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Diffusion Ignition Processes in MILD Combustion: A Mini-Review

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MILD combustion processes belong to new combustion technologies developed to achieve efficient and clean fuel conversion. The basic concept behind its implementation is the use of high levels of hot exhausted gas recirculation within the combustion chamber. They simultaneously dilute fresh reactants, to control system temperatures and pollutants emission, while promoting fuel complete oxidation. The combination of low maximum system working temperatures and high diluted mixtures with intense pre-heating delineates an oxidation process with unique chemical and physical features, such as uniformity of scalars at macroscale related to distributed reacting regions at microscale, extremely different from conventional flames. In turn, this requires the definition and characterization of new elementary processes, not ascribable to traditional deflagration or diffusive flame structures, which, in literature have been identified as "diffusion ignition." The present mini-review reports on several literature characterizations of such reactive structures under steady and unsteady conditions combining evidences from numerical, experimental, and/or theoretical studies. Both premixed and non-premixed configurations were analyzed in terms of system temperature, heat release, and species distributions as key parameters to describe the intrinsic nature of such new elementary processes. Analyses were realized changing the main system external parameters (mixture pre-heating temperature, dilution level in several feeding configurations) moving from traditional to MILD conditions. Results highlighted the "distributed ignition" nature of igni-diffusive structures, with implication on the thickness of the oxidation structures in the mixture fraction space, the presence/absence of a pyrolysis region, and the correlation of the maximum heat release with the mixture stoichiometric.

Keywords: MILD combustion, distributed ignition, igni-diffusion, dilution level, heat release

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INTRODUCTION

Elementary structures in combustion such as laminar diffusion layers have been characterized in the literature (Tsuji, 1982; Chao et al., 1991; Darabiha, 1992; Chen et al., 2012). Dilution and preheating of reactants pointed out some peculiarities that were examined through numerical, experimental, and theoretical studies in the last decades (de Joannon et al., 2009; Abtahizadeh et al., 2012; Sepman A. et al., 2013; Sorrentino et al., 2013). Diffusive ignition process under MILD combustion was deeply investigated (Cavaliere and de Joannon, 2004).

The relevance of such processes concerns several fields of application such as low heating value fuel oxidation (Maruta et al., 2007), destruction of VOC, or combustion in flows with internal recirculation.

Diffusion ignition can be described in 1-D spatial conditions, but usually they are very difficult to mimic in simple experiments due to such low dimensionality. 2-D experiments where fuel jets are injected in co-flowing or cross-flowing oxidizer streams are easier to realize for diluted and preheated conditions.

Jet in hot flows configuration was used to reproduce diffusion ignition processes (Adelaide or Delft jet in hot coflow) in both the laminar (LJHC) and turbulent (JHC) cases when inlet reactant temperatures are higher than ignition ones (Medwell et al., 2007, 2009; Choi et al., 2009; Oldenhof et al., 2011; Sepman A. V. et al., 2013). In particular, methane and CH_4/H_2 blends were analyzed by Medwell et al. (2007) and Al-Noman et al. (2016), whereas Oldenhof et al. (2010) analyzed the stabilization region of natural gas mixtures burning in a hot/diluted coflow by recording the flame luminescence with an intensified high-speed camera. The spatial distributions of the hydroxyl radical (OH), formaldehyde (H_2CO), and temperature imaged by laser diagnostic techniques were obtained for ethylene flames by Medwell et al. (2008) whereas Choi and Chung (2010) investigated the autoignition characteristics of laminar lifted ethylene flames in coflow air with elevated temperature over 800 K. Arndt et al. (2019) studied the autoignition of propane jet-in-hot coflow with high-speed OH^* chemiluminescence imaging and high-speed Rayleigh scattering for mixture fraction, temperature, and scalar dissipation rate measurements. Liquid fuels, such as ethanol (Correia Rodrigues et al., 2015; Ye et al., 2016), have been also studied in some preliminary works.

