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Turbulent Burning Velocities of Premixed CH₄/Diluent/Air Flames in Intense Isotropic Turbulence with Consideration of Radiation Losses

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This paper presents turbulent burning velocities, \( S_T \), of several premixed CH₄/diluent/air flames at the same laminar burning velocity \( S_L = 0.1 \) m/s for two equivalence ratios \( \phi = 0.7 \) and 1.4 near flammability limits with consideration of radiation heat losses from small (N₂-diluted) to large (CO₂-diluted). Experiments are carried out in a cruciform burner, in which the long vertical vessel can be used to provide a downward propagating premixed flame and the large horizontal vessel equipped with a pair of counter-rotating fans and perforated plates can be used to generate an intense isotropic turbulence in the central region between the two perforated plates. These turbulent flames, upon propagation, experience small downward/upward gas velocities near the top/bottom of the central turbulence region in the burner. The magnitudes of small downward and upward gas velocities are essentially equal, less than 5 ~ 10 % of turbulent flame speeds depending on turbulent intensities \( u'/S_L \), as confirmed by four different arrangements of pairs of ion-probe sensors in the burner. Hence, the effect of gas velocity on \( S_T \) measured in the central region can be neglected. Simultaneous measurements using the pressure transducer and ion-probe sensors show that the pressure rise due to turbulent burning has little influence on \( S_T \). These measurements prove the accuracy of the \( S_T \) data. At \( \phi = 0.7 \), the percentage of \( [(S_T/S_L)_{CO2} - (S_T/S_L)_{N2}]/(S_T/S_L)_{N2} \) decreases gradually from -4% to -17% when values of \( u'/S_L \) increase from 4 to 46, while at \( \phi = 1.4 \) such decrease is much more abrupt from -19% to -53% when values of \( u'/S_L \) only increase from 4 to 18. The larger the radiation losses, the smaller values of \( S_T \); this decrease effect is augmented by increasing \( u'/S_L \) and is particularly pronounced for rich CH₄ flames. When \( u'/S_L = 18 \), lean CO₂ and/or N₂ diluted CH₄ flames have much higher, 3.6 and/or 1.8 times higher, values of \( S_T/S_L \) than rich CO₂ and/or N₂ diluted CH₄ flames, respectively. It is found that lean (\( \phi = 0.7 \)) CH₄ flames are very difficult to quench globally even when methane mixtures are diluted with 41 % CO₂ having large radiation losses. Measurements of chemiluminescence intensities for these turbulent propagating CH₄/diluent/air flames using the photomultiplier are carried out. The peak light intensities of CH* and C₂* emitters are found to be qualitatively correlated with these aforementioned \( S_T \) data of lean and rich turbulent CH₄/diluent/air propagating flames, respectively. Finally, the scatter plot of \( S_T/S_L \) as a function of \( u'/S_L \) for the aforementioned N₂- and CO₂-diluted CH₄ flames can be well compressed and approximated by a general correlation of turbulent burning velocities in a form of \( (S_T - S_L)/u' = 0.06Da^{-0.58} \), where \( Da \) is the turbulent Damköhler number.

Key words: turbulent burning velocities, radiation heat loss, gas velocities and pressure rise, ion-probe sensors, turbulent premixed combustion

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1. Introduction

This is the third paper emanating from a series of experiments [1,2] designed to measure quantitatively turbulent burning velocities ($S_T$) that influence virtually all important properties of premixed turbulent flames [3]. The first article [1] described the generation of intense near-isotropic turbulence in a cruciform burner and presented a pair of specially-designed ion-probe sensors for global $S_T$ measurements of methane-air mixtures. It also discussed the effect of the energy-weighted rms turbulence intensity ($u'$) on $S_T$ for CH$_4$-air premixed flames. The second paper [2] reported measurements of global $S_T$ for both CH$_4$- and C$_3$H$_8$-air mixtures over a greater parameter range than hitherto measured. These turbulent burning rates ($S_T/S_L$) were then compared with earlier results using different burners [4-6], particularly with the explosion bomb measurements by Bradley’s group [4,7], where $S_L$ was the laminar burning velocity.

In brief review, the cruciform burner features a long vertical cylindrical vessel and a large horizontal cylindrical vessel. The former was used to provide a downward-propagating premixed flame with large surface area of about 10 cm in diameter. The latter was equipped with a pair of counter-rotating fans and perforated plates at the two ends to generate a region of intense near-isotropic turbulence for flame-turbulence interactions that are unaffected by ignition and unwanted turbulence from walls. This novel experimental design allows direct imaging and velocity measurements of the two-way interaction between premixed flames and homogeneous isotropic turbulence. Consequently, high-speed laser tomography, ion-probe sensors, particle imaging velocimetry measurements can be used in this configuration to develop a qualitative understanding and a quantitative analysis of various aspects of turbulent flame propagation, stretching, and global quenching [1-2,8-10]. In and by itself, we believe that the experimental design is a useful contribution to the study of premixed turbulent combustion.

