Numerical Study on the Stabilization of a Self-Sustaining Steady-State Premixed Cool Flame

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Abstract

The unexpected finding of the quasi-steady state cool flame onboard the International Space Station motivated increasing interests to study the dynamic behaviors of cool flames. One key scientific question is how to form and stabilize a self-sustaining steady-state cool flame in a burner with well-defined boundary conditions. This paper numerically studied the stabilization of a self-sustaining steady-state premixed dimethyl ether/O₂/N₂ cool flame. The dual S-curve response in the perfectly-stirred reactor was first analyzed and the results indicated three ways to form a self-sustaining premixed cool flame: 1) igniting the unburned fresh mixture by decreasing the residence time, 2) igniting the unburned mixture by increasing the temperature of the unburned fresh mixture, and 3) extinguishing an extremely lean or an extremely rich hot flame by decreasing the residence time. Using the counterflow configuration, the proposed three ways were successfully demonstrated by tuning the flow temperature and stretch rate. Moreover, double structured flames, i.e., a leading premixed cool flame front followed by a premixed warm flame, were numerically reported in the counterflow flame configuration. The thermal and species structures of the double flames were also discussed.

Keywords Cool flame · Ignition · Extinction · Counterflow · Double-structured flames

Nomenclature

Latin

$\dot{H}_{net}$	Net heat production rate, J/cm ³ ·s
L	Separation distance of the two stream exits in the counterflow flame, cm
p	Pressure, atm
T	Temperature, K
U	Velocity, cm/s
x	Mole fraction
z	Flame coordinate, cm

Greek

ϕ	Equivalence ratio
ρ	Gas density, g/cm ³
τ	Residence time, s

ω Elementary reaction rate, mole/m³·K

Subscript

i	Value of species i
m	Mixture
max	Maximum value
r	Reactor
u	Unburned

Abbreviation

CFE	Cool flame extinction
CFI	Cool flame ignition
HFE	Hot flame extinction
HFI	Hot flame ignition

Introduction

Near-limit flame dynamics has long been concerned as it is related to fire safety issues and initiating/terminating of combustion processes. In a near-limit flame, the chemistry slows down to a level that the characteristic chemical reaction time scale is at the same order of magnitude as, or even longer than the time scale of the gravity induced buoyant convection (Ronney 1998). Hence, gravity significantly affects the heat

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and mass transfer in the near-limit combustion process, making the near-limit flame behavior quite different at earth gravity (1 g) from the one at microgravity (μg) conditions. A number of microgravity near-limit flame experiments, e.g., (Ronney and Wachman 1985; Law and Faeth 1994; Maruta et al. 1996; Zhang and Egolfopoulos 2000; Zhang et al. 2011; Liu et al. 2015), have been conducted since the 1950s. The motivations of these efforts are: 1) to assess the fire hazards in spacecraft or onboard space station, and 2) to study the flame dynamic phenomena which are unrevealed at the earth gravity.

Recently, a series of exciting distinct phenomena (Nayagam et al. 2012; Farouk et al. 2015) have been discovered in the single alkane droplet combustion studies onboard the International Space Station (ISS). These phenomena have proved the existence of a quasi-steady diffusion “cool flame” in the low-temperature regime (Farouk and Dryer 2014). This cool flame found on ISS is different from the classical cool flame in the earlier literature, e.g., (Naidja 2003). The former one is a quasi-steady diffusion flame that sustains from seconds to $\sim$ half minutes (Nayagam et al. 2012), while the latter one refers to a transient propagating wave towards a premixed fuel/air fresh mixture leading to a high-temperature flame of the remaining reactants, e.g., (Griffiths and Inomata 1992). The discovery of the quasi-steady cool flame has excited the enthusiasm of the academia, as it is not only of richness in fundamental near-limit flame dynamics but also potentially provides an approach to study the low-temperature chemistry. Numerical simulations (Ju et al. 2011; Krisman et al. 2017) have also presented the existence of a cool flame front under engine-like conditions, and this cool flame significantly affects the high-temperature chemistry ignition (Krisman et al. 2017). Once a self-sustaining cool flame is established for a sufficiently long period of time, one can apply the existing detection techniques to seek insight into the low-temperature chemistry as well as related molecular transport. Thus, one of the key questions is how to form and stabilize a cool flame in a well-defined flame configuration.

