

## Highlights

### **On the effects of the fuel injection phase on heat release and soot formation in counterflow flames**

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- The fuel injection phase plays an important role in optimizing combustion processes.
- Ethanol flames consistently release more heat and less soot than n-butanol flames.
- Spray flames at low strain rates produce more heat and less soot than gas flames.
- Flames close to extinction produce more heat and less soot than at low strain rates.

# On the effects of the fuel injection phase on heat release and soot formation in counterflow flames

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## Abstract

Total rates of heat release,  $\dot{\omega}_{T,\text{tot}}$ , and soot emission,  $\dot{\omega}_{Y_s,\text{tot}}$ , are studied in ethanol-N<sub>2</sub>/air and n-butanol-N<sub>2</sub>/air counterflow flames. A gas flame is first established as a base case and fractions of the gaseous fuel are then replaced by droplets and nitrogen, keeping the total fuel mass flux constant. Several values of the liquid to total fuel mass ratio,  $\varphi_l$ , are employed, covering the entire range from a gas flame to a spray flame ( $0 \leq \varphi_l \leq 1$ ). Different initial droplet radii,  $R_0$ , are considered, as well as both low and close to extinction strain rates. Results show qualitative similarities for both fuels, even though quantitative differences are observed. In general, ethanol flames release more heat and less soot than n-butanol flames. For low strain rates,  $\varphi_l = 1$  leads to lower soot emissions than for the reference gas flame, independently of  $R_0$ . Also,  $\dot{\omega}_{T,\text{tot}}$  is higher for  $R_0 = 25$  and  $40 \mu\text{m}$ . Close to extinction, increasing  $\varphi_l$  notoriously improved  $\dot{\omega}_{T,\text{tot}}$  without considerably raising  $\dot{\omega}_{Y_s,\text{tot}}$

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for  $R_0 = 25 \text{ }\mu\text{m}$ . Also, for  $R_0 = 40 \text{ }\mu\text{m}$ , there are some particular values of  $\varphi_l$  for which similar situations occur.

These results show that the fuel injection phase plays an important role in optimizing combustion processes.

*Keywords:* biofuels, spray flames, counterflow, heat release, soot formation, evaporation

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## 1. Introduction

Oxygenated biofuels represent a very attractive alternative to conventional fuels. They are renewable (with a neutral  $\text{CO}_2$  cycle), have high energy density values, and tend to emit less soot than typically used fossil fuels, among other advantages [1, 2]. Ethanol and n-butanol are both in this category and they are widely recognized as promising candidates to replace gasoline in combustion engines [1, 3, 4]. This has motivated intensive research on the combustion characteristics of these biofuels, where both fundamental studies of flame structures in canonical configurations [5, 6, 7, 8, 9, 10] and applied studies of their performance in internal combustion engines have been performed [11, 12, 13, 14]. However, there are still several aspects requiring further investigation. Since both fuels are in liquid form at standard conditions, there is a great potential for optimization yet to be explored. More specifically, while it is well known that parameters such as the initial droplet radius, initial droplet velocity, equivalence ratio and the initial degree of pre-vaporization of the liquid fuel can strongly influence the structure of spray flames [15, 16, 17, 18, 19, 20, 21], the effects that varying them would have on the soot emission and heat release rates have not yet been systematically

studied and only few works have been reported in the literature addressing some of these and other related topics [22, 23, 24, 25]. These, however, have shown some very interesting results still requiring fundamental explanation, such as the fact that using liquid ethanol in engines leads to higher total heat release rates than employing liquid n-butanol [11, 12, 13, 14]. This observation, made for pure fuels as well as for blends with heptane, gasoline and diesel, is not consistent with the fact that the heating value of n-butanol is higher than that of ethanol. While the aforementioned works provide some suggestions regarding how evaporation could have influenced these results, a more elaborated explanation requires a detailed analysis and comparison of the structure of n-butanol and ethanol spray flames.

In this context, the counterflow configuration is a very attractive alternative for the fundamental study of different phenomena found in flames. It has a well defined flow structure, which can be appropriately described by means of a one-dimensional mathematical formulation. This leads to reduced computational costs and allows the inclusion of evaporation, soot formation and detailed chemistry and transport processes, among others. Varying boundary conditions in this configuration is also very easy, which facilitates carrying out parametric studies. All these advantages have been extensively used in previous works studying various fuels burning under several different conditions [26, 27, 28, 29, 30, 31]. Additionally, flamelet-based models developed for the simulation of turbulent flames, such as the ones found in internal combustion engines, consider counterflow flames as their most fundamental building block [32, 33, 34, 35, 36]. In particular, the spray flamelet model considers mono-disperse laminar spray flames generated with different initial

droplet radii (representing different Sauter mean diameters) for the generation of the required pre-tabulated spray flamelet libraries [37, 38, 39]. This represents an important additional motivation for the study of these canonical flames, since developing a sound understanding of them can lead to direct major improvements in the simulation of flames operating under engine-like conditions.

The general objective of this work is to develop new fundamental knowledge that can lead to innovative strategies for the improvement of combustion processes, with special emphasis on the effects of partial or total injection of the fuel in liquid phase. More specifically, we aim to determine whether, and under which conditions, it would be possible to simultaneously achieve higher total energy release and lower total soot emissions rates. For this, parametric studies of counterflow flames at low and close to extinction strain rates are systematically performed for ethanol and n-butanol flames. In particular, a gas flame with a stream of fuel and nitrogen (equal parts in mass) directed against an air stream is first established as a base case and then modified by replacing fractions of gas fuel by droplets and nitrogen, always ensuring the total fuel mass flux is not modified. Boundary conditions for velocities and temperatures are fixed for all cases studied. Simulations are then carried out for several values of the liquid to total fuel mass ratio,  $\varphi_l = \dot{m}_{F,\text{liq}}/\dot{m}_F$ , where  $\dot{m}_{F,\text{liq}}$  and  $\dot{m}_F$  are the initial mass flow rates of the liquid fuel and total fuel. The entire range from a gas flame ( $\varphi_l = 0$ ) to a mono-disperse spray flame ( $\varphi_l = 1$ ) is covered. For this analysis, initial droplet radii of 5, 25 and 40  $\mu\text{m}$  are considered for the mono-disperse spray. The paper has the following structure: First, we introduce the details of the mathematical

model employed for the numerical simulations. Then, a summary of the investigated cases are presented followed by the results and discussion. The paper closes with a summary of results and final conclusions.

## 2. Mathematical model

A two-dimensional Eulerian/Lagrangian formulation is adopted, which is transformed into a one-dimensional system for the gas phase equations by means of a similarity analysis. This model has been first presented by Continillo and Sirignano [15] and was later extended by Gutheil and Sirignano [16] to include detailed chemistry and variable transport properties. A mono-disperse spray is assumed, where heating and evaporation of spherically symmetric single-component droplets are considered. The droplet motion is formulated in a Lagrangian way [40]. The detailed chemical reaction mechanisms of Marinov [41] (57 species and 383 reactions) and Brady et al. [42] (120 species and 832 reactions) are adopted for ethanol and n-butanol, respectively. This model is coupled with the equations for the soot mass fraction and the soot number density originally proposed by Leung et al. [43] and later extended by Carbonell et al. [44]. A detailed description of the governing equations is given below.

