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Extinction and flame bifurcations of stretched dimethyl-ether premixed flames

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Abstract

Extinction limits and flame bifurcation of lean premixed dimethyl ether-air flames are investigated numerically using the counterflow flame with a reduced chemistry. Emphasis is paid to the combined effect of radiation and flame stretch on the extinction and flammability limits. A method based on the reaction front is presented to predict the Markstein length. The predicted positive Markstein length agrees well with the experimental data. The results show that flow stretch significantly reduces the flame speed and narrows the flammability limit of the stretched dimethyl ether-air flame. It is found that the combined effect of radiation and flow stretch results in a new flame bifurcation and multiple flame regimes. At an equivalence ratio slightly higher than the flammability limit of the planar flame, the distant flame regime appears at low stretch rates. With an increase of the equivalence ratio, in addition to the distant flame, a weak flame isola emerges at moderate stretch rates. With a further increase of the equivalence ratio, the distant flame and the weak flame branches merge together, resulting in the splitting of the weak flame branch into two weak flame branches, one at low stretch and the other at high stretch. Flame stability analysis demonstrates that the high stretch weak flame is also stable. Furthermore, a K-shaped flammability limit diagram showing various flame regimes and their extinction limits are obtained.

Key words: Dimethyl ether, extinction limit, radiation, bifurcation, stretch.
1. Introduction

Dimethyl ether (DME) has low soot emission, high tolerance to EGR, and causes no contamination of ground water. In addition, it can be synthesized from natural gas, coal, and bio-fuels and has been considered as a neat diesel substitute fuel and house cooking fuel. The study of DME combustion attracts great attentions [1-5]. Recently, detailed kinetic mechanisms for low and high temperature DME oxidations [6-8] have been established. Applications of DME on diesel engine have been investigated by real engine performance tests and engine CFD simulations [9-10]. One of the key issues to achieve best performance is to know how lean it can burn and how fast the flame can propagate in a turbulent flow.

The laminar burning velocity of DME-air mixtures was measured by Daly et al. [11] and Zhao et al. [12], respectively, using the spherical bomb and the counterflow flame. However, practical flames are always stretched. Although DME has a mean molecular weight larger than air, it decomposes quickly into CH₃ and CH₃O which have less and comparable mean molecular weights than air. As a result, DME and its decomposed species play opposite roles in affecting stretched flame burning. As such, it is of great interest to understand how flame stretch affects flame propagation and the flammability limit of DME-air combustion. Furthermore, since DME combustion involves high EGR, radiation impact on flame extinction via CO₂ emission has to be considered. Unfortunately, there is no report available on flame extinction for DME-air combustion.

The effect of radiation on stretched methane-air flames has been extensively investigated using counterflow flames [12-19]. The results showed that radiation flame interaction results in complicated flame bifurcations and radiation extinction. The previous study based on the one-step chemistry [19] demonstrated a transition from G- to K-shaped extinction curve at large Lewis numbers and the existence of highly stretched weak flame. However, the existence of this flame has not been confirmed using a practical fuel and detailed chemistry.

This study is motivated by the above discussions to investigate numerically the combined effect of stretch and radiation on the extinction limits and flame bifurcations of lean premixed dimethyl ether-air flames using a detailed chemistry. At first, the Markstein length is obtained by linearly extrapolating the flame speeds of weakly stretched lean DME-air flames to zero stretch rate. Several extrapolation methods based on different flame locations are employed and compared. Then, the flammability limit of planar flame is computed. This is followed by the
simulation of flame response with stretch rate using the radiative counterflow flame. Different flame regimes, limits, and bifurcations are discussed. Finally, flame stability on high stretched weak flame is examined and the flammability diagram is obtained.