For nonpremixed flames, cool flame behaviors were studied using the single fuel droplet configuration (Nayagam et al. 2012; Farouk and Dryer 2014; Farouk et al. 2015; Takahashi et al. 2018; Farouk et al. 2019) and the counterflow configuration. Several new phenomena were observed, such as multistage oscillation (Farouk et al. 2015), envelope-shaped cool flame (Takahashi et al. 2018) and a unique warm flame (Farouk et al. 2019) in the single fuel droplet combustion. The S-curve response of a cool flame was also numerically analyzed (Law and Zhao 2012; Zhao and Law 2013; Sun et al. 2014; Won et al. 2015; Lin et al. 2019) and the counterflow nonpremixed cool flames were subsequently stabilized (Deng et al. 2014; Sun et al. 2014; Won et al. 2015; Deng et al. 2017; Reuter et al. 2017a). In a more recent study, Novoselov et al. (2019) performed direct numerical simulations to

study isolated turbulent nonpremixed cool flame for the first time and assessed the applicability of flamelet models.

In nonpremixed cool flames, the counter-diffusion of the fuel and oxidizer controls the mixing and thereby the overall reaction rate, and the NTC chemistry plays a secondary role in the global flame behaviors. For a premixed flame, the flame dynamics inherently depends on the coupling of chemical kinetics and molecular transport (Law 2006). Consequently, the question of whether a premixed cool flame can be formed and stabilized was naturally raised. Quite a few attempts have been made to answer this question. As listed in Table 1, the premixed cool flames were studied using the configurations of 1D freely propagating flame, counterflow flame, Hecken flat flame, spherical flame, and wall stabilized stretched flame. Ju and coworkers conducted numerical study on 1D freely-propagating flames of DME/O₂ mixtures at ambient pressure with O₃ sensitization (Ju et al. 2015) and at elevated pressure without O₃ sensitization (Ju 2017), and revealed three flame regimes, 1) hot flames, 2) double (hot and cool) structured flames, and 3) cool flames. Subsequently, with O₃ sensitization, premixed cool flames were stabilized using counterflow flame (Reuter et al. 2016, 2017b) and Hencken flat flame (Hajilou et al. 2017; Hajilou and Belmont 2018). Meanwhile, Zhao et al.’s (2016) computation demonstrated that there is a stretch rate window within which a premixed DME/O₂/N₂ can be thermally ignited in a counterflow configuration without O₃ sensitization. According to these numerical results, Zhao et al. managed to experimentally establish self-sustained premixed cool flames with the equivalence ratio range of 0.4–1.8. Zhang et al. (2018) numerically simulated the outwardly-propagating spherical DME/air premixed flames and also found a double flame structure, i.e., a leading cool flame front propagating to the fresh reactants followed by a hot flame. This transient cool flame can last up to 0.5 s and then is caught up by the hot flame, and finally the two flame merges. Recently, Lee et al. (2019) also experimentally stabilized an O₃-free premixed DME/O₂ cool flame through impinging the reactant flow to a 700 K hot wall.

Based on the findings in the literature, the consensus has been reached that the self-sustaining premixed cool flames can be established by carefully controlling the preheating temperature and residence time. Some fundamental dynamic features of premixed cool flame are also obtained. The laminar flame speed of cool flames is normally <10 cm/s (Zhao et al. 2016; Ju 2017; Zhang et al. 2018). The cool flame can sustain outside a mixture’s “flammable range” that is conventionally determined by the high-temperature chemistry, i.e., the existence of cool flame extend the conventionally-defined flammability limits (Reuter et al. 2016; Ju 2017; Liang and Law 2017). Moreover, unlike the hot flame, the flame speed and extinction stretch rate of a premixed cool flame have a relatively weak dependence on equivalence ratio (Law and Zhao 2012; Zhao and Law 2013).