### 2.1. Gas phase

For steady, axisymmetric counterflow spray flames, the governing equations for total mass and momentum in the radial direction can be written as [16],

$$\frac{1}{x} \frac{\partial(x\rho u)}{\partial x} + \frac{d(\rho v)}{dy} = \dot{S}_v, \quad (1)$$

and

$$\rho \left( u \frac{\partial u}{\partial x} + v \frac{\partial u}{\partial y} \right) = -\frac{dp}{dx} + \frac{\partial}{\partial y} \left( \mu \frac{\partial u}{\partial y} \right) + \dot{S}_m, \quad (2)$$

respectively, where the boundary layer approximation and a low-Mach number assumption have been adopted. In these equations,  $y$  and  $x$  denote the axial and radial coordinates while  $v$  and  $u$  are the axial and radial components of the gas velocity.  $\rho$ ,  $p$ , and  $\mu$  are the density, the pressure and the dynamic viscosity of the gas, respectively.  $\dot{S}_v$  and  $\dot{S}_m$  are source terms due to the exchange of mass and momentum between both phases. A similarity transformation allows transferring the two-dimensional gas equations for the mass fractions,  $Y_k$ , of chemical species  $k$  ( $k = 1, \dots, N$ ), into a one-dimensional formulation [15, 16]. This yields

$$\rho v \frac{dY_k}{dy} = -\frac{d(\rho V_{ky} Y_k)}{dy} + \dot{\omega}_k + (\delta_{kF} - Y_k) \dot{S}_v, \quad (3)$$

where  $\dot{\omega}_k$  is the specific reaction rate, and  $\delta_{kF}$  is the Kronecker delta, where  $F$  refers to fuel. The term including the diffusion velocity is calculated as

$$\rho V_{ky} Y_k = -\rho D_k \frac{Y_k}{X_k} \frac{\partial X_k}{\partial y} - \frac{D_{T,k}}{T} \frac{dT}{dy} + \rho Y_k V_y^C, \quad (4)$$

where  $X_k$  is the mole fraction of species  $k$ .  $D_k$  and  $D_{T,k}$  are the molecular and thermal diffusion coefficients of species  $k$ , respectively. The velocity correction ensuring mass conservation is

$$V_y^C = \sum_{k=1}^N \left( D_k \frac{Y_k}{X_k} \frac{\partial X_k}{\partial y} + \frac{D_{T,k}}{T} \frac{dT}{dy} \right). \quad (5)$$

The similarity analysis also allows consideration of a one-dimensional transport equation for the gas temperature,  $T$ , which can be expressed as

$$\rho c_p v \frac{dT}{dy} = \frac{d}{dy} \left( \lambda \frac{dT}{dy} \right) + \rho \sum_{k=1}^N V_k c_{pk} \frac{dT}{dy} + \dot{\omega}_T + \dot{S}_e, \quad (6)$$

where  $c_{p,k}$  and  $c_p$  are the specific heat capacity at constant pressure of species  $k$  and of the gas mixture, respectively.  $\lambda$  is the thermal conductivity,  $\dot{\omega}_T$  is the heat release rate, and  $\dot{S}_e$  is the source term due to the exchange of energy between both phases. The heat release is

$$\dot{\omega}_T = - \sum_{k=1}^N h_k \dot{\omega}_k, \quad (7)$$

where  $h_k$  is the enthalpy of species  $k$ . All transport coefficients are computed following Kee et al. [45]. The gas phase equations are completed by the following equation of state:

$$\rho = \frac{p\overline{M}}{RT}, \quad (8)$$

where  $\overline{M}$  is the molecular weight of the mixture and  $R_u$  is the universal gas constant. Spatially integrating the chemical source of energy and the soot mass source rate, we define the total rates of heat release, fuel consumption and soot emission, respectively, as

$$\dot{\omega}_{T,\text{tot}} = \int_{-\infty}^{+\infty} \dot{\omega}_T dy, \quad (9)$$

$$\dot{\omega}_{F,\text{tot}} = \int_{-\infty}^{+\infty} \dot{\omega}_F dy, \quad (10)$$

$$\dot{\omega}_{Y_s,\text{tot}} = \int_{-\infty}^{+\infty} \dot{\omega}_{Y_s} dy, \quad (11)$$

where a perpendicular area of  $1 \text{ m}^2$  has been considered. The transport equations for the soot particle number density,  $N_s$  and the soot mass fraction,  $Y_s$ , are [43]

$$\rho \frac{\partial N_s}{\partial t} + \rho v \frac{\partial N_s}{\partial y} = - \frac{\partial}{\partial y} \left( 0.55 \frac{\mu}{\rho T} \frac{\partial T}{\partial y} \right) + \dot{\omega}_{N_s} - N_s \dot{S}_v \quad (12)$$

and

$$\rho \frac{\partial Y_s}{\partial t} + \rho v \frac{\partial Y_s}{\partial y} = - \frac{\partial}{\partial y} \left( 0.55 \frac{\mu}{\rho T} \frac{\partial T}{\partial y} \right) + \dot{\omega}_{Y_s} - Y_s \dot{S}_v, \quad (13)$$

respectively. In Equations 12 and 13, the diffusion term is expressed only by the thermophoretic effects [46], and molecular diffusion is neglected. Some additional quantities used in this study are the soot volume fraction

$$f_v = \frac{\rho Y_s}{\rho_s}, \quad (14)$$

the soot mean particle diameter

$$d_p = \left( \frac{6 Y_s}{\pi \rho_s N_s} \right)^{1/3}, \quad (15)$$

and the total soot surface per unit volume,

$$A_s = \pi d_p^2 \rho N_s, \quad (16)$$

all of which are dependent variables on  $N_s$  and  $Y_s$ .  $\rho_s$  denotes the soot density, estimated with a value of 2000 kg/m<sup>3</sup> [43]. The soot source terms in Eqs. (13) and (12) are

$$\dot{\omega}_{N_s} = \frac{2}{\text{NC}_{\min}} \frac{N_A}{M_s} \dot{\omega}_n - \dot{\omega}_a \quad (17)$$

and

$$\dot{\omega}_{Y_s} = \dot{\omega}_n + \dot{\omega}_g - \dot{\omega}_{ox, O_2} - \dot{\omega}_{ox, OH} - \dot{\omega}_{ox, O}. \quad (18)$$

In Equations 17 and 18, subscript “ $n$ ” denotes nucleation, “ $a$ ” denotes agglomeration, “ $g$ ” stands for surface growth, and “ $ox$ ” indicates oxidation caused by O<sub>2</sub>, OH, and O. NC<sub>min</sub> is the number of atoms of C in the incipient soot particles,  $N_A$  is the Avogadro number. Table 1 illustrates the source rate expressions, while the corresponding constants are presented in Table 2.

Table 1: Soot mass source rates [44].

| Reaction                                                                      | Molar rates                                                       |
|-------------------------------------------------------------------------------|-------------------------------------------------------------------|
| $\text{C}_2\text{H}_2 \rightarrow 2\text{C}_s + \text{H}_2$                   | $\dot{\omega}_n = k_n[\text{C}_2\text{H}_2]M_s$                   |
| $\text{C}_2\text{H}_2 + n\text{C}_s \rightarrow (n+2)\text{C}_2 + \text{H}_2$ | $\dot{\omega}_g = k_g[\text{C}_2\text{H}_2]M_s\sqrt{A_s}$         |
| $\text{C}_s + 0.5\text{O}_2 \rightarrow \text{CO}$                            | $\dot{\omega}_{ox,\text{O}_2} \rightarrow \text{NSC-model}$       |
| $\text{C}_s + \text{OH} \rightarrow \text{CO} + \text{H}$                     | $\dot{\omega}_{ox,\text{OH}} = 0.2k_{ox,\text{OH}}[\text{OH}]A_s$ |
| $\text{C}_s + \text{O} \rightarrow \text{CO}$                                 | $\dot{\omega}_{ox,\text{O}} = 0.2k_{ox,\text{O}}[\text{O}]A_s$    |

These constants were taken from the work of Carbonell et al. [44]. The agglomeration rate is expressed as [43]

$$\dot{\omega}_a = -2C_a d_p^{1/2} \left( \frac{6k_b T}{\rho_s} \right) (\rho N_s)^2, \quad (19)$$

where  $k_b$  is the Boltzmann constant and  $C_a = 9$  is the agglomeration constant [43]. Note that the soot model used in this work has been extensively validated for ethylene flames. However, no extensive validation has been performed for ethanol and n-butanol flames. We assume that the soot

Table 2: Soot rate constants  $k = AT^b \exp(-E/RT)$  (SI units)

| Soot rate constant  | $A$               | $b$ [-] | $E/R$ [K] | Ref. |
|---------------------|-------------------|---------|-----------|------|
| $k_n$               | $1 \cdot 10^4$    | 0.0     | 21, 100   | [43] |
| $k_g$               | $6 \cdot 10^3$    | 0.0     | 12, 100   | [43] |
| $k_{ox,\text{O}_2}$ | NSC-model         |         |           | [47] |
| $k_{ox,\text{OH}}$  | $1.27 \cdot 10^3$ | -0.5    | 0.0       | [48] |
| $k_{ox,\text{O}}$   | $5.54 \cdot 10^1$ | -0.5    | 0.0       | [49] |

formation mechanism for the two fuels investigated in this work could be described by the model with sufficient accuracy and that the rate constants are the same for both fuels. Future work should focus on the use of more advanced soot models with more detailed description of the soot formation process (e.g., sectional methods).