The maximum fan-stirred frequency ($f$) used in [2] was 170 Hz, corresponding to $u' \approx 7.85$ m/s and $Re_T = u' L_i / \nu \approx 24,850$ [8], where $L_i$ was the turbulent integral length scale estimated from Taylor’s hypothesis and Bradley’s correlation for zero mean velocities [1,7] and $\nu$ the kinematic viscosity of reactants. We have measured $S_T/S_L$ for CH$_4$- and C$_3$H$_8$-air mixtures over wide ranges of $u'$ up to 50 with equivalence ratios ranging from $\phi = 0.70 - 1.4$ for CH$_4$ mixtures and $\phi = 0.65 - 1.6$ for C$_3$H$_8$ mixtures using a pair of specially-designed ion-probe sensors, among several other methods [1,11]. These data can be fitted into a general correlation of the form $(S_T - S_L)/u' = 0.06 Da^{0.58}$, where $Da$ was the turbulent Damköhler number, defined as $Da = Re_T (u' L_i)^2$. This correlation, covering both corrugated flamelet (large $Da$) and distributed reaction (small $Da$) regimes, is found to be better than previous correlations. The reader is directed to Ref. [2] for details.

The difficulties in obtaining accurate measurements of $S_T$ should not be underestimated, because thermal expansion and heat losses at the flame front may induce both global and local
variations of the turbulent flow field. Some averaging procedures must be applied to estimate a global $S_T$. These experimental data commonly feature a large scatter even at a fixed $u'/S_L$ for a fixed $\phi$ of a given mixture in the same flow configuration [1,2,4-6]. How to model turbulent combustion for wide ranges of temporal and spatial scales of both flow and chemistry at high Reynolds numbers remains a difficult challenge [3]. Though we have put a great effort to measure quantitatively $S_T$ [1,2], some important questions still remain to be answered. Do the ion-probe sensors actually measure turbulent burning velocities? What is the effect of pressure rise due to turbulent burning in the cruciform burner on $S_T$ measurements? The first objective of this paper is to address these two questions by quantitatively evaluating the accuracy of $S_T$ measurements using the two ion-probe sensors and the pressure transducers at many different locations of the burner.

The second but equally important objective is to investigate the effect of radiation heat losses on turbulent burning velocities for the first time. There are many studies on the effect of radiation heat losses in laminar premixed flames. For instances, Law and his co-workers [12] measured values of $S_L$ for CH$_4$/CO$_2$/O$_2$ mixtures at various equivalence ratios, and some chemical effects of CO$_2$ on $S_L$ of premixed methane flames were studied numerically [13,14]. These results indicated that the radiation heat losses effect of CO$_2$ can significantly reduce values of $S_L$. Samaniego et al. [15,16] also measured flame temperature and CO$_2$*-chemiluminescence of premixed flames to investigate the effect of radiation heat losses, where the superscript * refers to the electronically-excited molecule. They found that CO$_2$-diluted flames experience more losses, up to four times more, than N$_2$-diluted flames. Furthermore, Candel and his co-workers [17] found that the global heat release rate of an acoustically excited premixed laminar-jet flame impinging on a water-cooled plate is proportional to chemiluminescence lights of radical emissions detected by a photomultiplier (PM). On the other hand, very few studies are available on the effect of radiation losses on turbulent burning velocities at high Reynolds numbers. This motivates the present experiments using CH$_4$/diluent/air mixtures near flammability limits at which these premixed flames are more sensitive to radiation losses. Both lean ($\phi = 0.7$) and rich ($\phi = 1.4$) CH$_4$/diluent/air flames at high Reynolds numbers are envisaged over a wide range of turbulent intensities. We use N$_2$ and CO$_2$ gases, as diluents, to alter the degree of radiation losses from small to large, while keeping $S_L \approx 0.1$ m/s for all CH$_4$/diluent/air flames for comparison. Note that CO$_2$-diluted flames experience greater radiation losses than N$_2$-diluted flames because heat losses occur by radiation from soot and emitting species, mainly CO$_2$ and/or H$_2$O, present in the burnt gases [16]. In this study we consider radiation heat losses and do not include conductive heat losses to walls, since the present measurements of these transient turbulent premixed flames are obtained in the central uniform region that is far away from the vicinity of walls.

By comparing values of $S_T/S_L$ between N$_2$ and CO$_2$ diluted flames of the same $S_L$ at fixed values of $u'/S_L$ and $\phi$, the effect of radiation losses on $S_T/S_L$ may be identified. It is worth noting that the importance of radiation losses on turbulent flame extinction has been anticipated by Sivashinsky [18], Poinso et al. [19], Shy et al. [20], among others. Therefore, we will also discuss the effect of radiation losses on global quenching for these CH$_4$/diluent/air turbulent flames at both lean and rich
conditions near flammability limits. Finally, measurements of chemiluminescence intensities of these propagating CH₄/diluent/air turbulent flames are carried out using the PM tube with several different narrowband optical filters. It will be shown that the peak light intensities of CH⁺ and C₂⁺ emitters are qualitatively correlated with values of $S_T/S_L$ for lean and rich CH₄/diluent/air flames, respectively.

The following section reviews experimental methods, including descriptions of (1) the apparatus and the near-isotropic turbulence, (2) measurements of turbulent flame speeds and gas velocities using ion-probe sensors, (3) pressure measurements, and (4) considerations of radiation losses and chemiluminescence light intensities measurements. These measurement results are then employed to analyze effects of gas velocities, pressure rise, and radiation losses on turbulent burning velocities as well as on global quenching of propagating turbulent premixed methane flames.