Table 1 Recent self-sustaining premixed cool flame studies in the literature

Literature	Fuel	Oxidizer	Assist.	Exp. or Num. ¹	Flame type	Equivalence ratio	Pressure
(Ju et al. 2015)	DME	O ₂	O ₃	Num	1D-FPF ² , CFF ³	0.02–0.3	1 atm
(Zhao et al. 2016)	DME	O ₂ with varied N ₂	–	Exp & Num	1D-FPF, CFF	0.4–2.0	1 atm
(Reuter et al. 2016)	DME	O ₂	O ₃	Exp & Num	CFF	0.06–0.24	1 atm
(Ju 2017)	DME	O ₂ with varied N ₂	–	Num	1D-FPF	0.1–10	1–20 atm
(Reuter et al. 2017b)	DME	O ₂	O ₃	Exp & Num	CFF	0.09–0.11	1 atm
(Liang and Law 2017)	<i>n</i> -C ₇ H ₁₆	Air	–	Num	1D-FPF	~0.3 - ~75 ⁴	1–5 atm
(Hajilou et al. 2017)	DME	O ₂	O ₃	Exp & Num	Hencken ⁵	0.4–1.4	7.3 kPa
(Hajilou and Belmont 2018)	C ₃ H ₈	O ₂	O ₃	Exp & Num	Hencken	0.17–1.0	17.3 kPa
(Zhang et al. 2018)	DME	O ₂ with varied N ₂	–	Num	SF ⁶	1, 2	1 atm
(Lee et al. 2019)	DME	O ₂	–	Exp & Num	WSSF ⁷ , CFF	0.2, 0.5	1 atm

1. Exp. – Experimental/Num. – Numerical; 2. 1D-FPF – 1D freely propagating flame; 3. CFF – counterflow flame; 4. In this literature the normalized equivalence ratio was used; 5. Hencken burner is usually operated in the nonpremixed mode but in this literature the fuel and oxidizer were premixed at the molecular level before burning; 6. SF – spherical flame; 7. WSSF – wall stabilized stretched flame

The existing studies partially uncovered the big picture of cool flame, but the premixed cool flame dynamics and related NTC chemistry are still not entirely understood. For instances, although the premixed cool flame sustains with the ozone sensitization, e.g., (Reuter et al. 2017b; Hajilou and Belmont 2018), the ozone chemistry brings in new complexity, making it not clean to study the NTC chemistry of fuel oxidation. Similarly, metal surfaces may induce considerable wall chemistry effect on the cool flame (Wan et al. 2019), it is also needed to carefully justify the flame-wall interaction when a cool flame is stabilized by a hot metal wall. How to initiate and then stabilize a premixed cool flame without external promoters is still an interesting question. Moreover, 1D freely-propagating flame simulation presented double structured flames, i.e., a leading cool flame followed by a hot flame on the fuel-lean side, while whether the double-structured flames can be steadily formed in a real flame configuration, such as a counterflow, remains unknown.

The present numerical study consists of three logically-related tasks. In Section 2, the NTC chemistry behavior with respect to the residence time was investigated and the effects of initial temperature (T_u), dilution, pressure (p) and equivalence ratio (ϕ) on the S-curve hysteresis were systematically assessed. The purpose of this task was to find reliable ways to initiate and stabilize a premixed cool flame without external promoters. In Section 3, three ways to establish a self-sustained premixed cool flame were demonstrated using the counterflow configuration. In Section 4, double structured flames, a leading cool flame front followed by a warm one, were presented in the counterflow configuration. The fuel chosen for the present study is DME, one of the smallest molecules exhibiting a strong NTC behavior. The present computation adopted Zhao et al.'s (Zhao et al. 2008) DME oxidation model which has been widely applied to study the cool flame dynamics. Although some recent studies (Deng

et al. 2017; Lee et al. 2019) reported that Zhao et al.'s model may not be perfectly-developed, it still gives correct trends and will not change the conclusion of the present analysis. Thus, Zhao et al.'s model still provides accurate underlying physics and is good enough for the present study.