2. Numerical Models

The axisymmetrical back-to-back counterflow premixed flames are considered in this study. The gaseous mixtures are issued from two opposed burners forming two planar flames near the stagnation plane. The governing equations based on the potential flow assumption were given in Ref. [17]. The fuel is DME (CH$_3$OCH$_3$), and the oxidizer is air. A reduced chemistry including 39 species (N$_2$, O$_2$, H$_2$, H$_2$O, H, HO$_2$, OH, O, H$_2$O$_2$, CO, CO$_2$, HCO, CH$_3$, CH$_3$OCH$_3$, CH$_4$, CH$_2$, CH$_2$O, C$_2$H$_4$, CH$_3$O, C$_2$H$_6$, C$_2$H$_5$, C$_2$H$_3$, CH$_3$OH, CH$_2$OH, CH$_3$O, CH$_3$OCH$_2$, CH$_3$OCH$_2$O, CH$_2$OCH$_2$O, CH$_3$OCH$_2$O$_2$, and 168 reactions was developed using the CSP reduced algorithm [20] from the full mechanism [7], which originally has 78 species and more than 300 reactions. As will shown later, this reduced mechanism can reproduce the experimental data [12] reasonably well and provides much better convergence and efficient computation time than the full chemistry.

In the radiation calculation, we employ the optically thin model. The radiation heat loss is determined by using the Planck mean absorption coefficients which are calculated for CO$_2$, H$_2$O and CO using the statistical narrow-band model. This is a reasonable and computational efficient choice for DME-air flames because the CO$_2$ concentration in thermal diffusion zone is at the same level of CH$_4$-air flame so that the radiation reabsorption effect can be neglected [21].

The unburned temperature of the mixture is 300 K and the pressure is 1 atm. Detailed transport properties are computed from the Chemkin database if they are available. For those species whose transport properties are not available, the transport parameters are estimated using the similar molecules. For steady-state solutions, the governing equations are solved by a revised version of the Chemkin code [22] with an improved arc-length continuation method [17]. For stability analysis, the initial stretch rate is perturbed by one percent and the unsteady solution is computed until the flame reaches steady state solution or jump to other flame branch. The time
step for unsteady computation is between one hundred and ten microseconds. The computation domain is 10 cm.

3. Results and Discussion

*Flammability limit and effect of flame stretch on flame speed*

Figure 1 shows the dependence of flame speeds of the one-dimensional (1D) planar DME-air flames ($S_{L0}$) on the equivalence ratio with and without radiation. It is seen that for the 1D flame, radiation plays a negligible role for equivalence ratio ($\Phi$) larger than 0.6. However, with the decrease of equivalence ratio, radiation increasingly reduces flame speed and causes flame extinction at $\Phi_0 = 0.454$. Hereafter, in order to differentiate the flammability limit of the stretched flame from that of the planar flame, we will call the extinction limit of the planar flame as the fundamental flammability limit. The experimental data by Zhao et al. [12] are also plotted in Fig.1. It is seen that the reduced chemistry can reproduce the experimental data reasonably well. In fact, the difference between the full chemistry and the present reduced chemistry is less than 5% for all lean DME-air flames.

For weakly stretched, adiabatic flames, the effect of flame stretch on flame speed ($S_L$) can be approximately treated as a linear function using the Markstein length $L$: $S_L/S_{L0} = 1 + (L/\delta_f)*K_a$, where $K_a$ denotes the normalized stretched rate and $\delta_f$ is the flame thickness. Therefore, the Markstein length for DME-air flames can be extracted directly from the dependence of $S_L$ on stretch rate using the counterflow flame. Two methods in the previous studies have been used in calculating the flame speed of the counterflow flame. One method is to use the minimum velocity in front of the flame [22] as the stretched flame speed ($S_{L_{\text{min}}}$) and the other method [23] is to use the mass flow rate at the peak chemical heat release to determine the stretched flame speed: $S_{L_{\text{max}}} = (\rho u)_{\text{max}} / \rho_u$. It should be noted that in counterflow flames the mass flow rate
decreases along the streamwise direction. Therefore, it is necessary to examine the sensitivity of the flame speed on the change of mass flow rate. For comparison, we choose two other locations: a. flame front (S_{L01}, 1% chemical heat release) and b. center of flame zone (S_{L50}, 50% chemical heat release), to obtain the stretched flame speed. The reason why the flame speed based on the minimum velocity increases with stretch rate is because the mass loss in the thermal diffusion zone between the minimum velocity point and the flame zone increases with the stretch rate, that is, the mass flux at the minimum velocity point over predicts the real burning flux.

