoichiometric ($\phi = 1.0$) mixtures, it is written here in function of the mixture fraction to allow its usage throughout the flame. It can be computed as $\phi = [Z/(1 - Z)] [(1 - Z_{st})/Z_{st}]$, where Z_{st} is the stoichiometric mixture fraction.

[Figure 9 about here.]

Typically, the intersection of the mass fractions of formaldehyde $Y_{\text{CH}_2\text{O}}$ and the hydroxyl radical Y_{OH} are used to indicate the reaction zone in gas flames [41]. By comparing the profiles plotted in Fig. A9 with the HRR depicted in Fig. A8, one may realize that this statement has also some correlation with spray flames. Particularly, formaldehyde is used to indicate the pre-heating zone, while the hydroxyl radical marks the oxidation zone. Considering the stoichiometric mixture quality, the behavior already observed for the previous quantities is also noticed here. Again, a gradual departure from the gas flame solution can be seen for $Y_{\text{CH}_2\text{O}}$. However, some changes are noticed in the production of the hydroxyl radical. In the ascending part of Y_{OH} curves, a gradual departure from the gas solution can be identified. Yet in their descending part, an overlap can be seen between spray profiles. Since OH formation is sensitive to temperature, it is not surprising that for spray flames its concentration is lower than the reference. Such an overall behavior gives an indication of the interference of the liquid phase with the reaction progress. A strong influence of the dispersed phase is visible for $\phi_{\text{over}} = 2.75$. As expected for gas flames, a peak of formaldehyde is noticed at the pre-heating zone. However, the descending part of such peaks does not reach the abscissa line and directly starts to increase to build a second local maximum. This bi-modality can be directly connected with the profiles of Y_{fuel} in Fig. A7 and demonstrates the decomposition of fuel as it is released by the liquid phase. Even more remarkable are the interference of droplets with the evolution of the hydroxyl radical. While the reference solution presents a smooth decay in its descending part, the decay of Y_{OH} is quite abrupt for spray flames. A possible explanation for that can be drawn from the heat removal introduced by the evaporative cooling combined with the enrichment of the mixture. Both increase as d_d^0 decreases

[Figure 10 about here.]

Up to this point, the modifications introduced by the liquid phase have been analyzed with intermediate species of the ethanol combustion. Certainly these indicate modifications in the reaction evolution but no significant changes in the composition of the exhaust products. This is not the case for the mass fractions of carbon dioxide and carbon monoxide presented in Fig. A10. At the pre-heating and reaction zones, a gradual departure from the reference solution is again observed for $\phi_{\text{over}} = 1.00$. However, as soon as the mixture becomes hotter, the oxidation of CO in CO_2 is clearly affected by the liquid phase. The decay of the gas temperature caused by the evaporative cooling leads to a lower degree of dissociation of CO_2 in CO in the burnt mixture. Another interesting aspect of the profiles of carbon monoxide is that the observed bi-modality may induce the production and consumption of intermediate species in a behavior not found for gaseous flames. This aspect reinforces the distinction of the two kind of combustion processes. As visualized for the previous quantities, the effects of the liquid phase are quite evident for $\phi_{\text{over}} = 2.75$. From their profiles, it is worth to note that the droplet size is able to cause modifications that permanence along the entire reactor. Such behavior is also observed for the profiles of the mass fractions of acetylene and hydrogen exhibited in Fig. A11.

[Figure 11 about here.]

The diffusion of H_2 and C_2H_2 through the flame front are used in [11] to clarify the higher propagation speeds obtained for mists when compared with single-phase cases. Both molecules behave as intermediate species for the stoichiometric cases and as

unburned products for the rich mixture. Indeed, the higher concentrations of H_2 early in the reaction zone may give support to the reasoning suggested by Neophytou and Mastorakos [11]. However, since the Lewis number found for acetylene is considerably high, another thermo-chemical mechanism than the diffusion transport is expected to induce its high concentrations at the early stages of the reaction zone. From analysis of single-phase flames, reactions close to stoichiometric conditions present higher peaks of acetylene. Perhaps, the combination of the gradual release of fuel with lower enthalpies favors its production at the early stages of the reaction zone.

3.2. Influence of heat losses

The inclusion of evaporative cooling effects is a common topic in the computations of dilute spray combustion. Often this phenomenon is omitted due to the complexities arising by its interaction with the chemistry or the extra computational demands. Since the present numerical setup offers a fruitful framework to evaluate the influences of the omission of heat losses in detail, such a simplification is investigated here.

As evaporative cooling one shall read here the heat exchange between phases associated with the latent heat. Following this assumption and the formulation used for CHEM1D, Eq. 13 reduces to

$$S_{h,i} = -\frac{\theta_i \dot{N}_p}{V_i} \Delta m_{p,i} h_f. \quad (18)$$

Hence, although no vaporization's heat transfer is considered, an injection of formation's enthalpy is done. As observed later, it modifies the absolute enthalpy along the flame and may reduce the gas temperature in the oxidation zone.