2. Experimental Methods

2.1. Cruciform burner and intense near-isotropic turbulence

Figure 1 shows schematic diagrams of the burner with a pair of ion-probe sensors positioned at several different positions with respect to the central region for $S_T$ measurements. In addition, ten pressure release valves are distributed to both the vertical and horizontal vessels for modulating the pressure rise in the chamber during turbulent combustion. The burner and its corresponding flow velocities and statistics have been reported [1,2], and recently we applied a high-speed particle imaging velocimetry and the wavelet analysis to further investigate small-scale intermittency of fan-stirred flow turbulence [21]. For completeness, here we briefly summarize these results and describe the new modification that we employ for chemiluminescence light measurements. A large region of nearly stationary homogeneous turbulence at high Reynolds number can be generated with roughly equal rms turbulent intensities in all three directions, negligible mean velocities, values of skewness and flatness being nearly zero and three, and energy spectra with a $-5/3$ slope [1,21]. It was found that the rms velocity fluctuations in both non-reacting and reacting cases are very close, especially when turbulence is intense. This large uniform region of turbulence may be up to 15x15x15 cm$^3$, which contains many large eddies allowing multiple interactions with propagating premixed flames. Consequently, these propagating turbulent flames can be free from the influences of ‘unsteadiness’ and large-scale dynamics. Before a run, the burner is evacuated and then filled with pure CH₄/air or CH₄/diluent/air mixtures at a selected $\phi$ at one atmosphere. A run begins by ignition on the top of the vertical vessel, generating a downward propagating premixed flame with a surface area of about 10 cm in diameter (Fig. 1a). This flame then enters the central region where interactions with turbulence take place without the influence from ignition, as demonstrated by a typical raw image directly taken from a high-speed CCD camera as shown in the middle of Fig. 1a.

2.2. Ion-probe sensor, turbulent flame speeds, and gas velocities

Two sets of ion-collecting electrodes supported by the bottom plate of the burner are used for $S_T$ measurements, see Fig. 1a. The ion-probe sensor is made of thin, long platinum wires, 0.2 mm in
diameter and about 5 cm for the exposed length, forming a circuit loop that is carefully shielded and supported by the two ceramic rods and stainless steel tubes, see the top left of Fig. 1a. The sensor can detect the abnormal ionization existing in the flame, corresponding to concentrations of ions in the reaction zone which are several orders greater than that in burned products [22,23]. These ion currents were converted into voltage signals via a 450 kΩ resistor and recorded at a sampling rate of 10 kHz. Figure 1b displays a typical example of the time evolution of voltage signals measured by the two sets of ion-probe sensors which are positioned respectively at levels ‘a’ and ‘e’ separated by 20 cm (these devices constitute the measurement I in Fig. 1a). The top sensor (channel 1 at level ‘a’) detects a spike signal, indicating the presence of a very thin, flat premixed flame. As the flame propagates downwards, it enters the central turbulence region and passes through the bottom sensor at level ‘e’, which features a short tail of the signal observed in channel 2 due to turbulent wrinkling. Hence, the turbulent burning velocity can be determined without ambiguity using the separation (0.2 m for measurement I) divided by the time between the two peaks (Δt = 0.153 s), yielding $S_T = 1.31 \text{ m/s}$.

One may ask if the ion-probe sensors actually measure the turbulent burning velocity. To verify this point, four different arrangements of the two ion-probe sensors were investigated. As indicated in Fig. 1a, measurements (I), (II), (III), and (IV) correspond to levels ‘a-e’ (20 cm apart), ‘a-c’ (10 cm), ‘c-e’ (10 cm), and ‘b-d’ (10 cm), respectively. Measurements of the turbulent flame speeds for these four different arrangements, respectively denoted as $S_{I,a-e}$, $S_{II,a-c}$, $S_{III,c-e}$, and $S_{IV,b-d}$, are carried out systematically under the same experimental conditions (fixed $\phi$, $S_L$, and $u'$). These four values obtained from different measuring positions will be compared to identify whether there are gas velocities ahead of propagating turbulent flames and thus evaluate the $S_T$ measurement accuracy, as discussed in the next section.

2.3. Pressure measurements

Concerning the pressure effect, as many as ten large pressure release valves are used to modulate the pressure rise during turbulent combustion in the burner, including four valves on the top of the burner, two on the sides of the horizontal vessel, and additional four valves on the front and back windows of the central region (see Fig. 1a). Three cases, with four, six and ten pressure release valves opened during turbulent combustion, are carried out independently to evaluate the pressure effect on $S_T/S_L$ over a wide range of $u'/S_L$. For each case of CH₄-dry air mixtures at a fixed value of $\phi$, we simultaneously measure the evolution of pressure due to turbulent combustion and turbulent burning velocities ($S_{I,a-e}$; measurement I, see Fig. 1a) as a function of the fan frequency ranging from 0 to 127 Hz. Thus, the effect of pressure rise on $S_T$ measurements in a confined chamber can be investigated.