Figure 2 shows the typical dependence of flame speeds of the stretched flames as a function of stretch rate at $\Phi=0.7$ using the above four different reference locations. It is seen that all the four methods yield linear dependences of flame speed on stretch rate. The three methods based on the reaction zone (S_{L01}, S_{L50} and S_{Lmax}) all predict negative dependence of stretched flame speed on the stretch rate. The methods based on the maximum and 1% chemical heat release, respectively, predict the lowest and the highest flame speeds. This is understandable because flow stretch causes flow leakage between the flame front and the location of maximum chemical heat release. This difference should be proportional to the product of the stretch rate and the flame thickness. However, the method based on the minimum flow velocity gives a positive gradient.

A comparison of the predicted Markstein number with the experimental data of the lean mixture at various equivalence ratios is shown in Fig.3. It is seen that all the three methods based on the reaction zone locations yield positive Markstein lengths. This means that flame speed will decrease with the increase of flame stretch for lean DME-air flames. The predicted results agree well with the recent experimental data [26]. Furthermore, it is seen that the method based on the reaction front predicts the best results. However, the method based on the minimum flow velocity predicts negative Markstein numbers for lean DME-air flame. This implies that flame speed will
increase with flame stretch. This is not reasonable for DME-air flame. Therefore, it is appropriate to use the method based on the flame front in counterflow flames to computationally extract the effect of flow stretch on the flame speed. The method based the counterflow flame provides a great advantage over the employment of the spherical flame because of its computation efficiency.

**Combined effect of radiation and stretch on near limit flames**

For non-adiabatic stretched flames, the dependence of maximum flame temperature on stretch rate for various equivalence ratios is shown in Fig.4. It is seen that at $\Phi = 0.5$, flame temperature decreases rapidly with the increase of stretch rate. At stretch rate ($a$) of 5.0 s$^{-1}$, flow stretch causes flame extinction. Therefore, flame stretch significantly weakens the flame. As a result, flames at equivalence ratios between $\Phi = 0.5$ and the fundamental flammability limit ($\Phi_0 = 0.454$) can only exist at very low stretch rates. Because these flame locations are very far away from the stagnation plane, hereafter we call them as the distant flame (DF).

As equivalence ratio increases up to $\Phi = 0.505$, in addition to the DF, there exists a new flame isola at moderate stretch rates. This is the so called weak flame (WF) [17, 18]. Compared to the DF, the weak flame has a much smaller flame separation distance and a lower flame temperature. In addition, the weak flame has two extinction limits, a radiation extinction limit at low stretch rate and a stretch extinction limit at high stretch rate. However, the DF only has one stretch extinction limit on the high stretch side. Moreover, the DF reduces to the planar flame in the limit of zero stretch rate. This is clearly seen in Fig.4 for $\Phi = 0.51$.

As the equivalence ratio further increases, Fig.4 shows that a new bifurcation occurs at $\Phi = 0.52$. Due to the merge of the DF and WF flame branches, the WF flame branch is divided into two flame branches $ef$ and $cd$, respectively at low and high stretch rates. This bifurcation has not been found for other practical fuels and is a unique phenomenon of DME-air flame. The low
stretch weak flame (LWF) has a radiation extinction limit at point $e$ and a jump limit at point $f$. A further decrease of stretch rate from point $e$ will result in extinction of the weak flame. On the other hand, a further increase of the stretch rate from point $f$ will cause the flame to jump to the DF flame branch. Similarly, on the high stretch weak flame (HWF) branch ($cd$), there are a stretch extinction point at $d$ and a jump limit at $c$. Therefore, for the moderately stretched lean DME-air flames, there exist multiple flame regimes. Furthermore, two flame regimes can coexist at the same boundary conditions in a wide range of stretch rates. Stability analysis in next section will show that the HWF branch is stable. As such, it is expected that a well designed microgravity experiment may be able to observe this flame regime.

As the equivalence ratio further increases, both the distant flame and the high stretch weak flame move to large stretch rate side. It is seen that the flammable range of the HWF becomes narrower and eventually disappears as equivalence ratio approaches 0.6. On the other hand, the low stretch weak flame shifts to lower stretch rate as the equivalence ratio increases. However, different from the 1D flame, the LWF branch never disappears even when the equivalence ratio becomes unity. Therefore, in summary, the combined effect of radiation and stretch causes distinct non-linear bifurcations and multiple flame regimes in moderate stretch rates for near limit DME-air flames.