Following the structure of section 3.1, initially the discussions are conducted with analysis of flame propagation speed. After that, profiles of selected quantities are used to discuss the evolution of combustion reactions. In order to avoid redundancies, the investigations presented in this section are restricted to just two numerical setups. These are mists with $\phi_{over} = 1.00$ and $\phi_{over} = 2.75$, $l_{inj} = 3.0$ cm, and $d_d^0 = 75\mu m$.

Analysis of the flame speed

The influences of heat losses in the flame propagation speed is clearly visible in Fig. A12. In this picture, as well as for all graphics of this section, results of the computation with evaporative cooling are included as a reference.

As soon as the evaporative cooling is neglected, the resulting spray flame speed is significantly higher. From Fig. A12 it is clearly evident that the omission of the heat losses give origin to overestimated flame speeds. Additionally, the effects observed for mists with $d_d^0 = 25\mu m$ and $d_d^0 = 35\mu m$ in Fig. A4 are present for flames without evaporative cooling. Namely, spray flames propagating faster than the maximum single phase cases. Different from the cases with heat losses, the solutions obtained for single-phase flames are directly comparable with the spray flames. For a specific ϕ_{over} spray and single-phase flames have the same value for h .

[Figure 12 about here.]

Thermo-chemical properties

As the departure from single phase solutions are already discussed in section 3.1, the evolution of thermo-chemical properties of gas flames are not included here. The comparisons are restricted to the solutions obtained for computations with and without evaporative cooling effects of flames propagating in droplet mists.

[Figure 13 about here.]

In Fig. A13 the evolution of fuel mass fraction and gas temperature are presented along the coordinate s for both overall equivalence ratios. Considering the characteristic of the mixture arriving at the flame, the amount of vapor increases when the heat losses are not accounted for both scenarios. This is an expected behavior as the gas temperature is kept at 300 K along the pre-vaporization zone and therefore it accentuates the evaporation process. For the rich mixture composition, the bi-modality of the Y_{fuel} profile is maintained, but the second local maximum is significantly reduced. Seemly, the higher temperatures reached in the oxidation zone intensifies the decomposition of fuel in intermediate species.

[Figure 14 about here.]

Regarding the reasoning given to the appearance of the faster flames induced by the higher diffusion of hydrogen in [11], the profiles plotted in Fig. A14 seem to partially contradict this theory. Despite the higher velocities of flames without heat losses, the flattening of H_2 profiles that would be expected from an increased diffusion transport is not apparent in the pre-heat zone. The higher concentration of this species seems to rather contributes to higher flame speeds. Such a flattering is slightly noticed in the oxidation zone. The development of the profiles of acetylene seems to depend on the overall mixture quality. As higher concentrations are observed for the solution without heat losses and $\phi_{\text{over}} = 1.00$, its concentration remains below the reference case in the pre-heating and reaction zones for $\phi_{\text{over}} = 2.75$. Just at the advanced stages of the oxidation zone, $Y_{\text{C}_2\text{H}_2}$ overcomes the curve of the simulation with evaporative cooling.

3.3. Influence of the diffusion transport

Part of the discussions conducted above uses the influences of the diffusive transport of chemical species and heat to interpret the reaction progress. This kind of transport becomes quite simplified and easier to interpret when the assumption of unitary Lewis number is applied. Namely, when the diffusive fluxes of heat and species are of the same intensity. If this condition is not attained, differential diffusion effects shall arise. Considering these characteristics, the interest to quantify the influences of the differential diffusion for spray flames arose along the development of this work. These are addressed in this section.

Similar to the last two sections, the analyses initiate here with a brief discussion about flame propagation speed. Afterwards, profiles of the same quantities presented in the last section are used to evaluate the features introduced by the diffusive transports into the reaction evolution. Exclusively for the analysis of flame speed, results are presented for droplet mists composed by different diameters. The remaining of the discussion is performed with the same numerical setup employed for the discussion about the importance of the evaporative cooling. For all cases, droplets are injected at 3.0 cm upstream the flame front.

Analysis of the flame speed

To better situate the present discussion, the profile of the propagation speed of single phase flames computed with unitary Lewis number is included in Fig. A15. As clearly noticed, the impacts of the differential diffusion are already perceived for gas flames. Differences in relation to the computations performed with the mixture averaged approach appear for lean and rich compositions. Nevertheless, these are more evident near to the stoichiometric mixture.

The results presented in Fig. A15 for the two-phase flows stem from three mists, which are distinguished by their initial droplet diameter (i.e. 25 μm , 50 μm , and 75 μm). In contrast to gas flows, the importance of the diffusive transport monotonically increases with ϕ_{over} . With respect to the droplet size, profiles approach each other as d_d^0 increases.