Dirichlet boundary conditions are considered for the gas phase equations. The fuel inlet is located at a distance  $L_F$  from the gas stagnation plane ( $y = 0$ ), while the air inlet is located at an infinite distance from that position. The gas strain rate is imposed at the fuel inlet, assuming a potential flow [15].

## 2.2. Liquid-phase

The equations for the droplet evaporation, heating, motion, and number density of the mono-disperse spray yield

$$-\frac{d}{dt} \left( \frac{4}{3} \pi R^3 \rho_l \right) = 2 \pi \rho_f D_f R \widetilde{Sh} \ln(1 + B_M), \quad (20)$$

$$\frac{\partial T_l}{\partial t} = \alpha_l \frac{1}{r^2} \frac{\partial}{\partial r} \left( r^2 \frac{\partial T_l}{\partial r} \right), \quad (21)$$

$$\frac{4}{3} \pi R^3 \rho_l \frac{d\mathbf{V}_l}{dt} = \pi R^2 \frac{1}{2} \rho (\mathbf{V} - \mathbf{V}_l) |\mathbf{V} - \mathbf{V}_l| C_D \quad (22)$$

and

$$\frac{1}{x} \frac{\partial(x n u_l)}{\partial x} + \frac{\partial(n v_l)}{\partial y} = 0, \quad (23)$$

respectively. In Equations (20)-(23),  $\rho_l$ ,  $T_l$ , and  $\alpha_l$  are the density, temperature and thermal diffusivity of the droplet, respectively.  $R$  denotes the instantaneous droplet radius, and  $n$  the droplet number density.  $\rho_f$  and  $D_f$  are the density and the molecular diffusivity of the gas mixture in the film,

respectively.  $v_l$  and  $u_l$  are, respectively, the axial and radial components of the droplet velocity,  $\mathbf{V}_l$ .  $B_M$  is the mass transfer number, and  $C_D$  is the drag coefficient [15].  $\widetilde{Sh}$  is the modified Sherwood number [40] and the vapor pressure is calculated using the Wagner equation [50]. Equation 23 can be solved analytically, which yields

$$n = \frac{n_0 v_{l0} x_{l0}^2}{v_l x_l^2}. \quad (24)$$

The equations for the gas phase and the liquid phase are coupled by means of the spray source terms. The effect of all droplets is represented by one size group at each grid point. Thus, the expressions for the source terms of mass, momentum and energy, cf. Eqs. (1), (2), (3) and (6), yield

$$\dot{S}_v = n\dot{m}, \quad (25)$$

$$\dot{S}_m = -n \left( m_l \frac{du_l}{dt} + \dot{m}(u - u_l) \right), \quad (26)$$

$$\dot{S}_e = -n (\dot{q}_L + \dot{m} [C_{p,f}(T - T_s) + L_v(T_s)]), \quad (27)$$

Table 3: Properties of the liquid fuels [50].

|                                     | Ethanol | n-Butanol |
|-------------------------------------|---------|-----------|
| Lower heating value (LHV) [MJ/kg]   | 26.84   | 33.11     |
| Density [kg/m <sup>3</sup> ]        | 787     | 810       |
| Boiling temperature [K]             | 351     | 391       |
| Critical temperature [K]            | 513     | 563       |
| Specific heat capacity [J/(mol K)]  | 112.3   | 177.1     |
| Latent heat of vaporization [kJ/kg] | 943.8   | 619.8     |

where  $\dot{q}_L = \dot{m} [C_{p,f} (T - T_s) / B_T - L_v(T_s)]$  is the heat transferred to the droplet and  $m_l$  is the droplet mass.  $t$  is the time,  $c_{p,f}$  is the average vapor specific heat in the film,  $B_T$  is the Spalding heat transfer number,  $T$  is the temperature of the surrounding gas,  $T_s$  is the temperature of the droplet surface, and  $L_v(T_s)$  is the latent heat of vaporization at temperature  $T_s$ . The properties of ethanol and n-butanol are listed in Table 3. The initial values of the radial position, velocity and temperature of the droplets are fixed. The temperature gradient of a droplet at its surface is given by

$$\left. \frac{\partial T_l}{\partial r} \right|_{r=R(t)} = \frac{\dot{q}_L}{4\pi R^2 \alpha_l \rho_l c_{pl}}, \quad (28)$$

where  $c_{pl}$  denotes the specific heat of the droplet. As a consequence of the spherical shape of the droplets, a zero-gradient condition is applied at  $r = 0$ .

### 3. Target flames

A gas flame ( $\varphi_l = 0$ ) with a mixture composed of 50% gaseous fuel and 50%  $N_2$  by mass at 470 K directed against a stream of pure air at 300 K is

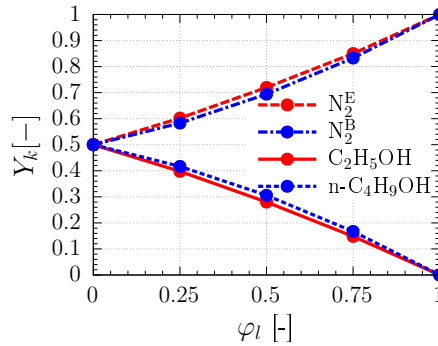


Figure 1: Mass fractions of fuel and  $N_2$  at the fuel inlet. Superscripts “E” and “B” are for ethanol and n-butanol flames, respectively.

first established as a reference case. After this, fractions of the gaseous fuel are replaced by liquid fuel (at 300 K) and  $N_2$ , systematically increasing  $\varphi_l$  from zero to one, while the initial flow conditions and the total fuel flow rate are kept constant. Figure 1 shows the mass fractions of gaseous fuel and  $N_2$  in the inlet gas as a function of  $\varphi_l$ . The pressure is equal to 1 atm and the analyses are conducted for low and high strain rate conditions, where the latter correspond to situations close to extinction. The details of the inlet conditions for all flames studied are shown in Table 4. The mass flow rates of the liquid and gas fuel can be obtained by multiplying the fuel mass flow rate from Table 4 by  $\varphi_l$  and  $(1 - \varphi_l)$ , respectively. Three different values of the initial droplet radius are considered: 5, 25 and 40  $\mu\text{m}$ , and the studied flames are classified using the following general notation:  $\gamma_{\varphi_l}^{R_0}$ , where  $\varphi$  indicates the fuel ( $E$  for ethanol or  $B$  for n-butanol), and the subscript and superscript denote the values of  $\varphi_l$  and  $R_0$ , respectively. For example, a n-butanol flame with  $R_0 = 5 \mu\text{m}$  and  $\varphi_l = 1.0$  is designated as  $B_{1.0}^5$ . It is important to remark that all the flames analyzed in this work have a non-premixed nature, which means that, despite having globally rich conditions, their reaction zones are located near to the position of stoichiometric mixture [18]. We also highlight

Table 4: Boundary conditions of the base case at the fuel inlet.

|                                      |       |
|--------------------------------------|-------|
| Fuel mass flow rate (Butanol) [kg/s] | 0.369 |
| Fuel mass flow rate (Ethanol) [kg/s] | 0.316 |
| Gas temperature [K]                  | 470   |
| Liquid temperature [K]               | 300   |
| Gas and liquid velocity [m/s]        | 0.7   |

that, even though laminar monodisperse spray flames are considered here, these are very relevant for the simulation of turbulent polydisperse spray flames by means of flamelet-based approaches [37, 38, 39].