2.4. Radiation heat loss and chemiluminescence considerations

Different percentages of N₂ and CO₂ dilution gases are used to alter the degree of radiation
heat losses from small to large. The laminar burning velocity is kept constant ($S_L \approx 0.1$ m/s) for all CH$_4$/diluent/air flames. Table 1 lists the corresponding thermochemistry properties for both lean ($\phi = 0.7$) and rich ($\phi = 1.4$) cases without and/or with N$_2$ and CO$_2$ diluents, including values of $S_L$, adiabatic flame temperature $T_{ad}$, and specific heat coefficients for reactants and products ($C_{p,react.}$ and $C_{p,prod.}$). It should be noted that values of $S_L$ with various percentages of diluents in Table 1 are obtained from direct measurements in the present burner using ion-probe sensors. The different percentages of diluents can be also obtained from Stone et al. [24]. Values of $T_{ad}$ and $C_p$ in Table 1 are obtained from computer codes of Turans [25], among others. Samaniego & Mantel [16] proposed a heat loss coefficient ($HL$), a ratio of the energy radiated in the flame zone to chemical energy release, to quantify radiation losses due to the presence of CO$_2$. These authors indicated that CO$_2$-diluted flames have much higher $HL$, up to four times that of N$_2$-diluted flames. It is thus possible to examine the effect of radiation losses on turbulent burning velocities of methane flames at a constant $S_L = 0.1$ m/s for both lean and rich cases near flammability limits.

Concerning chemiluminescence measurements, Kojima et al. [26] showed that chemiluminescence lights in the visible and UV spectra of hydrocarbon flames mainly originate from four natural emitters, CH$^*$, C$_2^*$, OH$,^*$, and CO$_2^*$. Figure 2a presents these emission intensities as a function of wavelength for laminar premixed lean ($\phi = 0.9$) and rich ($\phi = 1.4$) CH$_4$-air flames [26]. In the lean case, the main chemiluminescence spectrum is from CH$^*$ at the wavelength 431.5 nm, while C$_2^*$ at 516.5 nm contributes most significant emission intensities for rich CH$_4$-air laminar flames [26]. Light emission is detected by PM tubes equipped with different narrowband optical filters, respectively 310 ± 10 nm, 430 ± 10 nm, and 514.5 ± 10 nm for OH$^*$, CH$^*$, and C$_2^*$ emissions, as in Ref. [17]. The PM tube and the selected optical filter are closely clung to a small hole of only 3 mm in diameter on a black stainless steel plate, as shown on Fig. 1a. The corresponding light emission data are then recorded by a 330 kHz acquisition board (Advanced Technology, PCL-1800, Taiwan) operating at a sampling frequency of 5 kHz. Figure 2b shows a typical example of the time duration of the light intensities measured by the PM tube without any optical filters (all wavelength) for lean CH$_4$/CO$_2$/air flames at $\phi = 0.7$ with $S_L = 0.1$ m/s, where both laminar ($f = 0$ Hz; $u'/S_L = 0$) and turbulent ($f = 10$ Hz; $u'/S_L = 4.62$) cases are presented. Note that for the purpose of comparison, the peak intensities for both laminar and turbulent cases are shifted to 1 s, as indicated by the dashed line at 1 s in Fig. 2b. It can be seen that the peak light intensity in the turbulent case is higher, by about 1.5 times, than that detected in the laminar case. Also, the time duration of the turbulent case at $u'/S_L = 4.62$ is only 0.262 s, much shorter than 0.708 s in the laminar case (Fig. 2b). This is because turbulence increases the burning rate to be well above its laminar burning velocity. One finds for example $S_T/S_L \approx 3.75$ for $u'/S_L = 4.62$ in the case presented in Fig. 2b. The chemiluminescence light intensities obtained at different wavelengths will be compared for N$_2$ and CO$_2$ diluted flames of the same $S_L$ at fixed values of $\phi$ and $u'/S_L$. 
3. Results and Discussion

3.1. Effect of mean gas velocities

It is important to measure a possible mean gas motion ahead of the propagating flame. The best way to verify and address this question is to use high-speed digital particle image velocimetry (DPIV) [21]. Systematic tests indicate that good image quality can be obtained for DPIV processing if the cutoff fan frequency is less than 15 Hz [21] at which $u' \approx 4.62 f < 70 \text{ cm/s}$, which greatly limits the practical use of the setup. For higher values of $f$, up to 127 Hz corresponding to $u'/S_L \approx 50$ [1,2] as needed in the present study, an alternative method must be used. Pairs of ion-probe sensors are used to measure propagating turbulent flame speeds at different vertical portions near the intense near-isotropic region, from upper ($S_{II,a-c}$; measurement II) to central ($S_{I,a-e}$ and $S_{IV,b-d}$; measurements I and IV) to lower ($S_{III,c-e}$; measurement III) portions in the cruciform burner (see Fig. 1a). By carefully comparing these four measurements, gas velocities ahead of propagating turbulent flames can be extracted indirectly. Figure 3 presents these four turbulent flame speeds for CH$_4$-air mixtures at $\phi = 1$ with two different fan frequencies, (a) $f = 30 \text{ Hz}$ and (b) $f = 100 \text{ Hz}$, respectively. Each data point with the error bar shown in Fig. 3 is averaged from at least 5~8 runs under the same experimental conditions.