The main highlight of the analysis of the flame speed is that, the relevance of the differential diffusion transport seems to be milder for spray than for gas combustion. Particularly, for lean and slightly rich mixtures, no strong distinction is noticed for flame propagating in mists. The gradual distribution of fuel along the flame seems to alleviate the features introduced by the molecular transport as a whole. This overall outcome is quite relevant for the simplification of the chemistry in simulations of multi-dimensional and turbulent flows. Usually, the unitary Le assumption is adopted in such cases.

[Figure 15 about here.]

Thermo-chemical properties

The comparisons between the thermo-chemical properties of simulations with and without differential diffusion follow the same structure applied in section 3.2. Thus, the droplet mists are characterized by two overall mixture qualities ($\phi_{over} = 1.00$ and $\phi_{over} = 2.75$), $l_{inj} = 3.0$ cm, and $d_d^0 = 75\mu\text{m}$.

[Figure 16 about here.]

Influences of the differential diffusion effects can be already perceived in the pre-vaporization zones of both scenarios exhibited in Fig. A16. The omission of such mechanism induces higher concentrations of vapor arriving at the flame front. This is an indirect outcome of the assumption of $Le = 1$, since it is caused by the lower flame speeds. As the flame moves slower, the residence time of droplets in the pre-vaporization zone increases and consequently the amount of released vapor is more pronounced. Direct influences of the diffusion transport can be rather observed in the early stage of the reaction zone for $\phi_{over} = 1.00$. Despite the small changes in the ascending part of the temperature profile, the low diffusiveness of ethanol ($Le = 1.66$ - see Fig. A3) gives origin to a stronger gradient than for the cases with unitary Le assumption. Similar behavior is also noticed for $\phi_{over} = 2.75$. In this scenario, the bi-modality of the Y_{fuel} curve is also maintained for $Le = 1$ but not so extended as for the reference solution. Impacts on the profile of the temperature are more expressive in the oxidation zone. The marginally higher temperatures observed for the stoichiometric mixture can be justified by the slightly intenser HRR depicted in Fig. A17. The opposite functioning of this mechanism can be employed to explain the lower temperatures observed at the descending part of the temperature contour for $\phi_{over} = 2.75$. Downstream of it, higher HRR are visible for $\phi_{over} = 1.00$ and both temperature profiles approach each other asymptotically.

[Figure 17 about here.]

[Figure 18 about here.]

As the diffusivity of acetylene slightly increases with the unitary Le assumption, it is expected that its profiles would be flatter when compared with the mixture averaged approach. This is not observed in Fig. A18. However, in this discussion it is valid to account for the fact that the flame speeds obtained with the $Le = 1$ approach are lower. Certainly this aspect contributes to the steeper contours of the intermediate species. Perhaps, it explains the higher concentrations observed at the pre-heating zone for $\phi_{over} = 1.00$, since at this condition the flame speed does not strongly depend on the diffusion transport. Considering the mass fractions of hydrogen, the decrease in its overall diffusion is visible for both scenarios in Fig. A18.

4. Evaluation of the FGM with detailed chemistry computations

The simulations performed with the detailed description of the chemistry left quite clear that droplets interfere with the evolution of the combustion reactions. Regarding manifolds constructed with single-phase flamelets, such interferences are not explicitly accounted for. Specifically, single-phase flamelets are used to obtain the databases described in section 2.4. As the main objective of this paper, it is desired to know to what extent the constructed manifolds can represent spray flame characteristics.

As discussed in section 2, the investigations with tabulated chemistry are performed with FASTEST. Its unsteady formulation leads to increased computational efforts, since convergence is requested at each time step. Therefore, solutions with FASTEST require more computational time when compared with CHEM1D. In order to alleviate the computational demands, 36 one-dimensional flames are computed with FASTEST. They correspond to three values for d_d^0 , for four overall mixture qualities ($\phi_{over} = 1.00, 1.40, 1.80$, and 2.20) with and without the effects of evaporative cooling. While the table constructed for fresh reactants varying from 280 K to 900 K is applied to all cases, the other table is only employed in non-adiabatic simulations. Considering that both tables are constructed following the unitary Le assumption and fixed values for Pr and Sc , the solutions with detailed chemistry presented in this section also follow these simplifications. Accordingly, the transport equations of species mass fraction and absolute enthalpy solved in CHEM1D (see Eqs. 2 and 3) are simplified to

$$\frac{\partial(\dot{m}Y_i)}{\partial s} - \frac{\partial}{\partial s} \left(\frac{\mu}{Sc} \frac{\partial Y_i}{\partial s} \right) = \dot{\omega}_i + \delta_{ik} S_{\text{vapor}}^L, \quad \text{and} \quad (19)$$

$$\frac{\partial(\dot{m}h)}{\partial s} - \frac{\partial}{\partial s} \left(\frac{\mu}{Pr} \frac{\partial h}{\partial s} \right) = S_h^L. \quad (20)$$

The evaluation of the FGM databases with detailed chemistry follows the same structure adopted for the previous sections. In this sense, the discussion initiates with comparisons of flame speeds and it is followed by analyses of selected quantities.