## 4. Results and discussion

### 4.1. Low strain rate

In this subsection, flame structures obtained for a strain rate of 55/s are discussed. In Figs. 2a and 2b, the gas temperature profiles for both fuels

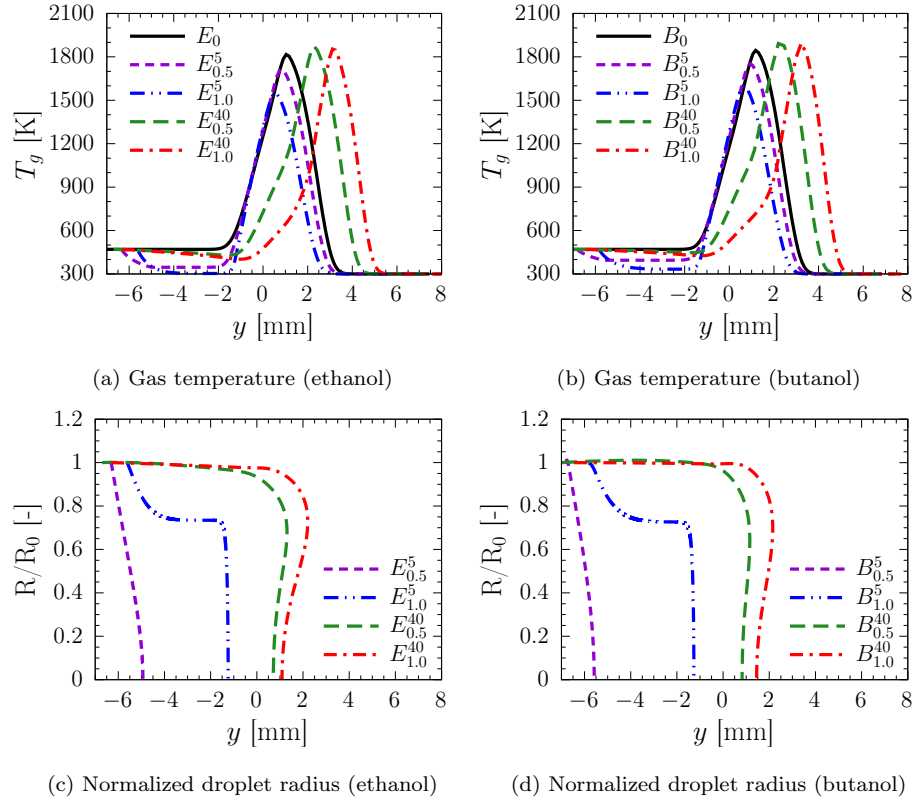


Figure 2: Gas temperature (top) and droplet radius (bottom) for five different flame configurations:  $\gamma_0$ ,  $\gamma_{0.5}^5$ ,  $\gamma_{1.0}^5$ ,  $\gamma_{0.5}^{40}$  and  $\gamma_{1.0}^{40}$ .

are presented. Since both fuels show qualitatively similar responses to variations of  $R_0$  and  $\varphi_l$ , the results will be analyzed in terms of the generic notation described in the previous section. Specifically, the flames studied in this subsection are:  $\gamma_0$ ,  $\gamma_{0.5}^5$ ,  $\gamma_{1.0}^5$ ,  $\gamma_{0.5}^{40}$  and  $\gamma_{1.0}^{40}$ . Compared to the gas flame,  $\gamma_0$ , a lower temperature is observed for flames  $\gamma_{0.5}^5$  and  $\gamma_{1.0}^5$ , especially in the air side and in regions close to the fuel inlet. In contrast, the temperature profiles for flames  $\gamma_{0.5}^{40}$  and  $\gamma_{1.0}^{40}$  present a displacement to the air side and somewhat higher maximum values than the gas flame. This is consistent with the enhanced droplet penetration into the reaction zone associated with a larger droplet radius and its corresponding higher momentum [16] (see Figs. 2c and 2d). Comparing these structures for both fuels, it can be observed that the gas temperature is more reduced on the left side of the flame due to the evaporation in that region. The n-butanol flames tend to be more displaced to the right than ethanol flames due to the higher momentum associated with the fuel droplets.

Figure 3 shows the heat release,  $\dot{\omega}_T$ , and the energy sink term due to evaporation,  $\dot{S}_e$ , for all flames under consideration. It is observed that for  $\gamma_{0.5}^5$  and  $\gamma_{1.0}^5$ , the fast evaporation of droplets close to the fuel inlet leads to higher values of the associated energy sink, which locally reduces the temperature in relation to the gas flame. This also leads to a corresponding reduction of  $\dot{\omega}_T$  in these flames. For cases  $\gamma_{0.5}^{40}$  and  $\gamma_{1.0}^{40}$ , on the other hand, the heat release is higher than for  $\gamma_0$ , and the peak of the profile is displaced to the air side. While the magnitudes of  $\dot{S}_e$  also increase for these cases, this is not enough to compensate the higher heat release and, therefore, the temperature increases. At  $R_0 = 5 \text{ } \mu\text{m}$ , the heat release for both fuels is

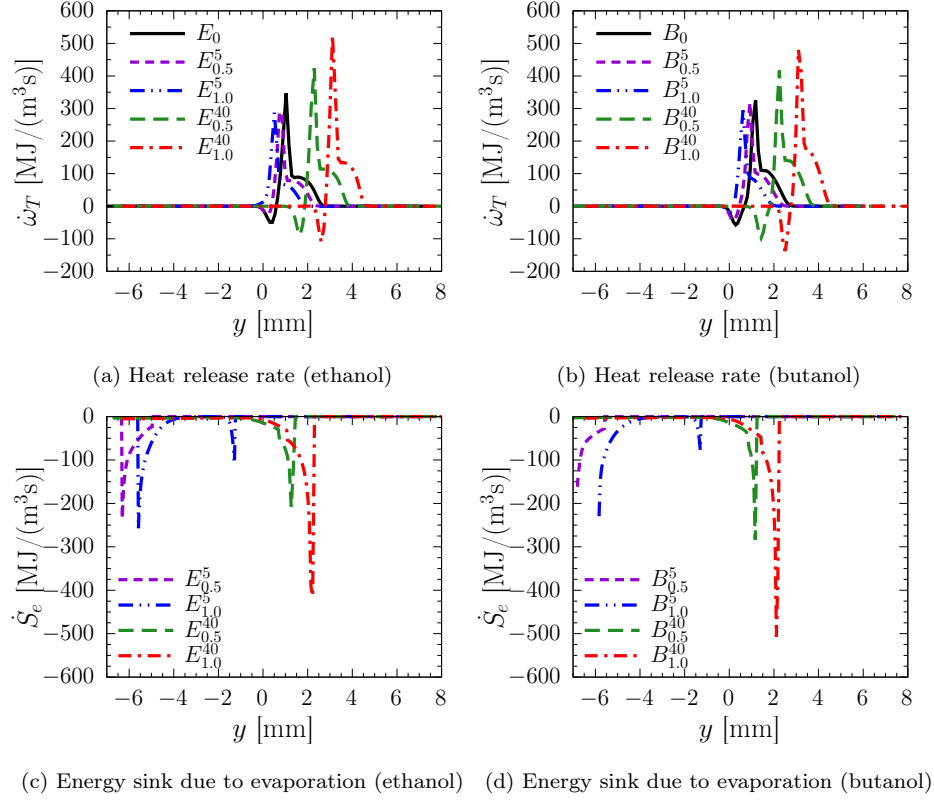


Figure 3: Local heat release (top) and energy sink due to evaporation (bottom) for five different flame configurations:  $\gamma_0$ ,  $\gamma_{0.5}^5$ ,  $\gamma_{1.0}^5$ ,  $\gamma_{0.5}^{40}$  and  $\gamma_{1.0}^{40}$ .

similar, but  $\dot{S}_e$  is higher for ethanol, which would explain the temperature decrease on the left side of the flame (Figs. 2a and 2b). For  $R_0 = 40 \mu\text{m}$ , the heat release of ethanol is higher than for n-butanol, but the energy sink is also high, and therefore, the temperature values are similar for both.