Figure 3 indicates that for $f = 30 \text{ Hz}$, the turbulent flame speeds are: $S_{II,a-c} \approx 2.82 \text{ m/s} > S_{I,a-e} \approx 2.54 \text{ m/s} > S_{IV,b-d} \approx 2.56 \text{ m/s} > S_{III,c-e} \approx 2.25 \text{ m/s}$, respectively. Similarly, $S_{II,a-c} \approx 4.61 \text{ m/s} > S_{I,a-e} \approx 3.97 \text{ m/s} > S_{IV,b-d} \approx 3.93 \text{ m/s} > S_{III,c-e} = 3.24 \text{ m/s}$ for $f = 100 \text{ Hz}$. Thus, the downward-propagating turbulent flame is slightly accelerating near the top portion of the central uniform region for which $S_{II,a-c}/S_{IV,b-d} = 1.10$ ($f = 30 \text{ Hz}$) and 1.17 (100 Hz), while the flame is slightly decelerating around the lower portion where $S_{III,c-e}/S_{IV,b-d} = 0.88$ ($f = 30 \text{ Hz}$) and 0.82 (100 Hz). There are small mean gas velocities ($S_g$) ahead of propagating turbulent flames near the upper and lower portions of the central region, where the upper $S_{g,u} = S_{II,a-c} - S_{IV,b-d}$ for flame acceleration near the upper portion and the lower $S_{g,l} = S_{IV,b-d} - S_{III,c-e}$ for flame deceleration at the lower portion, respectively. It is found that values of $S_{g,u}$ and $S_{g,l}$ are nearly the same over a wide range of $f$ from 10 Hz to 127 Hz and both values are no more than 5~10 % of turbulent flame speeds depending on turbulent intensities. It is also worth noting that the measurement I (a-e) is essentially equal to the measurement IV (b-d) within experimental uncertainties for which $S_{I,a-e} \approx S_{IV,b-d}$. When measurement (I) is used to measure $S_T$, mean gas velocities in the central region of the burner can be neglected. It is concluded that ion-probe sensors (measurement I) can correctly measure turbulent burning velocities, and that $S_{I,a-e}$ provides a good estimate of $S_T$.

3.2. Effect of pressure rise

To examine the pressure effect on $S_T$, tests are carried out by opening four, six, and ten pressure release valves during flame-turbulence interactions (see Fig. 1a). The evolution of pressure changes and turbulent burning velocities are recorded as a function of $f$ ranging from 0 to 127 Hz, for
methane-air mixtures at $\phi = 1$ using pressure transducers and ion-probe sensors. For simplicity, Fig. 4 displays data corresponding to four-valve and ten-valve opening. Each case includes six independent runs at $f = 0, 20, 40, 60, 80,$ and 100 Hz, respectively. When the propagating flame enters the central region, flame-turbulence interactions in the chamber cause a pressure increase due to thermal expansion, as can be clearly seen from Fig. 4. Here we shift the times at which the peak pressure occurs at various $f$ to $t = 1$ s for comparison. It is found that this pressure rise increases with $f$. For the case of 4-valve opening, values of the peak pressure increase from about 1.5 atm to 4.6 atm as $f$ increases from 0 Hz to 100 Hz. The pressure rise corresponding to an increased fan frequency is reduced in the case of 10-valve opening. There is no pressure rise at $f = 0$ Hz and the peak pressure only increases to 3.4 atm at $f = 100$ Hz.

By comparing values of $S_T$ measured by opening 4 valves with that when all 10 valves are opened using ion-probe sensors, it is found that $S_{T, 4V}/S_{T, 10V} = 1.02 \pm 0.05$ for stoichiometric CH$_4$-air flames over a wide range of $f$ varying from 0 Hz to 127 Hz. The burning velocities are found to be rather insensitive to pressure. This is because the reaction order of CH$_4$-air flames is close to 2. Similar results are also found for both lean and rich CH$_4$-air flames. Thus, it is concluded that the pressure rise in the burner has little influence on values of turbulent burning velocities, further validating the accuracy of the $S_T$ measurements.

3.3. Effect of radiation losses

The central idea of this study is that turbulent burning velocities are characterized by turbulent straining, equivalence ratio, and heat loss effects. The first two effects on $S_T$ have been examined [1,2] as a function of several non-dimensional parameters, such as $u'/S_L$, $\phi$, $Re_T$, $Da$, and a Bradley’s Karlovitz number $K = 0.157\left(\frac{u'}{S_L}\right)^2/(Re_T)^{0.5} = 0.157Ka$ [7] where $Ka$ is the commonly-used turbulent Karlovitz number. The effect of heat losses is here envisaged by comparing values of $S_T$ corresponding to N$_2$ and CO$_2$ diluted flames under the same experimental conditions.