Analysis of the flame speed

The flame propagation speeds resulting from the 36 cases are shown in Fig. A19 together with the reference solutions of CHEM1D. The computations with detailed chemistry are performed for a diameter range varying from $75\text{ }\mu\text{m}$ to the maximum value between $25\text{ }\mu\text{m}$ and the smallest diameter for which the mist could be made to burn.

[Figure 19 about here.]

An overall pretty good agreement of FGM flame speeds with DC computations can be observed in Fig. A19. Deviations noticed for simulations without evaporative cooling effects are small. For all cases, which have a corresponding DC computation, deviations of FGM computations are below 3.4%. Flames burning in mists with lower values of d_d^0 can be obtained for FGM computations when compared with DC results due to the interpolation methods employed in our FGM tables. These are necessary to interpolate flamelets data available at lower and upper flammability limits to the respective mixture fractions of 0 and 1 (see section 2.4). As a consequence, the mixture burning range is artificially extended in our tables. In Fig. A19, it can be noticed that flames computed with FGM for $\phi_{over} = 1.40$ can be made to burn up to $d_d^0 = 25\mu\text{m}$, while DC simulations present the minimum allowed value for d_d^0 of $29\mu\text{m}$. In this scenario, these artificial FGM results seem to behave as a prolongation of the profiles obtained with DC computations. However, as d_d^0 become considerably smaller than the minimum value obtained for DC cases, such a prolongation is no longer clear. This discussion is not extended here since it is beyond the scope of the present study.

Similar to computations performed without effects of the evaporative cooling, cases computed with the table FGM^s agree pretty well with their corresponding reference DC computations (“DC EC” in Fig. A19). Small underestimation of the laminar flame speed can be observed for mists composed by initial droplet diameter of $50\text{ }\mu\text{m}$. But these deviations are not higher than 4.2%. Differences on the values of $s_{l,s}$ computed with the reference FGM table (“FGM EC” in Fig. A19) from detailed chemistry simulations are also not high. Deviations are below 4.9% for the different initial droplet diameters. However, discrepancies are more evident when both tables are compared. The extension of the enthalpy range allowed with the table FGM^s yields lower values for the laminar flame speed when compared with our reference FGM table. These become more apparent as d_d^0 decreases. Such an outcome can be explained by the high enthalpy levels delivered by the reference table, which consequently increases the flame propagation speed. The overestimation of mists enthalpy levels are addressed in more detail in the discussions of thermo-chemical properties, where differences between tables are further investigated.

With respect to DC computations, one can notice that simulations considering evaporative cooling effects show a broader burnable range for decreasing d_d^0 values than cases without these effects. Such behavior is quite clear for mixtures showing $\phi_{over} > 1.00$. Despite sounding contradictory, an explanation for that could be drawn during the development of the present work. When evaporative cooling is considered, the mixture composition arriving the flame finds the physical limitation imposed by the saturation. Herein, the phase equilibrium is reached for droplet diameters below a certain value keeping l_{inj} constant. Such a behavior does not happen when EC is not accounted for, since the gas temperature is not reduced by heat transfer with the liquid phase. Under this condition, the mixture arriving the flame front becomes always richer when d_d^0 decreases or l_{inj} increases. As a result, cases without evaporative cool-

ing show richer mixture compositions and smaller droplet diameters arriving at the flame for a specific value of d_d^0 when compared with simulations considering EC. Since diameters are smaller, the evaporation is more pronounced in cases without EC when the combustion starts. Therefore, flames are more susceptible to the quenching caused by the heat removal in the rear part of the reaction zone. This mechanism is comprehensive discussed in section 3.1. Regarding general spray combustion simulations one can expect that, the omission of heat losses can not only lead to over-predicted mixture fractions and temperature but also deliver unrealistic flame quenching. From Fig. A19 one can also conclude that, the trends previously observed in section 3.2 are also present in FGM simulations. Namely, flames are faster when the evaporative cooling is neglected for a specific value of ϕ_{over} .

It is valid to highlight that, the agreement of the FGM spray flame speed with DC is a quite valuable outcome for the estimation of the flame topology in multidimensional simulations. Particularly, the propagation speed is a decisive characteristic to proper address such a feature [2]. As simplifications of the chemistry are typical in the simulation of turbulent flames, the application of FGM tables based on premixed flamelets is expected to suitably address the flame topology of lean or slightly rich burning spray flames (e.g. flames investigated in [2,42]).

Thermo-chemical and dispersed phase properties

The analysis of thermo-chemical and dispersed phase properties along the flame is an important procedure to get more understanding about the simplifications included with the FGM databases. The layout of this sub-section is divided in two parts. Initially, we focus on the stoichiometric overall mixture. Subsequently, results obtained for rich values of ϕ_{over} are presented.