The reactive structure features that were recognized in these studies can be summarized in such points:

1. Combustion Mode Type:
   - Diffusion–ignition
   - Standard deflagration or diffusion flame
   - Mixed-mode regimes (partially premixed).
2. Diffusion Ignition is Strongly Sensitive to Boundary Conditions More Than Standard 1-D Deflagrative/Diffusion Flame Processes.

The same type of analysis has been also performed in Jet in Hot Cross Flow configuration (Sidey and Mastorakos, 2015; Wagner et al., 2017) where fuel was injected into hot and vitiated air crossflow. Results suggested that autoignition is the dominant stabilization mechanism.

A wide numerical/theoretical characterization of igni-diffusive steady structures under MILD conditions was reported in the literature with 1-D counterflow configurations. Reactive structures were analyzed as a function of feeding configurations from several groups (de Joannon et al., 2012a,b; Zou et al., 2014; He et al., 2016).

In the following sections, the peculiarity of diffusion ignition processes will be analyzed for steady or unsteady conditions.

STEADY DIFFUSION IGNITION: HDDI AND HCDI

Distributed combustion regime occurs in technologies where dilution and preheating of reactants are used for efficient and clean energy conversion (Li et al., 2011; Khalil and Gupta, 2017; Perpignan et al., 2018). In practical systems, gas recirculation is adopted (Sorrentino et al., 2018a; Sabia et al., 2019). Low combustion temperatures and high dilution/pre-heating alter the fuel conversion with respect to traditional flames (Maruta et al., 2000; Minamoto et al., 2014; Minamoto and Swaminathan, 2015).

Elementary processes that undergo MILD combustion influence the controlling parameters (Cavaliere et al., 2016). In particular, the reactivity under stoichiometric conditions is comparable with the one related to fuel-lean or fuel-rich conditions (de Joannon et al., 2010). It follows that reactions are homogeneously distributed (Plessing et al., 1998; Özdemir and Peters, 2001).

In particular, the characterization of diffusion-controlled combustion processes developing in a steady 1-D layer was reported in the literature with several numerical studies by means of the opposed jets configuration (de Joannon et al., 2007; Cheong et al., 2017; Mameri et al., 2018).

This section takes into account numerical characterization of steady mixing layers with diluted and/or preheating streams with different feeding configurations as a function of main parameters.

In this review article, a sub-classification of steady diffusion ignition process will be used as reported in the following:

1. Reactants with homogenous mixture composition and preheating of inert flow as main controlling parameters [homogeneous charge diffusion ignition (HCDI)]
2. Non-premixed flows with reactants dilution and preheating as main controlling parameters [hot diluted diffusion ignition (HDDI)].

In the HCDI mode (de Joannon et al., 2007; Mastorakos, 2009; Goh et al., 2013), a homogeneous reactant charge interacts with inert (such as nitrogen or carbon dioxide) at high temperature (i.e., combustion products) and this yields a counter-diffusion layer. On the other hand, the HDDI is realizable for non-premixed streams with a certain level of dilution and/or preheating (de Joannon et al., 2009; Chen and Zheng, 2011; Ye et al., 2017). The initial conditions of HCDI processes are fixed on one side of spatial coordinate homogeneous reactants in a fixed composition and the species on the other side are inert (for instance combustion products) at high temperature, as reported in the sketch of the insert in **Figure 1A**. As is well-known, a premixed fuel/air mixture outside the flammability limit cannot support a deflagration process. Several experimental studies have reported the occurrence of this process when the heat source is a heated wall or a high-temperature inert flow in a laminar (Darabiha et al., 1988; Smooke et al., 1991; Zheng et al., 2002) or turbulent (Blouch et al., 1998) counterflow. In particular, Mastorakos et al. (1995) investigated the effects of simultaneous dilution and preheat of reactants by mixing with hot combustion products in turbulent counterflow flames formed near the impinging region of two opposed jets. Examples of this reactive structure are also obtained in a counterflow configuration as shown in the sketch of **Figure 1A** for a methane/oxygen stream that impinges toward high-temperature flows. The bulk flow velocities define the system strain rate. The sketch in **Figure 1A** shows the location of the reactive region (orange area) when the mixture is outside the flammability limits

