GLOBAL QUENCHING OF PREMIXED CH₄/AIR FLAMES: EFFECTS OF TURBULENT STRAINING, EQUIVALENCE RATIO, AND RADIATIVE HEAT LOSS

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Global quenching of premixed CH₄/air flames with turbulent strain, equivalence ratio, and radiative heat loss effects is explored in a cruciform burner. The burner equipped with a pair of counter-rotating high-speed fans and perforated plates provides downward propagating flames through near-isotropic intense turbulence, where flame-turbulence interactions are not influenced by ignition. Several CH₄/air flames with different degrees of radiative heat loss, from small (N₂-diluted) to large (CO₂-diluted), are investigated. Each case covers a range of the equivalence ratio (ϕ) with turbulent intensities (u'/S_l) as much as 100, where S_l is the laminar burning velocity, in which high rates of strain are achieved until, ultimately, global quenching of flames occurs. A Bradley's Karlovitz number, defined as \( K = 0.157(u'/S_l)^2 Re_T^{-0.5} = 0.157 K_a \), is used to quantify global quenching boundaries of these turbulent flames, where \( Re_T \) and \( K_a \) are the turbulent Reynolds and Karlovitz numbers, respectively. For pure CH₄/air flames, the critical value of \( K \) for global quenching of rich/lean CH₄ flames must be greater than 1.0/6.2. Values of \( K \) are very sensitive to \( \phi \) because \( K \) increases significantly as \( \phi \) gradually approaches 1 from either lean or rich sides, with the maximum \( K \) occurring possibly near \( \phi = 1 \). By comparing N₂- and CO₂-diluted flames of the same \( S_l \), it is found that global quenching of lean/rich CH₄ flames is/is not influenced by the radiative heat loss, respectively. The larger the radiative heat loss, the smaller the value of \( K \) for lean CH₄ flames, in which values of \( K \) decrease from 4 (N₂-diluted) to 3 (CO₂-diluted) where \( S_l \approx 10 \) cm/s and \( \phi = 0.62 \). On the other hand, \( K \approx 1.3 \) for both N₂- and CO₂-diluted rich CH₄ flames where \( S_l \approx 10 \) cm/s and \( \phi = 1.20-1.45 \). These experimental results are important to the understanding of global quenching processes for turbulent premixed combustion.

Introduction

Global quenching of premixed flames is of both fundamental and practical importance. When a premixed flame experiences external perturbations, such as aerodynamic stretch or heat losses, local quenching of the flame may occur if the perturbations are strong enough to reduce the reaction rate in the flame to a negligible value. There are many studies on local quenching of laminar premixed flames. For instances, asymptotic analysis by Libby et al. [1], numerical simulations by Darabiha et al. [2,3], and experimental studies by Ishizuka and Law and others [4-6] have agreed that quenching by stretch may occur if the flow is non-adiabatic or if the Lewis number (Le) > 1. Law [7] has described dynamics of stretched laminar flames. Poinset et al. [8] in a direct numerical simulation and Driscoll and coworkers [9,10] in an experimental study of flame/vortex interactions have further enhanced our understanding on local quenching processes of laminar premixed flames. On the other hand, only a few studies are available for global quenching (a complete extinction, not local quenching) of turbulent premixed flames (see Ref. [11]), because how to model turbulent combustion with a wide range of temporal and spatial scales of both flow and chemistry at high Reynolds numbers is an extremely difficult task [12]. This motivates us to study the problem experimentally.