Stoichiometric mixture ($\phi_{over} = 1.00$)

Temperature profiles of the gas phase are depicted in Fig. A20 for the three mists investigated, i.e. $d_d^0 = 25 \mu\text{m}$, $d_d^0 = 50 \mu\text{m}$, and $d_d^0 = 75 \mu\text{m}$. This quantity has been chosen here, since droplet evaporation strongly depends on gas temperature (see evaporation modeling description on section 2.2). Hence, when the computation of this temperature fails, deviations on the estimation of the liquid phase are expected. For all droplets sizes similar behavior can be observed. No significant differences are noticed in the pre-vaporization zone, as well as for the temperatures exiting the reactors between both methods. However, deviations are perceived at the late stage of the reaction zone in all cases. In this region, FGM underestimates the values delivered by DC computations. This characteristic is extended along the coordinate s as d_d^0 increases, since larger drops feature a more distributed release of fuel vapor along the reaction which consequently spreads the deformation in the temperature evolution. From these plots, one can state that the temperature evolution of the carrier phase computed with our assembly of single phase flamelets differs from the detailed chemistry computations. Since in mists composed by large values of d_d^0 the droplet penetration is higher, influences of the underestimation of the gas temperature become higher as the droplet diameter increases. The underestimation of gas temperature by tables based on single-phase reactions could also be noticed in [6]. When compared to the present simulations, those deviations are not well pronounced and reserved to a small region due to the strain imposed by the counter-flow setup. Hence, the presented results confirm the temperature deviations observed in [6]. At the final part of this section, additional

discussions about the deviations observed for the gas temperature are addressed.

[Figure 20 about here.]

[Figure 21 about here.]

The intensity of the combustion reactions can be expressed via the evolution of the source term of the progress variable, which is depicted in Fig. A21. This quantity is strongly connected with the flame propagation speed and is useful to draw an explanation for the results illustrated in Fig. A19. Focusing on the case with $d_d^0 = 50 \mu\text{m}$, one can realize that the profile obtained with FGM is not exactly the same as for the reference solution, but no strong distinctions are observed. In the case without heat losses, the shape and the maximum value obtained with the FGM computation are quite similar to those achieved with DC. Nevertheless, the profile obtained with detailed chemistry is slightly upstream the FGM profile. With respect to the case with evaporative cooling, the same happens when the table FGM^s is compared with DC results. On the other hand, higher values of $\dot{\omega}_{pv}$ can be noticed for the simulation performed with the reference table. Considering that the flame speed is influenced by the integral of $\dot{\omega}_{pv}$ along the flame, such higher values of $\dot{\omega}_{pv}$ by the “FGM EC” can explain the faster flames delivered with this table as shown in Fig. A19.

Regarding the properties of the dispersed phase, the evolution of the source terms of vapor and d_d are also plotted along the coordinate s in Fig. A21. FGM solutions agree reasonably well with DC computations. At the second half of the ascending part of vapor source term profiles, DC computations shown slightly higher values than FGM simulations. This is a direct outcome of the higher temperatures presented in Fig. A20. The maximum value of this quantity is well predicted by FGM simulations without EC, while the inclusion of heat losses in the table marginally underestimates this value. The evolution of droplets profiles is well characterized with the FGM computations. Differences appear when the reference table is applied to cases with heat losses. As previously discussed, this table cannot accurately describe the gas temperature in the pre-vaporization zone. Consequently, higher values of this temperature are considered. This leads to higher evaporation rates and, therefore, smaller droplets arriving to the reaction zone. These results motivate the analysis to the pre-vaporization zone of our flames, which is addressed in the following.

In Fig. A22 the evolution of gas temperature, absolute enthalpy, and mixture fraction is plotted against the coordinate s . In all cases investigated here, FGM simulations neglecting the evaporative cooling match solutions with detailed chemistry for different droplet diameters. From these plots, the influence of the extension of the enthalpy limits in a FGM table becomes evident. Results obtained with the table FGM^s agree quite well with DC computations and much better than those obtained with the reference table. The leak of information in low temperature levels of the reference table leads to higher temperatures, lower enthalpies, and higher mixture fractions arriving to the flame, as noticed for the case labeled as “FGM EC”.

[Figure 22 about here.]

[Figure 23 about here.]

Quantities depicted in Figs. A20 and A21 are mostly associated to the flame structure and do not precisely give details about intermediate reactions. In order to get more understanding about intermediate stages of the reaction evolution, the profiles of the mass fractions of CO_2 and OH along the coordinate s , as well as the relation of

both quantities are plotted in Fig. A23. The evolution of Y_{CO_2} resembles those seen for gas temperature, where deviations occur in reaction and oxidation zones. It is quite evident that FGM solutions underestimate the concentration of carbon dioxide. On the opposite side, mass fraction of OH are over-predicted by FGM tables. Deviations can reach the value of 20% from DC computations in cases including the evaporative cooling. Clearly, since the thermodynamic equilibrium is introduced in our FGM tables, as far the mixture flow within adiabatic reactors lower differences are noticed between FGM and DC computations for both quantities. To concentrate the analysis on the chemistry, the plot of Y_{OH} in function of Y_{CO_2} clearly exhibits the departure of FGM from DC solutions concerning intermediate species. For both scenarios (with and without heat losses), less OH is produced in DC computations.