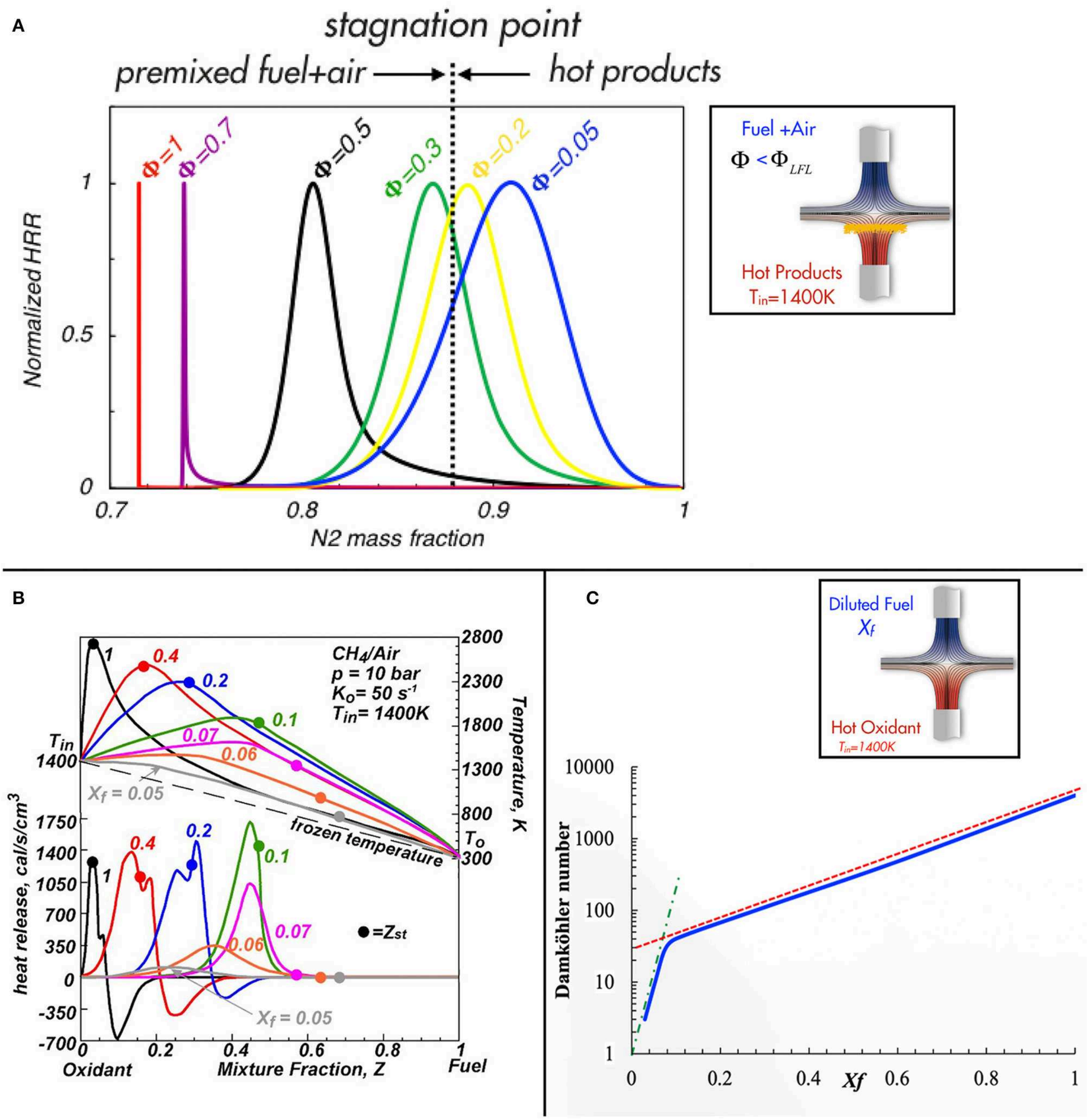

FIGURE 1 | Normalized heat release rate as a function of N_2 mass fraction in HCDI configuration **(A)**. **(B)** Reports temperature and heat release rate in a HDDI (HODF) configuration [adapted from de Joannon et al. (2009) with permission from Elsevier] and Damköhler number in **(C)**.