S-Curve Migration of DME/O₂/N₂ Homogeneous Combustion

In this Section, the system was simplified to homogeneous flow combustion without the molecular diffusion. The steady-state adiabatic perfectly-stirred reactor (PSR) was chosen as the physical model in which the residence time of the reactants was only determined by the bulk flow conventional transport. The PSR Fortran code (Glarborg et al. 1986) integrated with Chemkin II package (Kee et al. 1989) were used to compute the steady-state reaction temperature (T_r) and species composition (x_i) inside the reactor. Near the turning point of the S-curve, the bifurcation structure was computed using the following continuation method. T_r was imposed as an initial condition rather than a target of the solution. Either the residence time (τ), or the temperature (T_u) of the inlet unburned mixture was then released as an unknown to be solved. Thereby, either the T_r - τ S-curve, or the T_r - T_u S-curve formed as T_r continuously decreases or increased. Although the PSR model, free of molecular diffusion, is not exactly the same to a real flame, it captures the keys to the flame dynamics such as the ignition and extinction. Moreover, the two PSR parameters, T_u and τ , are also the keys to stabilizing a cool flame in a real flame. Taking the premixed counterflow flame as an instance, T_u corresponds to the temperature of the hot inert flow on which the premixed reactant flow impinges. τ in a counterflow flame is characterized by the inverse of the stretch rate (κ), i.e., $\tau \sim 1/\kappa$.

Figure 1 presents a dual S-curve response of T_r with respect to τ . As T_r decreases from point a to f , the $T_r - \tau$ curve passes four distinct turning points: b – hot flame extinction (HFE), c – hot flame ignition (HFI), d – cool flame extinction (CFE) and e – cool flame ignition (CFI). The similar dual S-curve response was also found in the literature (Shan and Lu 2012). Three branches on the $T_r - \tau$ curve, a - b , c - d , and e - f , are stable solutions while b - c and d - e are unstable solutions which physically not exist. The stable cool flame solution is the branch c - d .

The $T_r - \tau$ curve tells that a premixed cool flame can be established via tuning the τ value but only if $\tau_{\text{HFE}} > \tau_{\text{CFE}}$ or $\tau_{\text{HFI}} > \tau_{\text{CFI}}$ holds. Taking Fig. 1 as an example, since $\tau_{\text{HFI}} > \tau_{\text{CFI}}$, the cool flame can be formed by thermal ignition of an unburned mixture by gradually increasing the τ value, the unburned fresh mixture demonstrates two-stage ignition behavior following the path f - e - e' (cool flame)- c -hot flame. However, if a hot flame is established, decreasing the τ value only results in a thorough extinction of the hot flame to a no reaction state as $\tau_{\text{HFE}} < \tau_{\text{CFE}}$.

Figure 2 gives the revolution of the dual S-curves as the initial unburned temperature (T_u) changes from 500 K to 900 K. At $T_u = 500$ K, $\tau_{\text{HFI}} < \tau_{\text{CFI}}$ and $\tau_{\text{HFE}} < \tau_{\text{CFE}}$, which means that the cool flame can neither form via the cold-state ignition, nor via the hot flame extinction. For the cases of $T_u = 600$ K and 700 K, the S-curve behaviors are similar to the one demonstrated in Fig. 1. For the case of $T_u = 800$ K, the dual S-curve response migrates to a single S-curve, in which the cool flame ignition and extinction turning points disappear, forming a stretched monotonic response of T_r with respect to τ in the low-temperature zone. At $T_u = 900$ K, Fig. 2 clearly demonstrates that the cool flame branch disappears, which is straightforward that

Fig. 2 Dual S-curve response of the reactor temperature (T_r) with respect to the residence time (τ) at various initial unburned temperature (T_u) (DME/O₂/N₂ mixture, $\phi = 1.0$, $T_u = 500 - 900$ K, $x_{\text{N}_2,u}/x_{\text{O}_2,u} = 0$, $p = 1$ atm)

the low-temperature chemistry stops at 900 K. Figure 2 indicates that there is a T_u window (approximate 600 – 800 K) for the stabilization of a premixed cool flame.