Figure 5 shows the dependence of the flame locations of different flame regimes for $\Phi = 0.52$ as a function of stretch rate. It is clearly seen that the distant flame stays far away from the stagnation plane. As the stretch rate decreases, flame moves towards the stagnation plane and extinguishes at a finite distance. This is a typical stretch extinction phenomenon for mixture with a Lewis number larger than unity. On the other hand, the high stretch weak flame stands very close to the stagnation plane. With a decrease of the stretch rate, flame moves away from the stagnation plane and finally jumps to the DF flame branch at point $c$. The low stretch weak flame
also stays near the stagnation plane. In summary, for the very lean DME-air mixtures, the DF can only exist far away from the stagnation plane and the HWF and LWF only stay near the stagnation plane. Flame cannot exist at moderate separation distances because of the radiation effect.

In order to understand this phenomenon, we now examine the radiative heat loss effect. For planar flame, it is well known that the normalized flame speed \( m = S_L / S_{lad} \) depends on the normalized heat loss \( H \) as

\[
\ln m = -2H / m^2
\]

(1)

From the author’s previous analysis [25], by neglecting the Lewis number effect the dependence of normalized flame speed of the counterflow flame on heat loss can also be written as

\[
\ln m = H_r
\]

(2)

where \( H_r \) accounts radiative heat loss in both burned and unburned regions. Figure 6 shows the dependence of the radiation heat loss in counterflow flame as a function of normalized flame separation distance \( \eta_f = x_f \sqrt{a/2} \). It is seen that near the stagnation plane \( \eta_f = 0 \), the radiation heat loss is very small. This is why the weak flame can sustain near the stagnation plane. However, as the flame separation distance increases, there is a rapid increase of radiation heat loss. In addition, the radiation heat loss from the burned gas peaks at a moderate flame separation distance. This is why when the equivalence ratio is not high enough, neither the DF nor the WF can sustain in this region. At an infinite separation distance, Fig.6 shows that the normalized heat loss becomes unity on both burned and unburned sides. As such, Eq.(2) reduces to Eq.(1).

The radiation fraction (ratio of total radiation heat loss to the total chemical heat release) is shown in Fig. 7. It is seen that at the radiation extinction limit of the LWF branch \( e \), radiation
fraction becomes the maximum. On the other hand, on the jump limit \( f \), radiation fraction becomes the minimum. This branch is radiation dominant. On the HWF branch \( cd \), the competition between the stretch rate effect and the radiation heat loss plays a dominant role.

**Stability of the high stretched weak flame**

The stability of the low stretch weak flame has been examined in previous study. The appearance of the high stretch weak flame is a new phenomenon. To examine the stability of this flame branch, we choose two typical points on the HWF branch. One point is at \( a=9 \text{ s}^{-1} \) which is in the middle of this flame branch (Fig.4). The other point is at \( a=10 \text{ s}^{-1} \) which is close to the stretch extinction limit \( d \) in Fig.4). The steady state solution is first obtained at each stretch rate. Then, the steady state solution is perturbed by 1\% of temperature or 1\% of stretch rate changes at each grid point. The perturbed profile is used as the initial solution for unsteady computation. The unsteady computation continues until the steady state solution is reached. Figure 8 shows the time evolution histories of flame temperatures for \( a=9 \text{ s}^{-1} \) and \( 10 \text{ s}^{-1} \) after initial perturbation. For stretch rate of \( 9 \text{ s}^{-1} \), flame initially moves quickly away from the stagnation plane. As a result, flame temperature increases in the beginning. However, after the initial spike, flame temperature decreases and converges quickly to the steady state solution. For stretch rate of \( 10 \text{ s}^{-1} \), since flame is very weak, after a small temperature increase, flame immediately converges to its steady state solution. Therefore, the flame at the high stretch weak flame branch is stable. This is also consistent with the physical process because the high stretch weak flame at \( \Phi =0.52 \) was originally divided from the upper right part of the weak flame branch of \( \Phi =0.51 \).