Profiles of the  $\text{C}_2\text{H}_2$  mass fraction are presented in Figs. 4a and 4b. This species, which is the precursor for soot nucleation and a relevant factor for soot surface growth in the model used in this study, shows some relation with the temperature values. Similarly to temperature, for cases considering

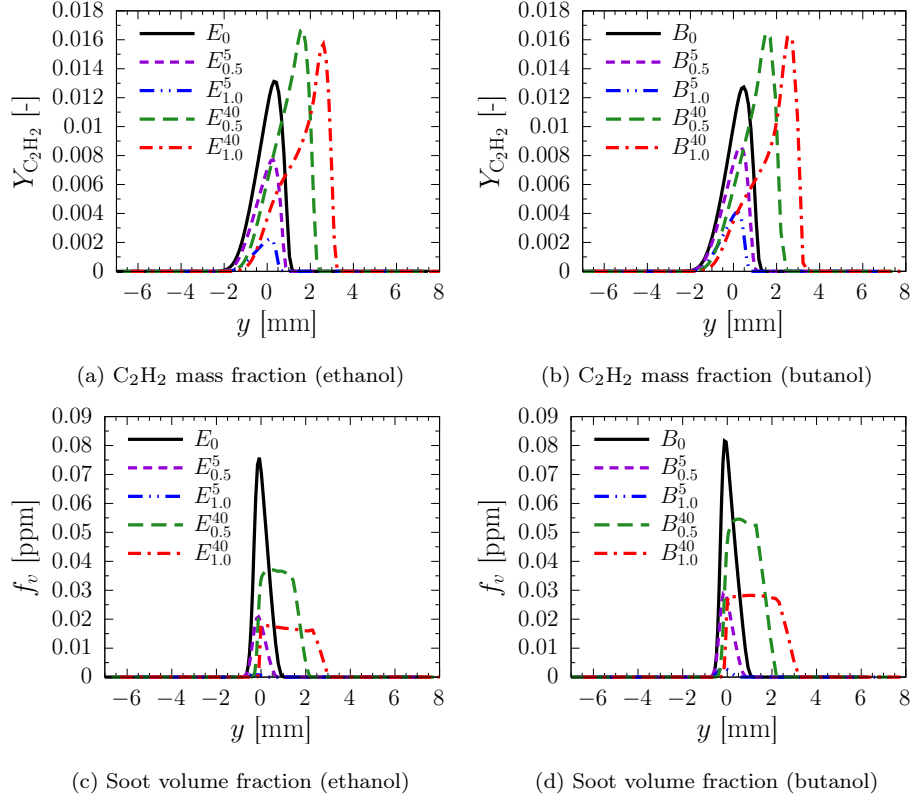


Figure 4:  $C_2H_2$  mass fraction (top) and soot volume fraction (bottom) for five different flame configurations:  $\gamma_0$ ,  $\gamma_{0.5}^5$ ,  $\gamma_{1.0}^5$ ,  $\gamma_{0.5}^{40}$  and  $\gamma_{1.0}^{40}$ .

small droplets,  $Y_{C_2H_2}$  is lower than for  $\gamma_0$ , and a higher concentration of this species is observed when larger droplets are employed. The corresponding soot volume fraction profiles are illustrated in Figs. 4c and 4d. For flames  $\gamma_{0.5}^5$  and  $\gamma_{1.0}^5$ , the soot concentrations are lower than for the gas flame, with the minimum values found for flame  $\gamma_{1.0}^5$ . This is a consequence of the low temperatures and  $C_2H_2$  mass fractions obtained for this case. While for  $\gamma_{0.5}^{40}$  and  $\gamma_{1.0}^{40}$  the maximum value of  $f_v$  is also lower than for the gas flame, it will be shown further below that the total soot formation rate is indeed increased

for these cases. The reduction of the peak value of  $f_v$  can be attributed to the displacement of the soot reaction zone to the air side, which enhances the transport of soot due to convection and diffusion from the reaction zone to the stagnation position. Also, the  $C_2H_2$  mass fraction and the soot volume fraction always reach higher values in the n-butanol flames than in the ethanol flames.

For a better understanding of the structures presented above, we proceed now to analyze the total rates of heat release,  $\dot{\omega}_{T,\text{tot}}$ , fuel consumption,  $\dot{\omega}_{F,\text{tot}}$ , and soot emission,  $\dot{\omega}_{Y_s,\text{tot}}$ , which are displayed in Fig. 5. These quantities are calculated by means of Eqs. 9, 10 and 11, as a function of  $\varphi_l$  and  $R_0$ . For  $R_0 = 5 \mu\text{m}$ , the total heat release monotonically decreases when  $\varphi_l$  is increased. In contrast, increasing  $\varphi_l$  for  $R_0 = 25$  and  $40 \mu\text{m}$  leads to a monotonic increase of the total heat release rate, with a higher slope for the highest droplet radius. These general tendencies are exhibited by flames of both fuels. All the studied flames show higher total heat release rates for ethanol than for n-butanol. This result contradicts the fact that the heating value and the inlet mass flow for n-butanol are higher than for ethanol

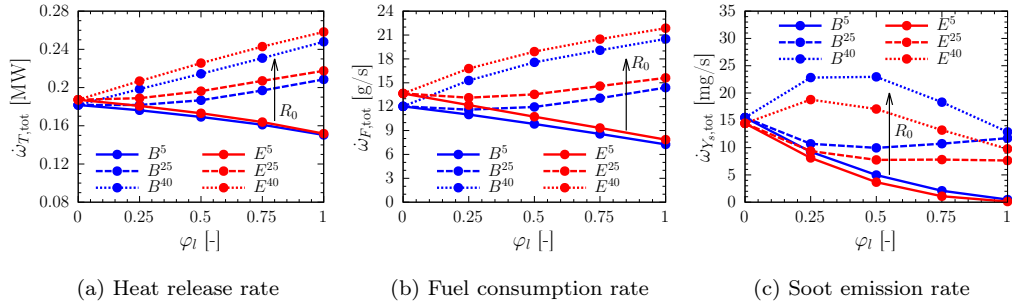
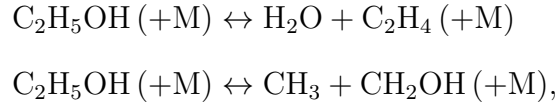
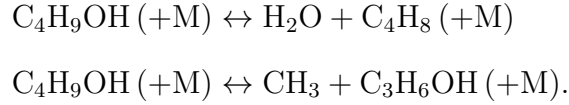


Figure 5: Total rates of (a) heat release, (b) fuel consumption and (c) soot emission in terms of  $\varphi_l$  and  $R_0$ .

(see Tables 3 and 4). However, this phenomenon can be explained by the higher total fuel consumption rates reached by the ethanol flame, which are consistently higher than the corresponding rates for n-butanol, as shown in Fig. 5b. This can be attributed to two different factors. First, it should be noted that 1 kg of n-butanol requires more mass of  $O_2$  (2.59 kg) than 1 kg of ethanol (2.09 kg) to be burnt in stoichiometric conditions and that, for the cases under consideration, the mass flow rate of n-butanol is actually higher than the one of ethanol (Table 4). Secondly, larger pre-exponential factors (since they are functions of the temperature) in the pyrolysis reactions of ethanol [41],



when compared with the pyrolysis reactions of n-butanol [42],



Thus, despite the higher heating value of n-butanol, and even when a higher inlet mass flow rate is considered for this fuel, higher heat release rates are observed for ethanol, since not all n-butanol is effectively reacting.