Figures 3 and 4 presents the effects of the inlet N₂/O₂ mole ratio ($x_{\text{N}_2,u}/x_{\text{O}_2,u}$) and the reactor pressure (p) on the revolution of the dual S-curve response, respectively. The values of τ_{HFE} , τ_{HFI} , and τ_{CFE} considerably increase as $x_{\text{N}_2,u}/x_{\text{O}_2,u}$ increases, while on the contrary, decrease as p increases. Either the variation of $x_{\text{N}_2,u}/x_{\text{O}_2,u}$ or that of p has a moderate effect on τ_{CFI} . The cool flame temperature decreases as $x_{\text{N}_2,u}/x_{\text{O}_2,u}$ increases, while increases as p increases. At higher $x_{\text{N}_2,u}/x_{\text{O}_2,u}$ or lower p , the cool flame sustains over a broader τ range.

Fig. 1 Dual S-curve response of the reactor temperature (T_r) with respect to the residence time (τ). (DME/O₂/N₂ mixture, $\phi = 1.0$, $T_u = 650$ K, $x_{\text{N}_2,u}/x_{\text{O}_2,u} = 2/3$, $p = 1$ atm)

Fig. 3 Dual S-curve response of the reactor temperature (T_r) with respect to the residence time (τ) at various inlet N₂/O₂ mole ratios ($x_{\text{N}_2,u}/x_{\text{O}_2,u}$) (DME/O₂/N₂ mixture, $\phi = 1.0$, $T_u = 600$ K, $x_{\text{N}_2,u}/x_{\text{O}_2,u} = 0-6$, $p = 1$ atm)

Fig. 4 Dual S-curve response of the reactor temperature (T_r) with respect to the residence time (τ) at various pressure (p) (DME/O₂/N₂ mixture, $\phi = 1.0$, $T_u = 600$ K, $x_{N_2,u}/x_{O_2,u} = 0$, $p = 0.2$ –10 atm)

Figure 5 presents the revolution of the dual S-curve as the equivalence ratio (ϕ) changes from 0.1 to 10. For the cases of moderately lean ($\phi = 0.5$), near stoichiometric ($\phi = 1$) and moderately rich ($\phi = 2.5$) mixtures, the dual S-curve responses are similar to the one shown in Fig. 1 in which $\tau_{HFI} > \tau_{CFI}$ and $\tau_{HFE} < \tau_{CFE}$. At the extremely lean condition ($\phi = 0.1$), the value of τ_{HFE} becomes greater than that of τ_{CFE} and a hysteresis between hot flame and cool flame forms, which is consistent with the finding in counterflow studies under extremely lean condition ($\phi = 0.08$) in the literature (Reuter et al. 2016). In addition, Fig. 5 also indicated that the “cool flame-hot flame” hysteresis also forms under extremely rich condition

Fig. 5 Dual S-curve response of the reactor temperature (T_r) with respect to the residence time (τ) at various equivalence ratios (ϕ) (DME/O₂/N₂ mixture, $\phi = 0.1$ –10, $T_u = 600$ K, $x_{N_2,u}/x_{O_2,u} = 0$, $p = 1$ atm)

($\phi = 10$). Thus, for extremely lean and extremely rich mixtures, the cool flame can be established via 1) igniting an unburned mixture by increasing τ , and 2) extinguishing a hot flame by decreasing τ .

Figure 6 presents another type of S-curve, namely, the S-curve response of T_r with respect to T_u , in which the thermal ignition and extinction dynamics are clearly demonstrated. Similarly to the finding in the literature (Zhao et al. 2016), there is a τ window for the establishment of a premixed cool flame. For the case of $\tau = 10^{-3}$ s, as shown in Fig. 6, two-stage ignition behaviors can be found as T_u increases. The detailed process is as follows ($\tau = 10^{-3}$ s):

- the cool flame can be ignited when T_u increases beyond the cool flame ignition temperature ($T_{u,CFI} \approx 644$ K);
- increasing T_u to the range of $T_u = 800$ –1000 K, the cool flame will be quenched as the low-temperature chemistry stops.
- as T_u increases beyond the hot flame ignition temperature ($T_{u,HFI}$), the second-stage ignition occurs and a hot flame forms.

For the case of $\tau = 10^{-1}$ s, a direct transition to the hot flame is demonstrated as T_u reaches $T_{u,CFI}$. For the case of $\tau = 10^{-5}$ s, the residence time is too short to sustain a cool flame and the mixture will not be ignited until T_u reaches $T_{u,HFI}$.