Since Karlovitz [13] first introduced the concept of flame stretch about five decades ago, the turbulent Karlovitz number, which was commonly defined as \( K_a = (u'/\lambda)/(S_l/\delta_k) = (u'/S_l)^2 Re_T^{-0.5} \), has been used to describe flame quenching by turbulent stretching. Here \( u' \), \( S_l \), and \( \lambda \) are the turbulent intensity, laminar burning velocity, and Taylor micro-length scale, respectively. \( \delta_k (=v/S_l) \) is the flame thickness and \( Re_T = u'L_4/v \), where \( L_4 \) and \( v \) are the turbulent integral length scale and the kinematic viscosity of reactants, respectively. In 1987, Bradley and coworkers [14] found that the Karlovitz number at global quenching of premixed turbulent flames, defined as \( K = 0.157(u'/S_l)^2 Re_T^{-0.5} = 0.157 K_a \), was unity using an explosion bomb. Bradley [15] further modified the global quenching condition as \( K Le \approx 6 \). Although the explosion bomb has an advantage of having high turbulent intensities with negligible
Fig. 1. Schematic of the cruciform burner (central) with left/right sequential images that display typical flame propagation/near quenching of premixed CH$_4$/air flames, where the experimental conditions are $\phi = 1.0/0.6$, $u'/S_L = 3.68/69.30$, and $K = 0.04/5.71$, respectively. The concentration of remaining CH$_4$ after turbulent combustion is measured by the gas chromatography.

mean velocities, it suffers some disadvantages that flame development and quenching are influenced by the ignition source and by non-uniform distributions of mean reactant temperature and pressure during the explosion. This makes the determination of actual global quenching conditions somewhat difficult. To avoid the ignition problem and further consider the effect of radiative heat loss on global quenching of premixed turbulent flames, a new methodology is presented.

Using a cruciform burner (Fig. 1), the effects of turbulent straining, equivalence ratio, and radiative heat loss on global quenching of premixed CH$_4$/air flames are investigated. The long vertical vessel provides a downward propagating flame with large surface area. The large horizontal vessel equipped with a pair of counter-rotating fans and perforated plates at the two ends provides intense near-isotropic turbulence in the core region between the two perforated plates \([16–18]\). Thus, flame-turbulence interactions are not influenced by the ignition source. Moreover, N$_2$ and CO$_2$, as diluents, are applied to study the effect of radiative heat loss. Note that the importance of radiative heat loss on turbulent flame extinguishment has been anticipated \([19–21]\) using liquid flames which have little heat losses and are extremely difficult to quench globally. Each case covers a range of the equivalence ratio ($\phi$) with values of $u'/S_L$ ranging from 0 to about 100, in which high rates of strain are achieved until, ultimately, global quenching of flames occurs. The following section reviews experimental methods used in this study and is followed by a description of dynamics of turbulent premixed flames from propagation and pocket formation to global quench. Both are then applied to determine the global quenching boundaries on a $K$-$\phi$ plot for these CH$_4$/diluent/air flames. Thus, the critical value of $K$, $K_c$, required to quench globally these premixed flames as a function of $\phi$ can be obtained. By comparing the behavior of N$_2$- and CO$_2$-diluted flames, the effect of radiative heat loss on $K_c$ is also determined.

**Cruciform Burner and Intense Near-Isotropic Turbulence**

Figure 1 shows a schematic of the cruciform burner, in which a large domain of intense turbulence can be generated in the core region ($\sim 15 \times 15 \times 15$ cm$^3$) between the two perforated plates. The turbulent intensities are very high with negligible mean velocities, values of skewness and flatness are nearly zero and three, respectively, and the corresponding energy spectrum has a $5/3$ slope. This burner has been used to quantitatively measure turbulent burning velocities ($S_T/S_L$) over a greater parameter range of $u'/S_L$ than hitherto. For details on the cruciform burner, the corresponding turbulence statistics (e.g., length scales), and values of $S_T/S_L$ as a function of $u'/S_L$, $Re_T$, $\phi$, and the Damköhler number, the reader is directed to Refs. \([16–18]\).

Before a run, the cruciform burner was evacuated and then filled with CH$_4$/air mixtures without or with diluting gases at a given $\phi$ ranging from 0.60 to 1.45 at 1 atm. A run began by ignition and simultaneous opening of four large venting valves at the top of the vertical vessel, generating a downward propagating premixed flame with large surface area. This
is 170 Hz, corresponding to velocities \([14,17]\).

hypothesis and Bradley’s correlation for zero mean view \([11.5]\) taken from the near-isotropic region with a field of given values of \(K\) for multitudinous interactions with flames. At any age of CH\(_4\) fuel (c/c\(_i\)) after a run, measured by the gas which many of large vortices (\(L\)) flame then enters the central uniform region in which many of large vortices (\(L\)) up to 5 cm) reside for multitudinous interactions with flames. At any given values of \(\phi\), the maximum fan frequency \((f_{\text{max}})\) is 170 Hz, corresponding to \(u’ = 785.4 \text{ cm/s}\) and \(Re_T = 24,850\), where \(L\) is estimated from Taylor’s hypothesis and Bradley’s correlation for zero mean velocities \([14,17]\).