Regarding the total soot emission rate (Fig. 5c), for  $R_0 = 5 \mu m$  this variable decreases considerably when  $\varphi_l$  is increased, which is consistent with the profiles displayed in Figs. 4c and 4d. For  $R_0 = 25 \mu m$ ,  $\dot{\omega}_{Y_s, tot}$  also decreases when  $\varphi_l$  is increased, but not as significantly as for  $R_0 = 5 \mu m$ . This is especially true for high values of the liquid to total mass flow rate. Note that  $\dot{\omega}_{Y_s, tot}$  increases from  $\varphi_l = 0.5$  to  $\varphi_l = 1.0$ , but its value in this interval

is always lower than for the gas flame. For  $R_0 = 40 \text{ }\mu\text{m}$ ,  $\dot{\omega}_{Y_s, \text{tot}}$  increases when  $\varphi_l$  varies from 0 to 0.25, which results from the higher temperatures and  $\text{C}_2\text{H}_2$  concentrations compared to the gas flame. When  $\varphi_l$  is further increased,  $\dot{\omega}_{Y_s, \text{tot}}$  starts to decrease, reaching a local minimum for  $\varphi_l = 1$ , which is lower than for the gas flame. This occurs as a consequence of the flame displacement to the air side, where  $\text{C}_2\text{H}_2$  concentrations are lower inside the reaction zone.

In conclusion, for the low strain rate situations analyzed in this subsection, spray flames ( $\varphi_l = 1$ ) are found to have lower soot emissions than the reference gas flame, independently of the initial droplet radius considered. Also, a simultaneous higher amount of total heat release is observed for initial droplet radii of 25 and 40  $\mu\text{m}$ . Moreover, it has been observed that ethanol flames consistently release more energy and lead to less soot than n-butanol flames, which is attributable to the higher total consumption rates of ethanol.

#### 4.2. Strain rates close to extinction

Flames at strain rates close to extinction are analyzed in order to determine how the results obtained in the previous subsection for low strain rate are modified under these conditions. For this, the extinction strain rate,  $a_{ext}$ , is first determined for each of the cases previously studied. Figure 6 shows  $a_{ext}$  as a function of the liquid to total fuel mass ratio,  $\varphi_l$ , for different initial droplet radii,  $R_0$ . For  $R_0 = 5 \text{ }\mu\text{m}$ , the extinction strain rate decreases continuously for both fuels when  $\varphi_l$  is increased, while for  $R_0 = 25 \text{ }\mu\text{m}$  the strain rate shows insensitivity to variations of  $\varphi_l$ . The latter indicates a similar stability between the gas flame and the spray flame. Finally, for  $R_0 = 40 \text{ }\mu\text{m}$

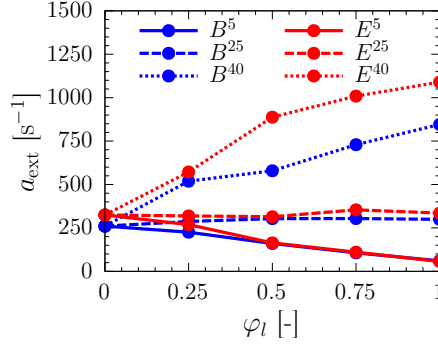


Figure 6: Extinction strain rate in terms of the liquid to total fuel mass ratio for different values of the initial droplet radius.

$a_{ext}$  monotonically grows when  $\varphi_l$  is increased. Additionally, ethanol presents higher extinction strain rates than n-butanol for most of the studied cases (exceptions are  $\gamma_{0.5}^{25}$  and cases for  $R_0 = 5 \mu\text{m}$  with  $\varphi_l \geq 0.5$ ). For  $R_0 = 40 \mu\text{m}$  and  $\varphi_l = 0.5$  the difference in the extinction strain rates reaches its maximum value, which corresponds to  $308.6 \text{ s}^{-1}$ . In the rest of this subsection, flames are classified according to the notation introduced in Section 3 ( $\gamma_{\varphi_l}^{R_0}$ ). We

Table 5: Extinction strain rates in terms of the liquid to total fuel mass ratio and the initial droplet radius.

|             | $R_0 = 5 \mu\text{m}$ |         | $R_0 = 25 \mu\text{m}$ |         | $R_0 = 40 \mu\text{m}$ |         |
|-------------|-----------------------|---------|------------------------|---------|------------------------|---------|
| $\varphi_l$ | Ethanol               | Butanol | Ethanol                | Butanol | Ethanol                | Butanol |
| 0           | 324                   | 260     | 324                    | 260     | 324                    | 260     |
| 0.25        | 269                   | 225     | 319                    | 287     | 572                    | 519     |
| 0.5         | 163                   | 160     | 314                    | 303     | 887                    | 579     |
| 0.75        | 110                   | 106     | 354                    | 304     | 1008                   | 729     |
| 1           | 56                    | 60      | 335                    | 299     | 1089                   | 845     |

highlight, however, that the strain rates considered here are the ones shown in Fig. 6, which are summarized in Table 5 for a better visualization.

Gas temperature profiles for ethanol and n-butanol are shown in Figs. 7a and 7b, respectively. As in the previous subsection, a local minimum due to the evaporation is observed at the left side of the configuration. This phenomenon is especially visible for  $\gamma_{1.0}^5$ , due to the high value of  $\varphi_l$  and the small initial droplet size. Additionally, it can be seen that flame  $\gamma_{1.0}^{40}$  is displaced to the air side, which is associated with the corresponding

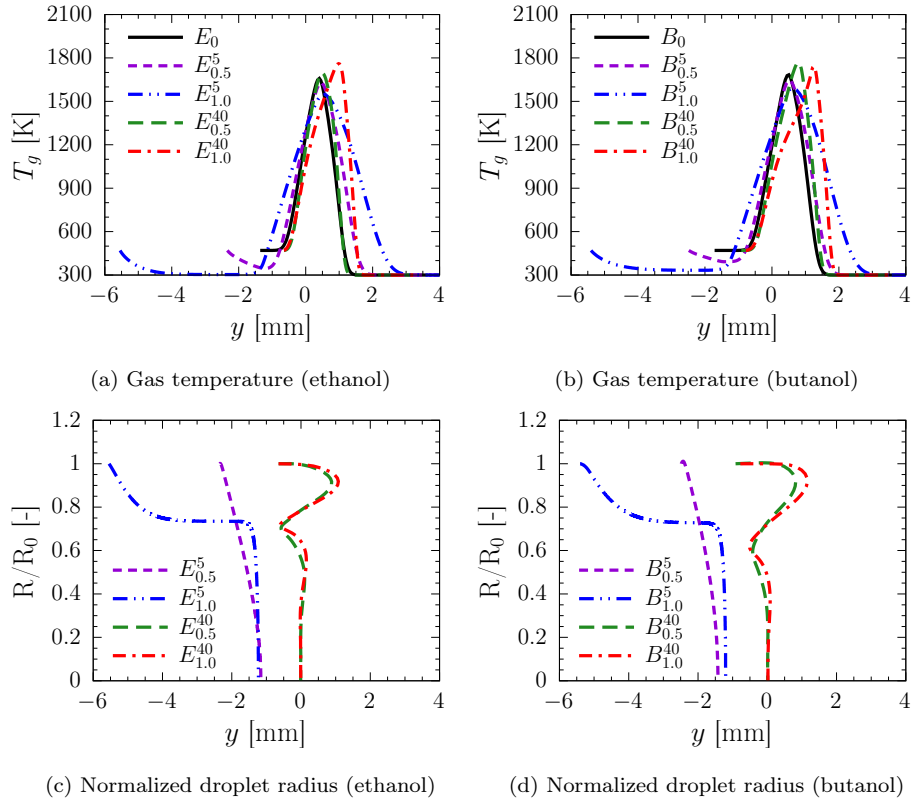


Figure 7: Gas temperature (top) and droplet radius (bottom) for five different flame configurations (close to extinction):  $\gamma_0$ ,  $\gamma_{0.5}^5$ ,  $\gamma_{1.0}^5$ ,  $\gamma_{0.5}^{40}$  and  $\gamma_{1.0}^{40}$ .

displacement of the region of high evaporation due to the enhanced droplet penetration. Figs. 7c and 7d show the normalized droplet radius for both fuels. For flame  $\gamma_{0.5}^5$ , droplets are quickly vaporized due to the small liquid fuel mass flow rate and small droplet size. In flame  $\gamma_{1.0}^5$ , however, droplets travel a much longer distance before they are completely evaporated, which is due to the reduced gas temperature at the left side of the flame discussed above. Despite the associated extended life time, droplets evaporate very quickly once inside the high temperature zone and they never reach the