In general, the PSR computation demonstrated three potential ways to form and stabilize a cool flame: a) igniting an unburned mixture via increasing the residence time, b) igniting an unburned mixture via increasing the unburned mixture temperature, and c) extinguishing an extremely lean or extremely

Fig. 6 Dual S-curve response of the reactor temperature (T_r) with respect to the initial unburned mixture temperature (T_u) at various residence times (τ) (DME/O₂/N₂ mixture, $\phi = 1$, $x_{N_2,u}/x_{O_2,u} = 1$, $p = 1$ atm, $\tau = 10^{-5}$ – 10^{-1} s)

rich hot flame via decreasing the residence time. In the next Section, the three ways will be demonstrated in the counterflow configuration, which can be realized in the practice.

Forming and Stabilizing Counterflow Cool Flames

The counterflow flame provides an ideal way to study laminar premixed flame dynamics. As schematized in Fig. 7, the counterflow consists of two opposed flows impinging on each other. The left flow is a N_2 stream heated to a certain temperature (T_{u,N_2}), and the right flow is the premixed DME/ O_2/N_2 mixture at the ambient temperature ($T_{u,m} = 300$ K). The temperature, species and momentum along the axis (z -axis in Fig. 7) of the counterflow flame were solved using the Fortran code OPPDIF (Lutz et al. 1997) integrated with CHEMKIN II subroutine library (Kee et al. 1989). The code was modified to account for the thermal radiation of CO, CO_2 , H_2O and CH_4 (Egolfopoulos 1994; Zhang et al. 2013). The computation domain, i.e., the separation distance (L) of the two stream exits, was 2 cm. The grid control parameters, GRAD and CURV were set to be 0.1 and 0.2 respectively to allow at least 300 grids along the axis. The stretch rate (κ) of the fuel stream is calculated using Eq. (1) (Law 2006).

$$\kappa = \frac{U_{u,m}}{L} \left(1 + \frac{U_{u,N_2} \sqrt{\rho_{u,N_2}}}{U_{u,m} \sqrt{\rho_{u,m}}} \right) \quad (1)$$

where ρ denotes the local density. In the present study, the quasi-steady state assumption was applied to the ignition or extinction state. This is reasonable as the chemical reaction is usually much faster than the changes of T_u and κ in laminar counterflow configuration.

Actually, in a previous study (Zhao et al. 2016), the premixed cool flame formation through increasing the initial unburned mixture temperature was numerically and experimentally demonstrated. In this section, more broadly, all the three ways found in Section 2 to form a premixed cool flame will be presented.

First, the counterflow computation was conducted to see whether a cool flame can be established through increasing the residence time. Figure 8 presents a two-stage ignition of a

Fig. 7 Schematics of the premixed counterflow flame configuration

Fig. 8 The maximum temperature (T_{max}) revolution during the two-stage ignition via decreasing the stretch rate (κ) (DME/ O_2/N_2 mixture, $\phi = 1$, $x_{N_2,u}/x_{O_2,u} = 1$, $p = 1$ atm, $T_u = 700$ K)

premixed DME/ O_2/N_2 mixture in the counterflow configuration. As the stretch rate κ decreases, a cool flame forms first followed by a hot flame ignition. The flashback occurs once the hot flame forms since the flame speed of the hot flame is much greater than the local flow velocity. To further confirm the existence of a premixed cool flame, Point Q on the curve in Fig. 8 was chosen as a typical cool flame case to check its thermal and species structure, as shown in Fig. 9. A positive heat production peak situated at the spot where $T \approx 720$ K, and the “cool flame marker”, i.e., CH_2O , is produced around this heat production peak. This confirms the cool flame is formed and stabilized by decreasing the stretch rate (increasing the residence time).

Fig. 9 Temperature, net heat production rate and CH_2O mole fraction distribution along the flame coordinate of a typical cool flame (Point Q in Fig. 8)

Second, the computation was conducted to demonstrate the ignition of a cool flame through increasing the initial unburned mixture temperature, and the results are in Fig. 10. The DME/O₂/N₂ mixture stream temperature ($T_{u,m}$) was kept at 300 K and the N₂ stream temperature ($T_{u,N2}$) was elevated from 300 K to 1000 K. Similarly, a two-stage ignition process is exhibited in Fig. 10. The cool flame forms when $T_{u,N2}$ reaches ~ 688 K, and quenches when $T_{u,N2}$ is in the range of 800–880 K because the low-temperature chemistry stops in this temperature range. Further increasing $T_{u,N2}$ results in an ignition of the hot flame. The results in Fig. 10 confirmed the feasibility of the cool flame forming and stabilization via the igniting an unburned mixture by increasing the unburned mixture temperature.