**Diagram of limits and flammable regions for different flame regimes**
Figure 9 shows the diagram of the extinction and flammability limits of various flame regimes. It is seen that the extinction curve is K-shaped. This result agrees well with the prediction of the G-K transition at large Lewis number based on the one-step chemistry and constant transport properties [19]. The G-K transition occurs at Lewis number larger than 1.4. This is also reasonable because the Lewis number of DME-air mixture is around 1.6.

In Fig.9, AB is the stretched extinction limits of normal flame which can be measured in ground experiments. BC is the stretch extinction limit of the distant flame. It is seen that only the extension of the distant flame to zero stretch rate generates the fundamental flammability limit ($\Phi = 0.454$). BD is the stretch extinction limit of the high stretch weak flame and BF is the jump limit of high stretch weak flame. DE is the jump limit of low stretch weak flame and FG is the radiation extinction limit of the weak flame. This K-shaped extinction curve agrees well with the numerical prediction using the one-step chemistry in the authors’ previous study. However, it is the first time to show that this bifurcation occurs for a real practical fuel.

4. Conclusions

The Markstein length of lean dimethyl ether-air premixed is extracted from the counterflow flames using a method based on the flame front. The predicted positive Markstein length agrees well with the experimental data. The results showed that flow stretch significantly reduces the flame speed and narrows the flammability limit of the lean dimethyl ether-air flame.

It is found that the combined effect of radiation and stretch results in a new flame bifurcation and multiple flame regimes for DME-air flames. At equivalence ratio slightly higher than the flammability limit of the planar flame, the distant flame regime appears at low stretch rate. With an increase of equivalence ratio, in addition to the distant flame, a weak flame isola appears at moderate stretch rates. With a further increase of equivalence ratio, the distant flame and the weak flame merge together which leads to the splitting of the weak flame branch into two new flame branches, a low stretch weak flame and a high stretch weak flame. Flame stability analysis demonstrated that the high stretch weak flame is also stable. A K-shaped flammability diagram showing various flame regimes and their flammable regions are obtained. The results demonstrated for the first time that the complicated bifurcations predicted using the one-step chemistry really happen for the DME-air flames.
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Figure Captions

Fig.1 Predicted flame speed and flammability limit of the one-dimensional, planar, DME-air premixed flame with and without radiation heat loss using reduced chemistry.

Fig.2 Flame speed vs. stretch rate for DME-air premixed flame at equivalence ratio of 0.7.

Fig.3 Comparison of predicted Markstein number with experiments for DME-air premixed flames.

Fig.4 Dependence of maximum flame temperature on the stretch rate at various equivalence ratios.

Fig.5 Dependence of flame separation distance on the stretch rate at equivalence ratio of 0.52.

Fig.6 Dependence of normalized radiative heat loss on the flame location in counterflow flame.

Fig.7 Dependence of radiation fraction on the stretch rate at equivalence ratio of 0.52.

Fig. 8. The time evolution history of high stretch weak flame after initial perturbation of stretch rate.

Fig.9. The diagram of flammable regions: extinction limits vs. equivalence ratio.
Fig. 1

DME-air

Flame speed, cm/s
Equivalence ratio

- full chemistry
- adiabatic (reduced)
- radiative (reduced)

Zhao et al. (2001)
Flame speed, $S_L$ (cm/s)

Stretch rate, $a$ (s$^{-1}$)

$\Phi = 0.7$

$S_{L\text{min}}$

$S_{L\text{max}}$

$S_{L01}$

$S_{L50}$

25.61 cm/s

Fig. 2
Fig. 3
Fig. 4
Flame position (cm) vs. Stretch rate, $a$ (s$^{-1}$)

$\Phi = 0.52$

Fig. 5
Fig. 6

Heat loss, $H_r$, normalized by $H/m^2$

- Burned gas side
- Unburned gas side
Fraction of radiation heat loss

Stretch rate, $a$ (s$^{-1}$)

DME-Air
$\phi = 0.52$

Fig. 7
Fig. 8

Maximum flame temperature (K)

\[ \phi = 0.52 \]
LWF branch

- \[ a = 9 \text{ s}^{-1} \]
- \[ a = 10 \text{ s}^{-1} \]
Stretch rate at extinction and jump limits, $s^{-1}$

Equivalence ratio, $\Phi$

DME-air

$\Phi = 0.454$

Fig. 9