($\Phi < \Phi_{\mathrm{LFL}}$). Such counterflow allows for the characterization of several HCDI or deflagration structures (de Joannon et al., 2007). In particular, the results shown in **Figure 1A** (Cavaliere et al., 2016) reports a homogeneous fuel/air charge continuously heated by a hot inert flow of N_2 (mimicking combustion products). A premixed CH_4-air flow, characterized by an inlet temperature (T_0) and velocity (V_0), is fed toward an opposed flow of nitrogen at high temperatures (T_{in}). The distance D between the jets is 2 cm. Numerical analysis was carried out by means of Oppdif application for counterflow configurations of ChemKin 3.7 package by using the GRI 3.0 mechanism as kinetics model. The enthalpy production reported in **Figure 1A** with a red curve at $T_{\mathrm{in}} = 1{,}400\,\mathrm{K}$ for $k = 50\ \mathrm{s}^{-1}$ is positioned on the premixed fuel/air side for $\Phi = 1$ with a very thin reactive region. By decreasing the inlet equivalence ratio, these heat release curves are different from those at $\Phi = 1$ as they are broadened, and are shifted closer to the stagnation point. Notably, the reactive zone location passes the stagnation point for $\Phi < 0.2$ when the mixture is outside the flammability limits. In this case, the oxidative structures are located on the hot products side.

In summary, the figure demonstrates that a great variety of oxidation structures may be stabilized in HCDI processes and the features of the oxidation process are related to the diffusion between the hot products and the air/fuel charge. Such behavior can be ascribed to MILD combustion and it has features that partially explain the distributed and noiseless characteristic reported for some flameless applications (Wünning and Wünning, 1997; Khidr et al., 2017; Zhou et al., 2017). In fact, the reactive process is distributed on a wide mixture fraction region and the transition from HCDI to deflagration is gradual (Sidey et al., 2014).

Combustion processes in which the reactants are separated yield diffusion flames and they can be obtained in a counter-diffusion reactor when it is fed with oxidant and fuel in opposite directions along the same axis. In MILD combustion, such streams can be diluted with inert species (such as combustion products) and can be heated to such high temperatures that the frozen temperature is higher than the ignition one (de Joannon et al., 2010; Sidey and Mastorakos, 2016).

In this case, the non-premixed configuration, as reported in the sketch of **Figure 1C**, is characterized by fuel at ambient temperature and molar fraction X_f that is fed vs. an opposed airflow at a fixed preheating temperature (T_{in}). The flow rates are kept constant to fix the bulk strain rates $K_0 = V_0/D$. In **Figure 1B**, the structure of the reactive zone was depicted for HODF where temperature and heat release rate profiles as a function of the mixture fraction (Z) were reported for several X_f values. The plots of **Figure 1B** (adapted by de Joannon et al., 2009) were obtained at $P = 10$ bar for a CH_4/Air system. They refer to diluted methane at ambient temperature diffusing in hot air at $T_{in} = 1{,}400$ K, i.e., a HODF (Hot Oxidant Diluted Fuel) feeding in a SICF (Steady Igniting Counter-Flow). Numerical tools and kinetics mechanisms are the same used for HCDI cases reported before. Methane content was varied from $X_f = 1$ to 0.05 by means of N_2 dilution. The stoichiometric mixture fraction location is indicated on the profiles with dots. The heat release profile (lower part of **Figure 1B**) reported with the black curve is related to an undiluted case and it exhibits a maximum in correspondence of Z_{st} whereas pyrolytic regions with negative heat release occur for fuel-rich region. A similar behavior was obtained for T (upper part) that reaches a maximum value of about 2,800 K. The fuel dilution strongly alters the oxidative structure as a function of its magnitude and it emphasizes the difference with respect to conventional conditions. For $X_f = 0.5$, the T_{max} values are shifted toward higher Z and the oxidation region in the heat release profile shifts and widens. The pyrolytic region is enlarged toward higher Z with strong reduction of the minimum heat release rate. This behavior becomes more clear by decreasing X_f, causing the vanishing of the pyrolytic region for dilution level higher than 85%. In fact, when the dilution level reaches 90% ($X_f = 0.1$), heat release profiles exhibit only a single maximum that diminishes its intensity with X_f. Moreover, the most important effect of the dilution is a shift of the oxidative region backward toward lower Z, in opposite direction with the stoichiometry (as reported by the colored dots that indicate the Z_{st} location).