**Flame Propagation, Pocket Formation, and Global Quenching**

On the left of Fig. 1, three sequential images, taken from the near-isotropic region with a field of view \(11.5 \times 10.0 \text{ cm}^2\) using a high-speed camcorder, display a typical flame propagation, which is statistically stationary as already verified \([17]\). Prior to global quench (the right side of Fig. 1), turbulent flames are largely disrupted, flames become very brushy (distributed-like), and the formation of pockets or islands is observed even though the visualization has an integrating effect. These disrupted flames with low burning rates can sustain in the near-isotropic region for a period that is much longer than typical flame propagation cases such as the one presented on the left of Fig. 1. After 76 ms, these distributed-like turbulent flames, upon slow propagation, approach the lower vertical vessel and finally consumed all remaining reactants. Here the time origin is chosen at the time of the first image presented in Fig. 1 for both flame propagation and near-quenching cases. If values of \(u’/S_L\) and/or \(K\) can be increased further, these aforementioned flames would be globally quenched as they enter the intense turbulence region (no flame propagation). Hence, it is unambiguous to determine whether global quench would occur using the high-speed camcorder.

The alternative method in determining flame quenching is via the gas chromatography. After a run, we sample the products in the region of interest (Fig. 1) to measure the remaining CH\(_4\) concentrations as a function of \(u’/S_L\) and/or \(K\). As a typical example, Fig. 2 shows variations of the normalized remaining percentage of CH\(_4\) fuel as a function of \(u’/S_L\) for both very rich \((\phi = 1.45)\) and very lean \((\phi = 0.6)\) CH\(_4\)/air flames. Also plotted are values of \(K\) just before and after global quenching. Across the transition of global quenching, the remaining CH\(_4\) concentration increases drastically for both rich and lean cases, as can be seen in Fig. 2. The critical value of \(K\) to quench globally rich CH\(_4\)/air flames at \(\phi = 1.45\) whose \(Le = 1.04\) is about 1.5. Note that even much higher values of \(K\) \((>6)\) are required for global quenching of lean CH\(_4\)/air flames at \(\phi = 0.6\) whose \(Le = 0.97\). Thus, lean CH\(_4\) flames are much harder to quench globally than rich CH\(_4\) flames, indicating an influence of \(Le\).

**Accessible/Inaccessible Domains**

The maximum \(f = 170 \text{ Hz}\) determines the accessible domain on a \(K-\phi\) plot using the present experimental configuration. For pure CH\(_4\)/air flames, both the maximum Bradley’s Karlovitz number \((K_{\text{max}})\) and \(S_L\) as a function of \(\phi\) are shown on Fig. 3a, where \(S_L\) is from measurements of Vagelopoulos et al. \([22]\). The shaded area in Fig. 3a represents the accessible domain that is determined by the curve of \(K_{\text{max}}\) and the two lines of \(\phi = 0.6\) and \(\phi = 1.45\), which are the leanest and richest mixtures in the cruciform burner. As can be seen from Fig. 3a, the accessible domain is very limited, because the larger the values of \(S_L\) the smaller the values of \(K_{\text{max}}\). Only very lean or very rich CH\(_4\)/air flames can experience high rates of turbulent straining \((K > 1)\) in the present cruciform burner.