Figures 5a and 5b show the effect of radiation heat losses on $S_T/S_L$ of CH$_4$/diluent/air flames over a wide range of $u'/S_L$ for both lean ($\phi = 0.7$) and rich ($\phi = 1.4$) cases. Note again that all diluted flames have the same $S_L = 0.1$ m/s and CO$_2$-diluted flames have a much higher degree of radiation losses than that of N$_2$-diluted flames. Figure 5 also includes previous data for $S_T/S_L$ as a function of $u'/S_L$ for pure CH$_4$/air flames without diluents at $\phi = 0.7$ and $\phi = 1.4$ [1], where $S_L$ used for normalization is from Vagelopoulos et al. [27]. Each of the data points presented in Figs. 5a and 5b is averaged over several runs with error bars, similar to those given in Fig. 3. These data show a strong bending effect on the slopes of $S_T/S_L$ vs. $u'/S_L$ plots. This phenomenon is even more pronounced for N$_2$ and CO$_2$ diluted CH$_4$ flames, particularly for the rich case ($\phi = 1.4$). At modest turbulent intensities for which $u'/S_L < 10$ at $\phi = 0.7$ (Fig. 5a), values of $S_T/S_L$ are not much different, slightly decreasing with increasing radiation losses from pure methane to 56 % N$_2$ to 41 % CO$_2$ diluents. This decrease due to radiation losses is then gently enhanced as $u'/S_L$ increases above 10,
where the bending effect becomes strong (Fig. 6a). The maximum values of $S_T/S_L$ of lean N$_2$ and CO$_2$ diluted flames at $\phi = 0.7$ are 10.2 and 9.5 at $u'/S_L \approx 28$ (Fig. 5a), whereas values of $(S_T/S_L)_{max}$ for rich N$_2$ and CO$_2$ diluted flames at $\phi = 1.4$ are only 5.2 and 3.0 at $u'/S_L \approx 12$, respectively (Fig. 5b). Further increasing turbulent intensities above some critical values, depending on $\phi$, $K$ and radiation losses, can actually lead to a decrease of turbulent burning rates. Similar results without considering radiation losses were also reported by Bradley [7] and Nakahara & Kido [28].

The propagation of lean ($\phi = 0.7$) turbulent CH$_4$/diluent/air flames continues without the occurrence of global quenching, even when the fuel is diluted with large amounts of CO$_2$ gases (41%; see Table 1) and turbulence is very intense where $f = 100$ Hz, $u'/S_L = 46$, and $K \approx 2.75$ (or equivalent to $Ka \approx 17.5$) at which $S_T/S_L \approx 8.2$ (Fig. 5a). In order to globally quench such a lean CO$_2$-diluted CH$_4$ flame of $S_L \approx 0.1$ m/s with large radiation losses, the critical value of $K$ has to be as large as 3.7 (or $Ka_c > 23.6$), corresponding to $f \approx 120$ Hz and $u'/S_L \approx 55$, as indicated by the symbol X in Fig. 5a. The occurrence of global quenching can be determined unambiguously in the burner because high-speed camcorders can readily discern whether or not flames propagate through the central turbulence region. The value of $K_c$ for global quenching of lean N$_2$-diluted CH$_4$ flames with small radiation losses is even higher ($K_c \approx 4.8$), revealing a sensitive influence of radiation losses on global quenching of lean premixed CH$_4$ flames. The smaller the radiation losses, the higher the value of $K_c$ that is required to globally quench lean CH$_4$ flames. On the other hand, these rich N$_2$ and CO$_2$ diluted CH$_4$ flames of the same $S_L = 0.1$ m/s at $\phi = 1.4$ are more vulnerable to global quenching, where $K_c \approx 1.3$ or equivalently $Ka_c \approx 8.3$ corresponding to $f \approx 50$ Hz and $u'/S_L \approx 23$. This is why Fig. 5b only shows four available data at $f = 10, 20, 30$, and 40 Hz. Unlike turbulent burning rates of rich CH$_4$ flames which are sensitive to the degree of the radiation losses, global quenching of rich CH$_4$ flames near the flammability limit is insensitive to the radiation losses. The reader is directed to Ref. [10] for a complete global quenching boundary of premixed CH$_4$ flames based on a $K_c$ vs. $\phi$ plot.

By comparing values of $S_T/S_L$ between CO$_2$ and N$_2$ diluted CH$_4$ flames at both $\phi = 0.7$ and $\phi = 1.4$ cases (Fig. 5), it is found that the radiation losses effect has much stronger influence on $S_T/S_L$ of rich CH$_4$/diluent/air flames than on lean CH$_4$/diluent/air flames. As can be seen from Fig. 6, the percentage of $[(S_T/S_L)_{CO2} - (S_T/S_L)_{N2}]/(S_T/S_L)_{N2}$ decreases linearly from -4% to $-17\%$ at $\phi = 0.7$ and/or from $-19\%$ to $-53\%$ at $\phi = 1.4$ when $u'/S_L$ increases from 4 to 46 and/or 4 to 18, respectively. The larger the radiation losses is, the smaller values of $S_T/S_L$ are. This effect is enhanced by increasing $u'/S_L$ and is particularly pronounced for rich CH$_4$ diluted flames.