Therefore, by summarizing, several peculiar characteristics have been pointed out as discriminative for the occurrence of different combustion regimes. They have been related to reactive region thickness, absence of a pyrolysis region, and the correlation of the maximum heat release with the Z_{st}. Following such criteria, the features of the reaction zone suggested a different name for the elementary process with respect to standard diffusion ones, and thus, HDDI was chosen for such non-premixed case.

The results above reported revealed that both the HDDI and HCDI modes support a "distributed ignition" behavior. Moreover, a strong extinction resilience was also reported in several literature papers as another important feature of such MILD processes (Sidey et al., 2016; Evans et al., 2017; Sidey and Mastorakos, 2018). All these features are linked to each other.

Another important synthetic representation of MILD features is reported with the blue curve in **Figure 1C** where Damköhler number is reported vs. the fuel mole fraction. Its evaluation is based on the ratio between a characteristic convective time (based on the strain rate) and a kinetic one. More specifically, it diminishes from about 7,000 to 50 by decreasing the X_f value from 1 to 0.1. Also, in this case, the most relevant variation is shown at 90% fuel dilution where Da undergoes a significant change. This is correlated to a "gradual" behavior of the reactive process that entails a continuous increase of chemical kinetics times with respect to a reference fluid-dynamic one. It is of interest that the Damköhler number change is three orders of magnitude with respect to undiluted cases. In particular, two asymptotic behaviors can be identified with the dashed red line for $0.15 < X_f < 1$ and with the dashed-dotted green one for $X_f < 0.1$. Such paradigm shift in the reactive structures at a fixed dilution level was also identified in several literature papers for diluted conditions in SICF (Fotache et al., 1997, 1998; Abtahizadeh et al., 2012) and confirmed also for low-heating value fuels (Kwiatkowski and Mastorakos, 2016).

UNSTEADY DIFFUSION IGNITION

Igni-diffusive structure characterizations are possible in 1-D configurations by following the temporal evolution of diffusion layer ignition/oxidation for diluted and preheated conditions in wide parameter ranges. In this case, unsteady elementary processes strongly depend on the boundary conditions and fluid-dynamics pattern. In particular, literature evidenced two main possibilities for transient diffusion-controlled combustion processes in 1-D configurations: IML (Igniting Mixing Layer) and ICF (Igniting Counter-Flow).

IML can be defined as a "universal" or "zero-order" configuration because it represents the natural evolution of diffusive-reactive process (Abtahizadeh et al., 2015). Therefore, it is of interest as reference case because the temporal evolution refers to an initial unmixed mixture where the initial mixture fraction gradient is not prescribed by a fixed strain rate