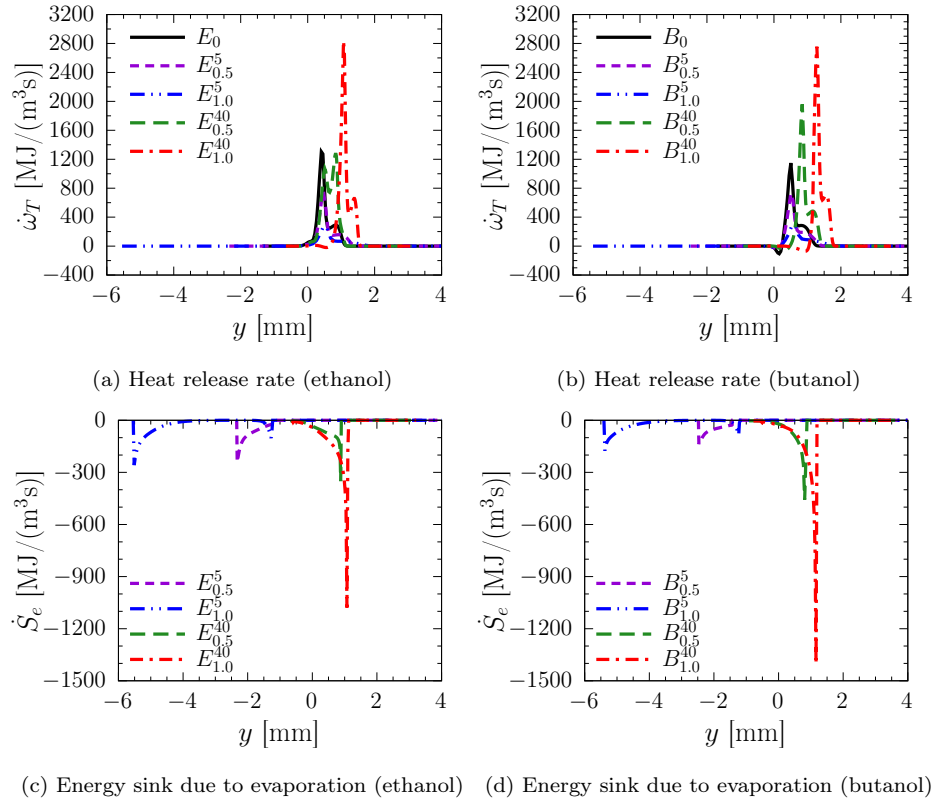


Figure 8: Local heat release (top) and energy sink due to evaporation (bottom) for five different flame configurations (close to extinction):  $\gamma_0$ ,  $\gamma_{0.5}^5$ ,  $\gamma_{1.0}^5$ ,  $\gamma_{0.5}^{40}$  and  $\gamma_{1.0}^{40}$ .

stagnation plane. For  $R_0 = 40 \text{ }\mu\text{m}$ , droplets oscillate multiple times around the stagnation plane due to their high momentum (especially for flame  $\gamma_{1.0}^{40}$ ), distributing the fuel mass in its vicinity.

Profiles of heat release and energy sink due to evaporation are shown in Fig. 8. Considering flame  $\gamma_0$  as a reference case, for both fuels it is observed that the heat release values decrease for  $R_0 = 5 \text{ }\mu\text{m}$  when  $\varphi_l$  is increased, which, in combination with the higher values of  $\dot{S}_e$  near the fuel inlet, leads to a temperature reduction. This is similar to the situation observed for the corresponding low-strain rate case. On the other hand, for  $R_0 = 40 \text{ }\mu\text{m}$ ,  $\dot{\omega}_T$  increases considerably for both fuels as  $\varphi_l$  is raised, which results in a notorious temperature increase for this case (see Figs. 7a and 7b).

Figures 9a and 9b present profiles of  $\text{C}_2\text{H}_2$  mass fraction and soot volume fraction,  $f_v$ . Analogously to the behaviors observed for the low strain rate situations (Fig. 4), for cases with small droplets,  $Y_{\text{C}_2\text{H}_2}$  is lower than for  $\gamma_0$ . For  $R_0 = 40 \text{ }\mu\text{m}$ , different behaviors are found depending on the fuel. For ethanol, moving from the gaseous reference flame to  $\varphi_l = 0.5$  slightly decreases the acetylene mass fraction, which however, increases again for  $\varphi_l = 1$ , reaching considerably higher values than for  $E_0$  in that limit. Conversely, for n-butanol,  $Y_{\text{C}_2\text{H}_2}$  monotonically increases when  $\varphi_l$  is raised. The corresponding soot volume fraction profiles are illustrated in Figs. 9c and 9d. For flames  $\gamma_{0.5}^5$  and  $\gamma_{1.0}^5$ , the soot concentrations are higher than for the reference gas flame, with the maximum value found for flame  $\gamma_{1.0}^5$ . This effect can be attributed to the low strain rates existing in the latter. Similarly to the behavior observed for  $Y_{\text{C}_2\text{H}_2}$ , for flames with  $R_0 = 40 \text{ }\mu\text{m}$ , different results are found depending on the fuel. For ethanol, in the interval from  $\varphi_l = 0$  to

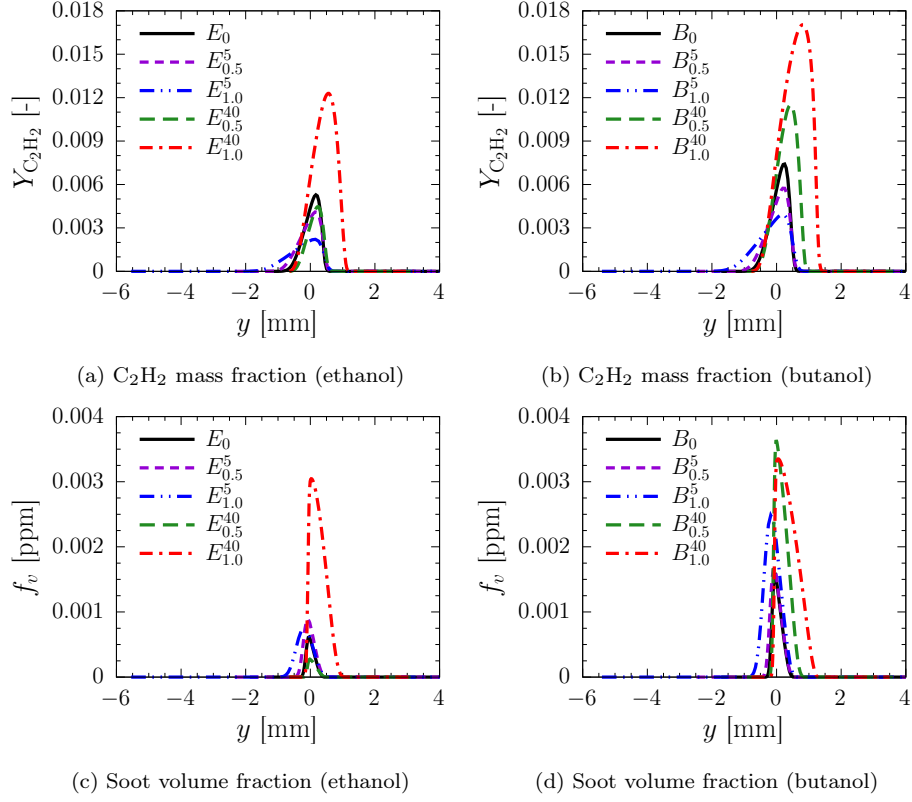


Figure 9: C<sub>2</sub>H<sub>2</sub> mass fraction (top) and soot volume fraction (bottom) for five different flame configurations (close to extinction):  $\gamma_0$ ,  $\gamma_{0.5}^5$ ,  $\gamma_{1.0}^5$ ,  $\gamma_{0.5}^{40}$  and  $\gamma_{1.0}^{40}$ .