These experimental results may be understood by examining the structure of premixed CH$_4$/air flames: one may recall that such flames consist of a chemically inert preheat zone, a chemically reacting inner layer, and an oxidation layer [3,29]. The behavior of the inner layer is responsible for the fuel consumption that keeps the reaction process alive. For rich CH$_4$/air flames, only one
reacting inner layer exists without the oxidation layer, while both layers coexist for lean CH₄/air flames. The present data of $S_{f}/S_{L}$ reveal that one layer is easier to disrupt than two layers when turbulence is strong enough to influence the inner layer. Moreover, the radicals during the fuel consumption in the inner layer are depleted by chain-breaking reactions, and the rate-determining reaction in the inner layer is rather sensitive to the presence of H radicals. Since the depletion of H radicals is much faster in rich CH₄/air flames than in lean CH₄/air flames [30,31], the decrease in $S_{T}$ due to the radiation losses is more severe in rich flames than in lean flames, especially when turbulent intensities are large. The $S_{f}/S_{L}$ measurements of both lean and rich CH₄/diluted/air flames in Fig. 5 also indicate that turbulent flames are statistically dominated, in a global sense, by positive stretching that increases/decreases the reaction rate of diffusionally unstable/stable turbulent premixed flames, similar to what is found for laminar flames [32].

3.4. Measurements of chemiluminescence intensities

To estimate the radiation losses form the flames which are being compared, we use the PM tube with different narrow-band optical filters to measure chemiluminescence intensities of these N₂ and CO₂ diluted flames of the same $S_{L}$, $\phi$, and $u'$. On the one hand, a detailed analysis of the all wavelength and OH⁺ light signals does not provide correlations which could lead to an estimation of values of $S_{T}$. On the other hand, peak intensities of free radicals CH⁺ and/or C₂⁺ existing in the reaction zone seem to have some correlations with $S_{f}/S_{L}$ of lean and/or rich CH₄/diluent/air flames, respectively. This may be due to the fact that CH⁺ and/or C₂⁺ emissions are proportional to the global heat release rate $Q$ of lean and/or rich CH₄/air flames, as found by Kojima et. al. [26], Schuller et al. [17], among others. Therefore, peak CH⁺ and/or C₂⁺ light intensities may be used as an estimate of the radiation losses from these lean and/or rich CH₄/diluent/air flames.

As a typical example, Fig. 7 presents the time duration of chemiluminescence intensities from CH⁺ and C₂⁺ emitters for both N₂ and CO₂ diluted flames at $\phi = 0.7$ and 1.4, where all diluted flames have the same $S_{L} = 0.1$ m/s and $u'/S_{L} = 4.62$. For comparison, we shift the peak value of these chemiluminescence data to 0.5 s, and these light intensity data near the peak around 0.5 s with varying time durations are best fitted, as indicated by solid lines on Fig. 7. Other cases with higher values of $u'/S_{L}$ (not shown) are similar, except that the higher values of $u'/S_{L}$ are, the smaller the time duration is. Figure 8 shows peak values of CH⁺ and C₂⁺ chemiluminescence intensities, taken from these fitting curves in Fig. 7, as a function of $u'/S_{L}$, including both (a) lean ($\phi = 0.7$) and (b) rich ($\phi = 1.4$) CH₄/diluent/air flames of the same $S_{L} = 0.1$ m/s. Also plotted are mean turbulent burning rates without error bars from Fig. 5 for comparison. For the lean ($\phi = 0.7$) case, there are two points of similarity, in a qualitative sense, between peak CH⁺ light intensities and turbulent burning velocities. (1) Both peak CH⁺ light intensities and values of $S_{f}/S_{L}$ for N₂-diluted flames are higher than that of CO₂-diluted flames; (2) these values first increase with $u'/S_{L}$ up to 28 and then level off (Fig. 8a). For rich ($\phi = 1.4$) CH₄/diluent/air flames, the correlations between peak C₂⁺ intensities and turbulent burning velocities are more pronounced. It is worth noting again that the
present N\textsubscript{2} and CO\textsubscript{2} diluted flames used for comparison have the same \( \phi, S\textsubscript{L}, u' \), and with nearly the same \( T\textsubscript{ad} \) and \( C\textsubscript{p} \) (Table 1). Thus, the differences in peak chemiluminescence intensities and values of \( S\textsubscript{T}/S\textsubscript{L} \) between N\textsubscript{2} and CO\textsubscript{2} diluted flames are mainly due to different degrees of radiation losses existing in these two flames. The larger the losses, the smaller values of \( S\textsubscript{T}/S\textsubscript{L} \) and peak CH\textsuperscript{\*} (lean) or C\textsubscript{2}\textsuperscript{\*} (rich) intensities. In addition, it is found that rich N\textsubscript{2}-diluted flames have much higher peak C\textsubscript{2}\textsuperscript{\*} light intensities (more heat release) and thus much higher values of \( S\textsubscript{T}/S\textsubscript{L} \) than that of rich CO\textsubscript{2}-diluted flames, further revealing the strong influence of radiation losses to turbulent burning velocities of rich premixed CH\textsubscript{4} flames. From Fig. 8, one could estimate the percentage differences of peak light intensities between CO\textsubscript{2} and N\textsubscript{2} diluted flames using \( (I\textsubscript{CO\textsuperscript{2}} - I\textsubscript{N\textsubscript{2}})/I\textsubscript{N\textsubscript{2}} \) at both \( \phi = 0.7 \) and \( \phi = 1.4 \), where the peak intensity \( I = CH\textsuperscript{\*} \) at \( \phi = 0.7 \) and \( I = C\textsubscript{2}\textsuperscript{\*} \) at \( \phi = 1.4 \). One finds that the decreasing percentages of \( (I\textsubscript{CO\textsuperscript{2}} - I\textsubscript{N\textsubscript{2}})/I\textsubscript{N\textsubscript{2}} \) are nearly constant (nearly independent of \( u'/S\textsubscript{L} \)), only – 9 \% for the lean case (\( \phi = 0.7 \)) but as much as – 72 \% for the rich case (\( \phi = 1.4 \)). Alternatively, the ratios of peak light intensities \( I\textsubscript{N\textsubscript{2}}/I\textsubscript{CO\textsuperscript{2}} \) are found to be only about 1.1 for lean diluted flames (\( \phi = 0.7 \)) but up to 4 for rich diluted flames (\( \phi = 1.4 \)). Note again that the depletion of H radicals which dominates the rate-determining reaction in the inner layer is much swifter in rich CH\textsubscript{4}/air flames than in lean CH\textsubscript{4}/air flames. Therefore, the decrease in \( I\textsubscript{CO\textsuperscript{2}} \) due to the radiation losses is more severe in rich flames than in lean flames. These chemiluminescence results are consistent, in a qualitative sense, with the aforementioned \( S\textsubscript{T}/S\textsubscript{L} \) data (see Fig. 8). Nonetheless, because the transient nature of the turbulent propagating flames in the burner limits the available collecting time period of the PM tube, the peak light emission data should be viewed with some cautions.