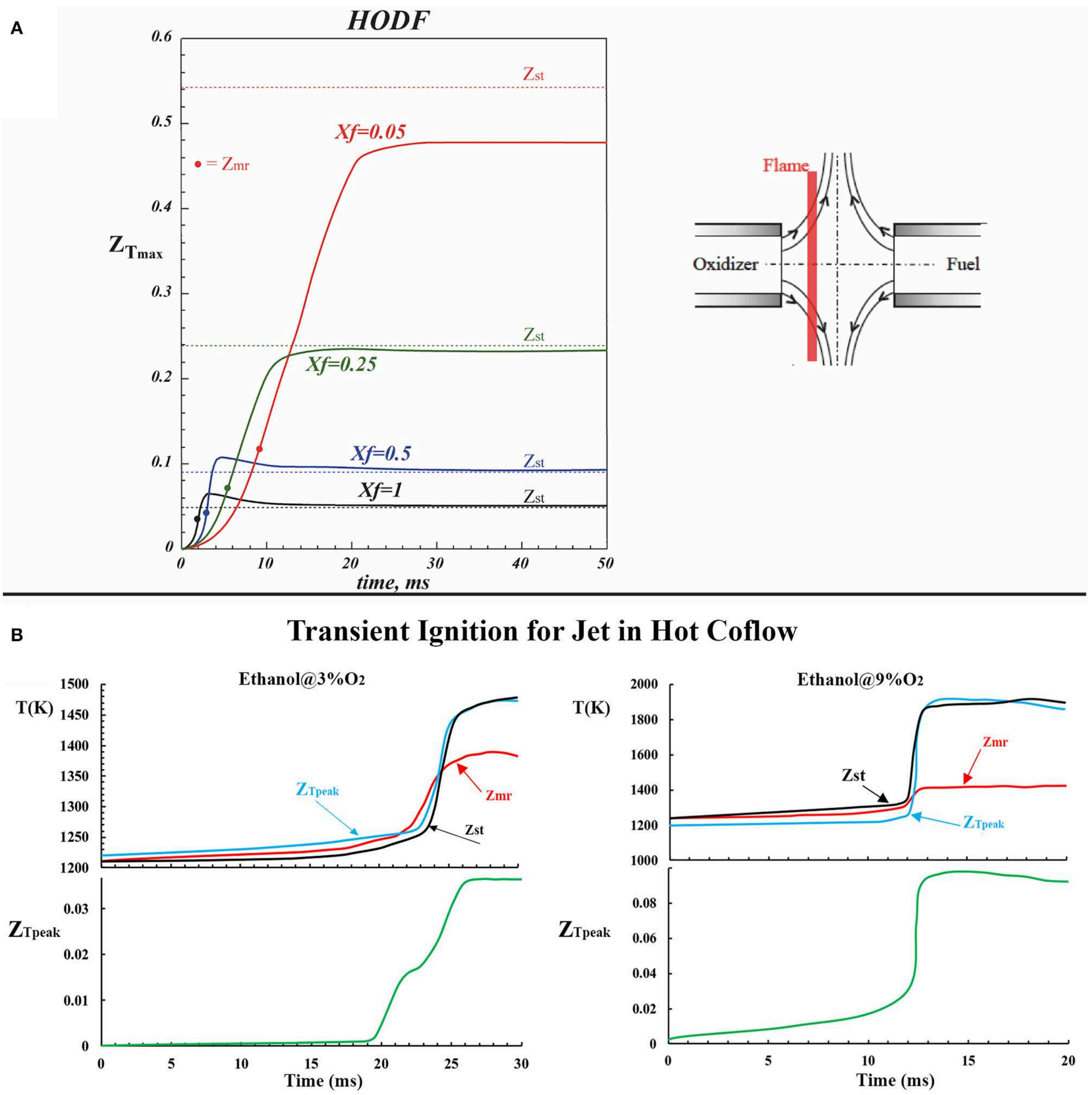

FIGURE 2 | Temporal evolution of mixture fraction where the peak temperature occurs for HODF conditions **(A)** adapted from Sorrentino et al. (2013) with permission from Elsevier. **(B)** Reports transient numerical computations for ethanol in JHC flames with the boundary conditions obtained from the experiments by Ye et al. (2018). Temperature and Z_{Tpeak} as a function of time were reported for two oxygen levels.

but is controlled by molecular diffusion. It is a key process also for inferring tabulated chemistry information for complex systems (Sorrentino et al., 2018b). IML was reported in the literature to evaluate ignition characteristics (most reactive mixture fraction) of diluted fuels in comparison with Auto-Ignition and Homogeneous Ignition (Mastorakos et al., 1997; Im et al., 1998; Sreedhara and Lakshmisha, 2000; Hilbert and Thévenin, 2002; van Oijen, 2013).