To expand the accessible domain for higher \(K_{\text{max}}\), N\(_2\) and CO\(_2\) gases are diluted with CH\(_4\) fuel to reduce \(S_L\), as shown on Fig. 3b and c, respectively. Values of \(S_L\) for N\(_2\)- and CO\(_2\)-diluted flames in Figs. 3b, c are from Stone et al. \([23]\). Concerning the radiative heat loss, CO\(_2\)-diluted flames experience more heat losses than N\(_2\)-diluted flames, because of a higher concentration of CO\(_2\) in the products. Smaniego and Mantel \([24]\) used a heat loss coefficient \((HL)\), a ratio of the energy radiated in the flame zone to chemical energy release, for quantifying radiative heat losses due to the presence of CO\(_2\). They reported that CO\(_2\)-diluted flames have much higher
Fig. 3. Variations of the maximum Bradley’s Karlovitz number and laminar burning velocities with the equivalence ratio, showing the accessible domain when the maximum $f = 170$ Hz is operated. (a) CH$_4$/air mixtures; (b) CH$_4$ diluted with 20%–60% N$_2$; (c) CH$_4$ diluted with 20%–60% CO$_2$; and (d) combined plots of these maximum-$K$ ($f_{\text{max}} = 170$ Hz) lines from (a–c) for comparison.

$HL_c$, up to four times more, than N$_2$-diluted flames [24]. For clarity, the accessible/inaccessible domains for CH$_4$/air, CH$_4$/N$_2$/air, and CH$_4$/CO$_2$/air flames, respectively from Fig. 3a–c, are plotted together on Fig. 3d (values of $K_{\text{max}}$ at both leanest and richest sides of these diluted flames are also marked). For example, at $\phi = 0.6$, values of $K_{\text{max}}$ increase from about 9.5 to 50.3 when CH$_4$ fuel is diluted with 60% CO$_2$ (Fig. 3d). Note that $K \sim 50.3$ is equivalent to $Ka \sim 520.4$, because $Ka = 6.37 K$. As can be seen from Fig. 3d, the accessible domain for CH$_4$/CO$_2$/air flames are significantly expanded.

**Global Quenching Regime and Its Anticipated Curve**

The central idea of this work is that global quenching of premixed flame is characterized by turbulent straining (the effect of $K$), equivalence ratio ($\phi$), and heat loss effects. The latter may be determined by comparing the behavior of the N$_2$- and CO$_2$-diluted flames of the same $S_L$. Here attempts are made in determining regimes of global quenching on these $K$-$\phi$ plots (Fig. 3a–c) for CH$_4$/air, CH$_4$/N$_2$/air, and CH$_4$/CO$_2$/air flames, respectively. At any given $\phi$, hundreds of experiments with the same mixtures but different values of $K$, which can be increased from 0 to $K_{\text{max}}$ ($f = 170$ Hz), are conducted. Thus, values of $K_c$ for global quenching of these premixed flames may be obtained. Fig. 4a–c presents variations of $K_c$ with $\phi$ on these accessible domains, respectively, for CH$_4$/air, CH$_4$/N$_2$/air, and CH$_4$/CO$_2$/air flames. Only a limited range of $\phi$ on both lean and rich ends can be scrutinized to identify $K_c$, because we cannot study the inaccessible domains where $K_c \gg K_{\text{max}}$.

For pure CH$_4$/air flames (Fig. 4a), it is much harder to globally quench very lean flames ($\phi = 0.6; K_c \gg 6.22$) than very rich flames ($\phi = 1.45; K_c > 1.54$). When $\phi$ is only slightly increased from 0.6 to 0.62, there is absolutely no sign of global quenching of lean CH$_4$/air flames even as $K_{\text{max}} = 7.7$ (Fig. 4a). These no quench data points, as overlapped by the symbol X on the $K_{\text{max}}$ lines, are also plotted in Fig. 4a–c. For rich CH$_4$/air flames, when $\phi$ is decreasing from 1.45 to about 1.38, the corresponding values of $K_c$ increase from 1.54 to 2.11. Hence, the global quenching regimes, as indicated by the shaded areas with dashed grid lines, can be determined.