The deviations observed in Figs. A20 - A23 indicate that our FGM tables cannot exactly describe spray combustion reactions. Namely, the chemistry evolution of flames propagating in droplet mists cannot be strictly decomposed in an ensemble of reactions derived from single-phase freely propagating flamelets. Deviations are noticed in the flame structure and on the evolution of major and minor intermediate species. This aspect is reinforced in the plots presented in Fig. A24, which exhibit the evolution of Y_{pv} , h , and gas temperature as functions of mixture fraction. Considering the profiles of Y_{pv} , the flow field can be divided in four parts: (1) pre-vaporization zone where Z increases with Y_{pv} zero, (2) vigorous increase of Y_{pv} up to an inflection point, (3) smooth increase of Y_{pv} up to Z reaches its maximum, and (4) increase of Y_{pv} with Z constant. According to this classification, solutions obtained with FGM without EC match DC results except for part (3) which corresponds to the zones with deviations in Figs. A20 and A21. Such discrepancies can be explained by the different natures of the reaction evolution in both methods. In detailed chemistry computations, the fuel released by droplets is distributed along the computational domain. A portion of this fuel mixes with fresh reactants before arriving at the flame, while another amount is injected in an evolving reactive process when droplets are large enough. In this second situation, fuel is decomposed in high temperature mixtures composed by diverse chemical species. Since reactions evolve according to the overall mixture composition and state, the presence of fuel is perceived in this context. In case of the FGM constructed with single-phase flamelets, the fuel released by droplets causes the increase of the mixture fraction in the gaseous mixture. When Y_{pv} is zero, the increase of Z corresponds to the increase of fuel mass fraction. This effect is correctly represented with the present FGM tables. However, when fuel is injected during the reaction progress, the increase of Z does not necessarily correspond to a strict increase of the fuel mass fraction. Part of the released fuel vapor is interpreted by the table as not completely burnt products, which depict different reaction evolution as the pure introduction of fuel in similar mixture composition and state. Deviations in part (3) of cases including evaporative cooling effects are also associated the deviations observed in Figs. A20 and A21. By comparing results obtained with both tables, it is evident that the deviations encountered in the pre-vaporization zone by the reference table (part (2)) do not significantly affect the evolution of the chemical reactions on part (3). Such an outcome is reinforced by the close agreement between both tables in Fig. A23. Similar to cases without heat losses, computations performed with the table FGMs agree with DC except in part (3).

The discrepancies observed for the enthalpy in Fig. A22 disappear in the plots of Fig. A24. The similarity between profiles computed with DC and FGM in this last figure indicates that the modifications in the chemistry are rather reserved to the evolution of intermediate species instead of absolute enthalpy. Such a reasoning is confirmed

with the plots presented for gas temperature in Fig. A24. Herein, the discrepancies in zone (3) evidence the difference of the sensible enthalpy of the mixture. Since absolute enthalpy matches while its sensible component varies, modifications are indirectly realized in values of formation's enthalpy of the mixture. This last quantity is strongly connected with the mixture composition.

[Figure 24 about here.]

Rich mixtures

With respect to the rich overall mixture compositions, the evolution of the thermochemical and dispersed phase properties intensifies the aspects visualized for the stoichiometric case. Figure A25 depicts the profiles of the temperature of the carrier phase for three values of ϕ_{over} . The under-predictions in the reaction zones already noticed in Fig. A20 are observed for all cases. Indeed, the main influence of the presence of the liquid on the flame emerges along the reaction progress. Such influence is more pronounced for rich mixtures due to the higher amount of evaporated fuel throughout the flame. Also noteworthy are the differences among the curve shapes describing the gas temperature. While mists with $\phi_{over} = 1.80$ and 2.20 properly delineate the profiles observed in the reference solutions, differences are noticed in the temperature profiles of leaner mixtures. Again, the FGM simulations with the reference table agree quite well with DC solutions in the burnt gas zones.

[Figure 25 about here.]

[Figure 26 about here.]

Figures A26 and A27 shows similar evolution of corresponding profiles for $\phi_{over} = 1.00$. From the four zones identified for the stoichiometric mixture, the first three are observed here. Again, zone (3) coincides with regions presenting discrepancies for gas temperature (see Fig. A25). As previously mentioned, such discrepancies are more pronounced in rich mixtures. Simulations performed with table FGM^s approach DC solutions better than the reference table for cases with evaporative cooling effects, but are not able to reproduce droplet-flame interactions in the final stages of the combustion reaction. As previously noticed for stoichiometric cases, deviations observed for the reference FGM table in the pre-vaporization zone (part (2)) do not affect the predictions in part (3). Computations performed with both tables agree pretty well with each other in part (3). Altogether, these results reinforce the reasoning already given for stoichiometric mists, in which our manifolds (constructed only with single-phase flamelets) cannot recover the chemistry evolution of flames propagating in droplet mists.