ICF in steady strain flow field is the configuration in which the transient diffusion ignition process evolves in a developed mixing layer originated by a mixture fraction in two opposed laminar jets, where the initial mixture fraction is distributed according to frozen mixing due to applied strain. This kind of process was one of the most characterized in the literature because it can be obtained through coflow configurations for both the undiluted and diluted cases, as already underlined in the *Introduction* section (Cabra et al., 2002; Dally et al., 2002; Chung, 2007). In this case, several regimes such as autoignition, flame propagation, and diffusion ignition can occur (Medwell et al., 2016; Schulz et al., 2017).

Unsteady 1-D reactive diffusive layer evolution has been depicted for HODF conditions in **Figure 2A** by reporting the numerical results obtained in ICF configuration and adapted by the results of Sorrentino et al. (2013).

The profiles in **Figure 2A** show the temporal evolution of the maximum temperature location in the mixture fraction space. The solid dots depict the location of the most reactive mixture fraction (Z_{mr}) whereas the dashed lines are related to the position of Z_{st}. It is worthwhile to note that, for undiluted or slightly diluted conditions ($0.2 < X_{\mathrm{f}} < 1$), the location of Z_{mr} and Z_{st} is very close to each other. It means that ignition and stabilization phenomena occur in the same mixture fraction and time intervals (Im et al., 2000). Therefore, in such cases, the diffusive structure growth in time is limited to a narrow mixture fraction space. For $X_{\mathrm{f}} < 0.1$, the main effect of MILD combustion (because the reactant dilution slows down the oxidative chemical kinetics) is to increase the distance between the "most reactive" mixture fraction and the "stoichiometric" one. In each case, the most reactive mixture fraction is always placed on the high-temperature side (preheated oxidizer in **Figure 2A**) and the location is not affected by dilution (Viggiano and Magi, 2004; Yoo et al., 2011).

The aforementioned behaviors were confirmed for ethanol-diluted flames in JHC configuration, as reported in the numerical results of **Figure 2B**, where boundary conditions were obtained from Ye et al. (2018). Unsteady flamelet equations were solved in time and mixture fraction space using the FlameMaster program (Pitsch, 1998). It shows the peak temperature as a function of time for ethanol at 3 and 9% of O_2. The mixture fraction where the peak temperature occurs is denoted as Z_{Tpeak}. As the oxygen level reduces from 9 to 3%, the ignition process becomes prolonged with a reduced temperature increase. Thus, flames in the 3% O_2 case are close to diffusion ignition cases, being distributed with a low temperature increase.

Recent DNS studies (Doan et al., 2018) confirmed the importance of dilution and Z_{mr} location on the stabilization but they also depicted that non-premixed turbulent MILD combustion is a complex process with spatial and temporal ignition fronts, non-premixed, and propagating flames that are strongly convoluted and interacting. In particular, multiple igniting mixing layers can interact between themselves and the annihilation of peripheral isolines may strongly influence the reactive structures (Sorrentino et al., 2017).

CONCLUSIVE REMARKS

The analyses reported so far have emphasized that the occurrence of MILD combustion in diffusion-controlled conditions strongly differs from deflagration and diffusion flames. These characteristics are of interest in comparison to the conventional flame structure where the mixture fraction range in which the reaction takes place is quite narrow and changes slightly with dissipation rate. In contrast, the oxidation structure of MILD combustion shifts along the mixture fraction space according to different boundary conditions (temperature and dilution). The features of diffusion ignition are related to several peculiarities such as distributed ignition, flameless oxidation, and heat release not correlated with stoichiometric mixture fraction, differently from flame structured processes.

Strong extinction resilience and gradual ignition are also distinctive characteristics of such reactive structures that were highlighted by analyzing the unsteady evolution of the diffusive reactive layer under preheated and diluted conditions.

AUTHOR CONTRIBUTIONS

GS and PS were involved in the bibliographic research to include in the manuscript the most important literature contributions on the diffusion ignition process in MILD combustion. They have also strongly contributed in writing several parts of the article. AC and MJ gave their support in the theoretical background regarding categorization and classification of MILD combustion processes. RR gave important insights and hints in the physical description of diffusion ignition and its connection with dimensionless numbers. He also strongly revised the English language in the manuscript.

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