Third, the stabilization of a premixed cool flame via hot flame extinction was further proved in the counterflow configuration, as demonstrated in Fig. 11. The DME/O₂/N₂ was extremely rich ($\phi = 10$). A hot flame is first stabilized at a low stretch rate, as depicted in Fig. 11. As κ increases, the flame undergoes the first-stage hot flame extinction, forming a cool flame, and then the second-stage cool flame extinction. In this situation, the extinction limit is extended by the existence of the cool flame, which also theoretically proves the feasibility of the establishment of a premixed cool flame by the hot flame extinction.

Double-Structured Flames in a Counterflow

One interesting finding in the counterflow calculation is that self-sustaining steady-state decoupled double-structured

Fig. 10 The maximum temperature (T_{max}) revolution during the two-stage ignition via increasing the N₂ stream temperature ($T_{u,N2}$) (DME/O₂/N₂ mixture, $\phi = 1$, $x_{N2,u}/x_{O2,u} = 0.25$, $p = 1$ atm, $\kappa = 13$ s⁻¹)

Fig. 11 The maximum temperature (T_{max}) revolution during the two-stage extinction via increasing the stretch rate (κ) (DME/O₂/N₂ mixture, $\phi = 10$, $x_{N2,u}/x_{O2,u} = 1$, $p = 1$ atm, $\kappa = 13$ s⁻¹)

flames, i.e., a leading cool flame front followed by a premixed flame front with much higher (> 1000 K) flame temperature, are formed and stabilized. Taking Point W in Fig. 11 as an instance, the net heat production rate distribution curve presents two decoupled heat release peaks. The thermal, species and reaction rate structures of the doubled structured flames are demonstrated in Figs. 12, 13 and 14. The low-temperature peak situates at the location where $T \approx 740$ K under which the low-temperature chemistry dominates (Fig. 12). Correspondingly, CH₂O mole fraction curve demonstrates a “bump” at this location (Fig. 13). The relatively higher temperature peak situates at the spot where $T \approx 1038$ K (Fig. 12). At $T \approx 1038$ K, the CH₂O greatly converts to CO and the high-

Fig. 12 The thermal structure of the double flames (Point W in Fig. 11, DME/O₂/N₂ mixture, $\phi = 10$, $x_{N2,u}/x_{O2,u} = 1$, $p = 1$ atm, $\kappa = 2.8$ s⁻¹)

Fig. 13 The species structure of the double flames (Point W in Fig. 11, DME/O₂/N₂ mixture, $\phi = 10$, $x_{N_2,u}/x_{O_2,u} = 1$, $p = 1$ atm, $\kappa = 2.8$ s⁻¹)

temperature chemistry dominates (Fig. 13). Figure 14 depicts the reaction rate distributions of three elementary reactions: $H + O_2 = OH + O$ (R1), $H_2O_2 (+M) = 2OH (+M)$ (R20) and $CH_2OCH_2 + O_2 = CH_2OCH_2O_2$ (R249). R1 is a characteristic chain branching reaction of high-temperature chemistry. R20 is a characteristic chain branching reaction of intermediate-temperature chemistry. R249 is a typical reaction of low-temperature chemistry. It is clearly demonstrated in Fig. 14 that the first flame, namely, the low-temperature peak on the heat release curve, is controlled by the low-temperature chemistry, being a cool flame front. For the heat release peak at ~ 1038 K, the intermediate-temperature chemistry dominates