[Figure 27 about here.]

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6. Summary

Investigations of laminar flames propagating in quiescent droplet mists are conducted in a numerical context. Focus is given to the description of the chemistry and its simplifications typically applied to address turbulent spray combustion. The obtained results demonstrate to what extent these simplifications can be suitably employed.

In a first part, analyses are strictly conducted in a detailed chemistry (DC) context. It could be clearly demonstrated that the presence of droplets creates modifications on the flame characteristics and the reaction evolution. Extension of flammability limits and faster flames when compared with single-phase cases could also be observed here. These general modifications are sensitive to the droplets size, injection position, and the overall mixture composition. The relevance of the evaporative cooling and the differential diffusion transport are separately evaluated. The absence of heat transfer due to phase exchange strongly modifies the resulting flame structure and composition of the mixture exiting the reactor. Modifications of the flame structure are also noticed with the omission of diffusive transports with the unitary Le assumption. Nevertheless, less impact was perceived for spray than for pure gas flames. In both cases (heat losses and differential diffusion), impacts become significantly small for lean and slightly rich mixtures. Such an outcome indicates that for these mixture compositions, typical simplifications applied to address turbulent spray combustion are suitable.

In a second part, the simplification of the chemistry by FGM tables based on single-phase flamelets is assessed. Results demonstrate that the constructed manifolds offer a good approach to spray flame properties. In fact, the propagation flame speed agrees pretty good with detailed chemistry computations. Particularly, this indicates that such FGM tables can be a suitable tool for simulations aiming to predict the topology of flames burning lean or close to stoichiometry. In these cases, tools based for instance on the Artificially Thickened Flame (ATF) and Filtered Tabulated Chemistry for LES (F-TACLES) are suitable candidates to address such a problem. Analogously, results also reveal that the evolution of thermo-chemical properties estimated with our manifolds is not far from results determined with DC. Nevertheless, for situations in which the chemical equilibrium is not expected, the application of FGM tables constructed with single-phase freely-propagating flames shall deliver erroneous results. The release of pure fuel vapor along a progressing reaction clearly demonstrates this. This kind of problem is supposed to be managed when effects of heterogeneity are explicitly included in a FGM manifold, for instance by constructing manifolds with spray flamelets. Such an approach is in agreement with the work conducted by Olghin and Gutheil [10], who proposed the tabulation of counter-flow spray flamelets. Another possibility to address this problem is by means of combinations of different gaseous flamelet archetypes as proposed by Franzelli et al. [9]. However, it remains to be investigated whether these both methods will work for the current configuration, i.e. freely propagating spray flames.

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Appendix A.

In flames propagating freely in quiescent mists, relative movement between phases exists. As already mentioned in section 1, it is exclusively induced by the thermal expansion of the mixture. For this reason, the momentum exchanges occur in the proximities of the reaction zone. The importance of these exchanges is evaluated here by means of one-dimensional simulations conducted with the code FASTEST, where the momentum equations are always part of the solution procedure. By enabling and disabling the two-way coupling for these equations, the influences of the phase interaction can be assessed by comparisons.

[Figure 28 about here.]

Three diameters are chosen to evaluate the effects of the momentum exchanges: $25\text{ }\mu\text{m}$, $50\text{ }\mu\text{m}$, and $75\text{ }\mu\text{m}$. The overall mixture quality of $\phi_{over} = 1.40$ is chosen due to the broad variety of flame speeds obtained by the selected droplet sizes (see Fig. A19).

The comparisons of the evolution of gas and droplets velocities presented in Fig. A28 clearly demonstrate that the two-way coupling of momentum is not relevant in the present context. No significant changes are perceived either for gas or droplets velocities. The profiles of both quantities are also insensitive to variation of the droplet's diameters, which are able to modify the flame speed. Despite not included here, no impact is noticed in the propagation speeds of all tested flames.

Figure A1. Schematic of the structure of freely propagating laminar flames in mists. Adapted from [11] and [19].

Figure A2. Illustration of the strategy used to build a non-adiabatic Flamelet Generated Manifolds. Both plots are derived from the manifold where $T_{low} = 270$ K.

Figure A3. Lewis number for the analyzed components of the mechanism proposed by Marinov [35] at an ambient pressure of 101,325 bar.

Figure A4. Flame propagation speed in mono-dispersed droplet mists composed by different diameters. The propagation speed of adiabatic gas flames (dashed black) and an auxiliary line highlighting their maximum value are added as references.

Figure A5. Flame propagation speed in mono-dispersed droplet mists for different values of l_{inj} . The propagation speed of adiabatic gas flames (dashed line) is added as a reference

Figure A6. Evolution of gas temperature (continuous) and $\dot{S}_{vapor}$ (dashed) along the coordinate s for flames propagating in droplet mists with different diameters at their respective ϕ_{up} .

Figure A7. Evolution of the fuel mass fraction Y_{fuel} (continuous line) and the gas temperature (dashed line) along the coordinate s . *Left:* $\phi_{\text{over}} = 1.00$. *Right:* $\phi_{\text{over}} = 2.75$.