Similar to Fig. 4a, global quenching regimes and their anticipated curves on the $K_c$-$\phi$ plot for N$_2$- and CO$_2$-diluted flames are presented in Fig. 4b and c, respectively. It becomes clear that values of $K_c$ are very sensitive to $\phi$, since $K_c$ increases drastically as $\phi$ gradually approaches 1 from either lean or rich sides. For highly diluted flames, the leanest and richest values of $\phi$ that can be conducted in the cru-
Fig. 4. (a–c) Similar to Fig. 3a–c for the accessible domains, but showed the critical values of Bradley’s Karlovitz number for global quenching as a function of the equivalence ratio. (d) Values of $K_c$ plotted against $\varphi$ for CH$_4$/air, CH$_4$/N$_2$/air, and CH$_4$/CO$_2$/air flames, respectively, where the solid lines are real quenching lines obtained from the actual data points from (a–c) and the dashed lines are the anticipated quenching lines.

ciform burner are shrunk. For instance, values of $\varphi$ vary from 0.6 to 0.72 for the leanest mixtures and from 1.45 to 1.25 for the richest mixtures, as CH$_4$ fuel is diluted with 60% CO$_2$ gases. The global quenching boundary for 60% CO$_2$-diluted flames, consisting of both real data points (the solid line) and the anticipated curve (the dash line) as shown on Fig. 4c, suggests a complete variation of $K_c$ with $\varphi$, where the maximum $K_c$ is assumed to occur near $\varphi = 1$. Based on the same trend, we predict the anticipated curves for both pure CH$_4$/air flames and 60% N$_2$-diluted flames. These results with both real (solid lines from Fig. 4a–c) and anticipated (dash lines) data are plotted on Fig. 4d for comparison. Assuming that these anticipated curves are correct, the required $K_c$ for global quenching of stoichiometric CH$_4$/air flames (little heat losses) must be as much as 25 ($K_c \approx 160$; see Fig. 4d). This result ($K_c \approx 160$) seems to support the prediction by Peters [12]. He proposed a kinetic foundation of thermal flame theory, in which a premixed CH$_4$/air flame consisted of a chemically inert preheat zone, a chemically reacting inner layer, and an oxidation layer [25]. Peters argued that turbulence could only broaden the preheat zone without much thickening the inner layer until, ultimately, very high rates of strain ($K_c \sim 100$) are achieved, leading to flame quenching [12]. Thus, the vitality of premixed turbulent CH$_4$/air flames is very impressive.

### Positive Stretching and Radiative Heat Loss

The experimental evidence shows that lean CH$_4$/air flames are much more difficult to quench globally by turbulence than rich CH$_4$/air flames. This can be understood from the behavior of the inner layer that is responsible for the fuel consumption and thus keeping the reaction process alive. For rich CH$_4$/air flames, the oxidation layer disappears and only one reaction layer (the inner layer) exists. On the other hand, both the inner layer and the oxidation layer coexist in lean CH$_4$/air flames. It is thought that one layer is easier to disrupt than two layers if turbulence eddies are strong enough to penetrate into the inner layer. As the fuel is consumed in the inner layer, the radicals are depleted by chain-breaking reactions. Moreover, the rate-determining reaction in the inner layer is very sensitive to the presence of H radicals [25], and the depletion of H radicals is much more rapidly in rich CH$_4$/air flames than in lean CH$_4$/air flames [26,27]. Thus, rich CH$_4$/air flames are more vulnerable to quench by turbulence than lean CH$_4$/air flames. This result reveals that premixed turbulent flames may be statistically dominated by positive stretching in a global (not local) sense.

Figure 5 presents the effect of radiative heat loss on global quenching of premixed CH$_4$/air flames. By comparing the behavior of N$_2$- and CO$_2$-diluted flames of the same $S_L \approx 10$ cm/s, it is found that
the radiative heat loss has an important influence on global quenching of lean CH$_4$/air flames. At $\phi \approx 0.64$, $K_\phi$ is about 1.3$^{278}$ for N$_2$-diluted flames, while $K_\phi = 4.1$ for CO$_2$-diluted flames (Fig. 5); the larger the radiative heat loss, the smaller the value of $K_\phi$. For lean CH$_4$/air mixtures with constant $S_L$, values of $K_\phi$ increase with $\phi$ for both N$_2$- and CO$_2$-diluted flames, in which the difference in values of $K_\phi$ between these two flames also increases with $\phi$. On the other hand, global quenching of rich CH$_4$/air flames is not much influenced by the radiative heat loss, because $K_\phi \approx 1.32$ for both N$_2$- and CO$_2$-diluted flames (Fig. 5).