$\varphi_l = 0.5$ , the soot volume fraction drops, and then it grows again for  $\varphi_l = 1$ , substantially surpassing the magnitudes of the gas flame. Conversely, for n-butanol  $f_v$  monotonically raises as  $\varphi_l$  is increased. While the maximum value of  $f_v$  in flame  $B_{0.5}^{40}$  is higher than in  $B_{1.0}^{40}$ , it will be shown later that the total soot formation rate is higher in the second case. Consistently with results found for conditions of low strain rate, the C<sub>2</sub>H<sub>2</sub> mass fraction and the soot volume fraction are always higher for n-butanol flames than for ethanol flames.

We proceed now to analyze the total rates of heat release  $\dot{\omega}_{T,\text{tot}}$ , fuel consumption  $\dot{\omega}_{F,\text{tot}}$ , and soot emission  $\dot{\omega}_{Y_s,\text{tot}}$ , which are shown in Fig. 10. In Fig. 10a, the total heat release rates show a qualitatively similar behavior to the one observed for flames with low strain rate. For  $R_0 = 5 \mu\text{m}$ , the total heat release continuously decreases when  $\varphi_l$  is raised. In contrast, increasing  $\varphi_l$  for  $R_0 = 25$  and  $40 \mu\text{m}$  leads to a monotonic growth of the total heat release rate, with a higher slope for the latter. These general observations are attained for flames of both fuels. Similarly to the low strain rate cases, the ethanol flame structures close to extinction consistently present higher total heat release rates than the n-butanol flames, which can be attributed to the higher consumption rate of ethanol (see Fig. 10b). In general, both the total heat release and fuel consumption rate present higher values for flames close to extinction than for flames at low strain rate.

In flames close to extinction, the total soot emission rate (Fig. 10c) presents considerably smaller values than for cases with a low strain rate (Fig. 5c), which can be attributed to the shorter residence times associated with the increased strain rates. In n-butanol flames, for  $R_0 = 5 \mu\text{m}$ , raising

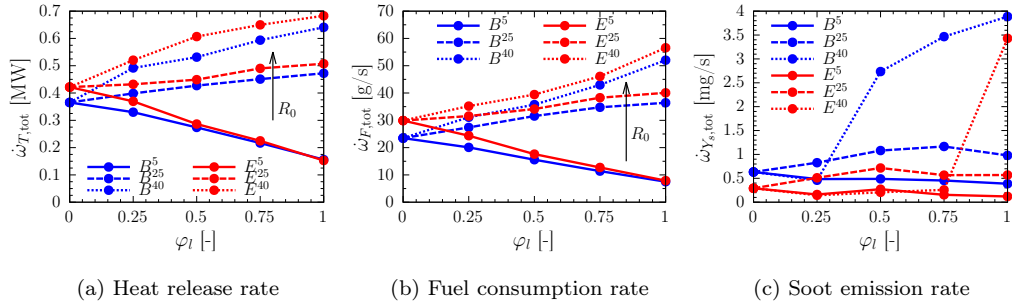


Figure 10: Total rates (close to extinction) of (a) heat release, (b) fuel consumption and (c) soot emission in terms of  $\varphi_l$  and  $R_0$ .

$\varphi_l$  results in a slight, but monotonic decrease of  $\dot{\omega}_{Y_s, \text{tot}}$ . For ethanol, however,  $\dot{\omega}_{Y_s, \text{tot}}$  decreases when  $\varphi_l$  is increased from 0 to 0.25, then it increases for  $\varphi_l = 0.5$  and finally, it drops again for  $\varphi_l = 0.75$  and 1. The increase observed for  $\varphi_l = 0.5$  could be a consequence of the temperature profile (Fig. 7a), which is wider for this flame than for  $E_0$  due to the high droplet penetration. This leads to an enhancement of soot source rate in the regions without evaporation. For  $R_0 = 25 \text{ } \mu\text{m}$ , as  $\varphi_l$  is increased, n-butanol flames show a continuous growth from the reference gas flame to  $\varphi_l = 0.75$ , and then it decreases until  $\varphi_l = 1$ . In ethanol flames, for the same droplet size,  $\dot{\omega}_{Y_s, \text{tot}}$  starts increasing from  $\varphi_l = 0$  to  $\varphi_l = 0.5$ , and then it drops until  $\varphi_l = 1$ . Finally, for  $R_0 = 40 \text{ } \mu\text{m}$ ,  $\dot{\omega}_{Y_s, \text{tot}}$  starts decreasing for both fuels from  $\varphi_l = 0$  to  $\varphi_l = 0.25$ , which is attributable to the higher extinction strain rate associated with those flames. When  $\varphi_l$  is further increased, two different behaviors can be observed again depending on the fuel. For n-butanol, the total soot emission rate grows monotonically and substantially until  $\varphi_l = 1$  as a consequence of the temperature increase. For ethanol, on the other hand,  $\dot{\omega}_{Y_s, \text{tot}}$  slightly grows from  $\varphi_l = 0.25$  to 0.75, and then it increases considerably for  $\varphi_l = 1$ , again, due to the high temperature generated in this case. Consistently with the soot volume fraction profiles, for all the conditions studied, n-butanol flames have higher soot emission rates than ethanol flames.

In summary, for the flames close to extinction analyzed in this subsection, only few combinations of  $\varphi_l$  and  $R_0$  simultaneously increasing the heat release rate and reducing the total soot emissions were identified. In particular, for  $R_0 = 40 \text{ } \mu\text{m}$  such situations were observed in ethanol flames within the interval  $0.25 \leq \varphi_l \leq 0.75$ , and for a single n-butanol flame with a specific

value of  $\varphi_l = 0.25$ . Also, and even when our double optimization objective was not achieved, it is remarkable that for  $R_0 = 25 \text{ }\mu\text{m}$  heat release could be significantly improved varying  $\varphi_l$ , without greatly increasing the soot emission rates. Indeed, this positive effect was reached for all values of the liquid to total fuel mass ratio considered. It is also highlighted that all flames close to extinction show higher total heat release rate values and lower total soot emission rates than for the corresponding low strain rate situations. Finally, and consistently with the results obtained with low strain rates, ethanol flames are found to consistently release more heat and less soot than n-butanol flames.

## 5. Conclusions

Numerical simulations of several ethanol-N<sub>2</sub>/air and n-butanol-N<sub>2</sub>/air counterflow flames have been performed considering different initial droplet radii and liquid to total fuel mass ratios. Both low and high strain rate (close to extinction) conditions have been studied. Based on the results, conditions have been identified for which an increment of the total heat release rate and a reduction of the soot formation rate could be simultaneously achieved.

For low strain rates, spray flames ( $\varphi_l = 1$ ) with initial droplet radii of 25 and 40  $\mu\text{m}$  were found to satisfy the aimed double objective. For situations close to extinction, on the other hand, only few combinations of  $\varphi_l$  and  $R_0$  could do it. Particularly, for  $R_0 = 40 \text{ }\mu\text{m}$  only ethanol flames in the interval  $0.25 \leq \varphi_l \leq 0.75$ , and a single n-butanol flame with a specific value of  $\varphi_l = 0.25$  could achieve this.

Despite failing in the attainment of the soot reduction goal, it is worth

noting here that, for the high strain rate situations, increasing the amount of fuel injected in liquid phase for  $R_0 = 25 \mu\text{m}$  allowed a significant improvement of the total heat release without greatly increasing soot emission rates. In fact, this favorable behavior was observed for all cases studied with that initial droplet radius and  $\varphi > 0$ .

Regarding the comparison between the performance of both fuels, it has been found that ethanol flames release more heat and less soot than n-butanol flames. This result is not consistent with the fact that the heating value and the inlet mass flow considered for n-butanol are higher than for ethanol. However, this can be explained by the fuel consumption rate, which is consistently lower for n-butanol.

These results show how the phase in which fuel is injected plays an important role in optimizing combustion processes and simultaneously accomplishing higher energy release and lower soot emissions. Moreover, this work represents a starting point for the development of more detailed analyses in the future, where special emphasis should be given to the study of soot particle size distributions by means of more detailed soot models.

### **Declaration of competing interests**

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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