Figure A8. Evolution of the heat release rate (HRR - continuous line) and the gas temperature (dashed line) along the coordinate s . *Left:* $\phi_{over} = 1.00$. *Right:* $\phi_{over} = 2.75$.

Figure A9. Evolution of the mass fractions of formaldehyde $Y_{\text{CH}_2\text{O}}$ (continuous line) and the hydroxyl radical Y_{OH} (dashed line) along the coordinate s . *Left:* $\phi_{\text{over}} = 1.00$. *Right:* $\phi_{\text{over}} = 2.75$.

Figure A10. Evolution of the mass fractions of carbon dioxide Y_{CO_2} (continuous line) and carbon monoxide Y_{CO} (dashed line) along the coordinate s . *Left:* $\phi_{over} = 1.00$. *Right:* $\phi_{over} = 2.75$.

Figure A11. Evolution of the mass fractions of hydrogen Y_{H_2} (continuous line) and acetylene $Y_{C_2H_2}$ (dashed line) along the coordinate s . *Left:* $\phi_{over} = 1.00$. *Right:* $\phi_{over} = 2.75$.

Figure A12. Flame propagation speed in mono-dispersed droplet mists with $d_d^0 = 75\mu\text{m}$ including and omitting the evaporative cooling (EC) effects. The propagation speed of adiabatic gas flames (dashed black) and an auxiliary line highlighting their maximum value are added as references.

Figure A13. Evolution of the fuel mass fraction Y_{fuel} (continuous line) and the gas temperature (dashed line) along the coordinate s for flames with and without evaporative cooling (EC). *Left:* $\phi_{\text{over}} = 1.00$. *Right:* $\phi_{\text{over}} = 2.75$.

Figure A14. Evolution of the mass fractions of hydrogen Y_{H_2} (continuous line) and acetylene $Y_{C_2H_2}$ (dashed line) along the coordinate s for flames with and without evaporative cooling (EC). *Left:* $\phi_{over} = 1.00$. *Right:* $\phi_{over} = 2.75$.

Figure A15. Flame propagation speed in mono-dispersed droplet mists including and omitting differential diffusion transport. The propagation speed of adiabatic gas flames following the mixture averaged (dashed black) and unitary Le (dashed gray) approaches are added as references. Auxiliary lines highlighting their respective maximum values are also included.

Figure A16. Evolution of the fuel mass fraction Y_{fuel} (continuous line) and the gas temperature (dashed line) along the coordinate s . *Left:* $\phi_{\text{over}} = 1.00$. *Right:* $\phi_{\text{over}} = 2.75$.

Figure A17. Evolution of the heat release rate (HRR - continuous line) and the gas temperature (dashed line) along the coordinate s . *Left:* $\phi_{over} = 1.00$. *Right:* $\phi_{over} = 2.75$.

Figure A18. Evolution of the mass fractions of hydrogen Y_{H_2} (continuous line) and acetylene $Y_{C_2H_2}$ (dashed line) along the coordinate s . *Left:* $\phi_{over} = 1.00$. *Right:* $\phi_{over} = 2.75$.

Figure A19. Evaluation of the prediction ability of the FGM based on gas flamelets with detailed chemistry (DC) for flames propagating in droplet mists.

Figure A20. Evolution of the gas temperature along the coordinate s for different diameters and $\phi_{over} = 1.00$.

Figure A21. Evolution of $\dot{\omega}_{pv}$, $\dot{S}_{vapor}$, and droplet diameter along the coordinate s for $d_d^0 = 50 \mu\text{m}$ and $\phi_{over} = 1.00$.

Figure A22. Evolution of the gas temperature, enthalpy, and mixture fraction at the pre-vaporization zone (see Fig. A1) along the coordinate s for $d_d^0 = 50 \mu\text{m}$ and $\phi_{over} = 1.00$.

Figure A23. Evolution of Y_{CO_2} and Y_{OH} along the coordinate s , as well as the profile of Y_{OH} in function of Y_{CO_2} for $d_d^0 = 50 \mu\text{m}$ and $\phi_{\text{over}} = 1.00$.

Figure A24. Evolution of Y_{pv} , h , and gas temperature in function of the mixture fraction for $d_d^0 = 50 \mu\text{m}$ and $\phi_{over} = 1.00$.

Figure A25. Evolution of the gas temperature along the coordinate s for different values of ϕ_{over} and $d_d^0 = 50 \mu\text{m}$.

Figure A26. Evolution of Y_{pv} in function of the mixture fraction for different values of ϕ_{over} and $d_d^0 = 50 \mu\text{m}$.

Figure A27. Evolution of gas temperature in function of the mixture fraction for different values of ϕ_{over} and $d_a^0 = 50 \mu\text{m}$.

Figure A28. Comparisons between profiles obtained with and without the inclusion of the two-way coupling of momentum for gas and droplets velocities.