Conclusions

This study contributes to our understanding of global quenching processes of premixed turbulent CH$_4$/air flames that may be characterized by multifarious interactions among turbulent straining, flame chemistry, and heat losses. The present measurements reveal the following.

1. The condition for global quenching of premixed CH$_4$/air flames can be determined by a critical turbulent Karlovitz number and the equivalence ratio. Using the cruciform burner, global quenching regimes for CH$_4$/air and CH$_4$/diluent/air flames are identified for the first time on $K_\phi$-$\phi$ plots.

2. Lean CH$_4$/air flames are much harder to quench globally by turbulence than rich CH$_4$/air flames, suggesting that turbulent premixed flames may be statistically dominated by positive stretching in a global sense. Values of $K_\phi$ increase largely as $\phi$ gradually approaches 1 from either lean or rich sides, with the maximum $K_\phi$ occurring possibly near $\phi = 1$. The vitality of premixed turbulent CH$_4$/air flames at $\phi = 1$ is astonished, because the anticipated value of $K_\phi$ is as high as 25 or $K_\phi \approx 160$ for the occurrence of global quenching.

3. Global quenching of lean/rich CH$_4$/diluent/air flames is not influenced by the radiative heat loss, respectively. For lean mixtures, the smaller the radiative heat loss, the larger the values of $K_\phi$. Specifically, values of $K_\phi$ increase from 4.1/3.2 to 5.1/4.3 as $\phi$ increases from 0.64 to about 0.7–0.8 for N$_2$-/CO$_2$-diluted flames with $S_L \approx 10$ cm/s, respectively. In contrast, $K_\phi$ remains nearly constant at a value of about 1.5 for both rich N$_2$- and CO$_2$-diluted flames with $S_L \approx 10$ cm/s in the range of $\phi \approx 1.20$–1.45.

These results are important to turbulent premixed combustion and may be relevant to gasoline engines and atmospheric explosions.

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REFERENCES

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COMMENTS

Kazuhiro Yamamoto, Toyohashi University of Technology, Japan. You added CO2 to the fuel to estimate the radiation effect, compared with the case of N2 addition. Since the heat capacity of CO2 is much higher than that of N2, the flame temperature is much lower even if the radiation heat loss is not considered. It cannot be concluded that the unstable behavior is caused only by increased radiation effect in the case of CO2 addition. Is it possible to compare two mixtures with the same heat capacity to clarify the radiation effect?

Author’s Reply. CO2-diluted flames experience more heat losses than N2-diluted flames because of a higher concentration of CO2 in the products and because the two diluent gases have different heat capacity (e.g., specific heat @1200 K, C\text{pCO2} = 56.2, C\text{pN2} = 33.7, and C\text{pH2O} = 43.9 kJ/kmol K). This is the reason why we kept S\text{L} constant (~10 cm/s) for both N2- and CO2-diluted flames, thus enabling us to make a detailed comparison of the flame response to turbulent straining (the effect of critical Bradleys’s Karlovitz number, K\text{c} for global quenching) and equivalence ratio (\varphi). As shown in Fig. 5, by comparing N2- and CO2-diluted flames of the same S\text{L}, it is found that global quenching of lean/rich CH4 flames is/is not influenced by the radiative heat loss, respectively. In addition, we have estimated average values of C\text{p} and adiabatic flame temperature T\text{ad} for both N2- and CO2-diluted flames, and these values of C\text{p} and T\text{ad} for both N2- and CO2-diluted flames are very close at a given \varphi with S\text{L} ~ 10 cm/s (Fig. 5).

The vitality of premixed turbulent CH4/air flames is very impressive. It is anticipated that K\text{c} has to be as much as 25 (\varphi/0.5 \text{Re}^{0.5} \text{St}) in order to quench pure stoichiometric CH4/air flames globally. Even for CO2-diluted flames with large radiative heat loss, the required K\text{c} for global quenching is greater than unity (K\text{c} > 6.37 because K\text{c} \sim \text{Re}^{0.5} \text{St}) as shown in Figs. 4 and 5. To the author’s best knowledge, this is the first experiment to determine the effect of radiative heat loss on global quenching of turbulent CH4 flames, and the conclusion is carefully verified.