Fig. 14 The reaction rate distributions of the key elementary reactions of the double flames (Point W in Fig. 11, DME/O₂/N₂ mixture, $\phi = 10$, $x_{N_2,u}/x_{O_2,u} = 1$, $p = 1$ atm, $\kappa = 2.8$ s⁻¹, R1: $H + O_2 = OH + O$, R20: $H_2O_2 (+M) = 2OH (+M)$, R249: $CH_2OCH_2 + O_2 = CH_2OCH_2O_2$)

and the high-temperature chemistry plays a second role, while the effectiveness of the low-temperature chemistry in this zone is negligible. Thus, the second flame, at ~ 1038 K, should be regarded as an example of the “warm flame” (Lin et al. 2019) based on its thermal and chemical nature. The results in Fig. 14 gives firm evidence supporting that the two flames are controlled by different chemical mechanisms. The double-structured flames found in the present study is essentially different from those double flames in the literature (Ju et al. 2015; Reuter et al. 2017b; Zhang et al. 2018). In Reuter et al.’s experiments (Reuter et al. 2017b), the observed double flames are due to the “partially premixed-diffusion” configuration, and consequently their double flames are a combination of a premixed flame and a diffusion flame. In Ju et al.’s simulation (Ju et al. 2015), the double flames are found in the 1D freely-propagating flame model which is difficult to realize in the practice. Moreover, the double flames in the 1D freely-propagating flame model are limited to the fuel-lean side (Ju et al. 2015). In Zhang et al.’s computation (Zhang et al. 2018), the double flames are indeed a transient state that lasts no more than 0.5 s.

For comparison, the thermal and species structures of Point E (cool flame) of Fig. 11 are presented in Figs. 15 and 16, respectively. Only a single structure cool flame is found, which situated at the place where $T = 712$ K (Fig. 15). CH₂O and CO mole fraction distributions also clearly indicate that these products are only produced in the low-temperature cool flame zone.

One interesting question is whether the double-structured premixed flames can also form in fuel-lean and near-stoichiometric flames. The authors have tried their best but failed to numerically establish the similar double-structured flames for fuel-lean or near-stoichiometric mixtures. This is

Fig. 15 The thermal structure of the single cool flames (Point E in Fig. 11, DME/O₂/N₂ mixture, $\phi = 10$, $x_{N_2,u}/x_{O_2,u} = 1$, $p = 1$ atm, $\kappa = 5$ s⁻¹)

Fig. 16 The species structure of the single cool flames (Point E in Fig. 11, DME/O₂/N₂ mixture, $\phi = 10$, $x_{N_2,u}/x_{O_2,u} = 1$, $p = 1$ atm, $\kappa = 5$ s⁻¹)

not concluding that the similar double-structured premixed flames are impossible in fuel-lean or near-stoichiometric flames. Physically, the cool flame situates in the upper-stream where the local flow velocity is higher. Thus, a necessary requirement to form the double-structured flames is that the cool flame speed needs to be higher than the hot or warm flame, so that both flames can find the suitable locations to establish local dynamic balances. The attempt to find double-structured premixed flames of fuel-lean and near-stoichiometric mixtures remains as an open question and will be a future study.

It still needs experimental evidence to further prove the three ways of establishing a self-sustaining steady-state premixed cool flame as well as the double-structured flames which are numerically demonstrated in the present study. One should also note that the premixed cool flame is a near-limit weak flame. The buoyance induced natural convection may greatly affect the flame and may conceal the premixed cool flame dynamic behaviors if the experiments were carried out on earth gravity. Micro-gravity experiments, e.g., experiments in a drop tower, onboard of an aircraft or a satellite, may be needed if accurate premixed cool flame data are desired.

Conclusions

The formation and stabilization of a self-sustaining steady-state premixed cool flame without external sensitization were numerically studied. By analyzing the PSR dual S-curve behavior, three ways to form and stabilize a self-sustaining premixed cool flame were indicated. The three ways are: 1) igniting the unburned fresh mixture by decreasing the residence time, 2) igniting the unburned mixture by

increasing the temperature of the unburned fresh mixture, and 3) extinguishing an extremely lean or an extremely rich hot flame by decreasing the residence time. The three proposed ways were then successfully numerically demonstrated in the counterflow premixed flame configuration. It was also found that there were double-structured premixed flames, namely, a leading cool flame front followed by a warm flame, formed under the extremely rich condition. Further micro-gravity experiments are needed to confirm the numerical findings in the present study.

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