3.5. General correlation

One may now examine whether a general correlation of turbulent burning velocities in a form of \( (S\textsubscript{T} - S\textsubscript{L})/u' = aDa^b \) where a and b are empirical constants can be derived under the consideration of radiation losses. Thus, values of \( (S\textsubscript{T} - S\textsubscript{L})/u' \) of the present CH\textsubscript{4}/diluent/air flames with consideration of radiation losses are plotted as a function of the turbulent Damköhler number, \( Da = \text{Re}_T(u'/S\textsubscript{L})^{-2} \), ranging from about 6 to 34, as shown in Fig. 9. Also plotted for comparison are previous data of pure methane-air flames [1], as in the annex of Fig. 9, where the range of \( Da \) is from 12 to 900 and the shaded area is the domain of the present data. Note that these scatter plot of \( S\textsubscript{T}/S\textsubscript{L} \) data as a function of \( u'/S\textsubscript{L} \) as shown in Fig. 5 for N\textsubscript{2} and CO\textsubscript{2} diluted flames can be compressed and well approximated by the general correlation, \( (S\textsubscript{T} - S\textsubscript{L})/u' = 0.06Da^{0.58} \). This correlation is represented by a solid line in Fig. 9 where the three error bars correspond to previous data ranges [1] presented in the annex of Fig. 9. In addition, the dashed line of the form \( (S\textsubscript{T} - S\textsubscript{L})/u' = 0.08Da^{0.5} \) in Fig. 9 corresponds to the so-called distributed reaction zone (DRZ) model [1] associated with disrupted flame structures as initially described by Damköhler [33]. The ratio \( S\textsubscript{T}/S\textsubscript{L} \) is then principally influenced by increased diffusive transport inside the broadened flame front without modification of the reaction rate. The general correlation, \( (S\textsubscript{T} - S\textsubscript{L})/u' = 0.06Da^{0.58} \), is a good approximation. However, values of \( (S\textsubscript{T} - S\textsubscript{L})/u' \) for rich CO\textsubscript{2}-diluted flames are smaller than those of rich N\textsubscript{2}-diluted flames at various values of \( Da \) due to the influence of radiation losses.
4. Conclusions

An experimental study of premixed CH₄/diluent/air flames in intense isotropic turbulence has been carried out to investigate effects of gas velocities, pressure rise, and radiation losses on turbulent burning velocities, $S_T$. The measurements and analyses reveal the following.

1. The accuracy of the $S_T$ measurements using ion-probe sensors is confirmed, because gas velocities in the burner are negligible and the pressure rise due to turbulent burning has little influence on $S_T/S_L$ over a wide range of $u'/S_L$.

2. Radiation losses have much larger influence on rich CH₄ flames than lean CH₄ flames. The larger the radiation losses, the smaller values of $S_T/S_L$. This influence is enhanced by increasing $u'/S_L$ and is particularly pronounced for rich CH₄ flames. At $u'/S_L = 18$, lean CO₂ and N₂ diluted CH₄ flames have higher, as much as 3.6 and 1.8 times higher, values of $S_T/S_L$ than rich CO₂ and N₂ diluted CH₄ flames, respectively.

3. From the present $S_T/S_L$ data of both lean and rich CH₄/diluted/air flames, turbulent premixed flames are statistically dominated, in a global sense, by positive stretching that increases/decreases the reaction rate of diffusionaly unstable/stable turbulent premixed flames, similar to what is found for laminar flames.

4. The peak light intensities of CH* and C₂* emitters are found to be qualitatively correlated with these aforementioned $S_T$ data of lean and rich turbulent CH₄/diluent/air propagating flames, respectively. The larger the losses, the smaller values of peak CH* (lean) and/or C₂* (rich) intensities. Therefore, peak CH* and/or C₂* light intensities may be used as an estimate of the radiation losses from these lean and/or rich CH₄/diluent/air flames.

5. A general correlation of turbulent burning velocities in a form of $(S_T - S_L)/u' = 0.06Da^{0.58}$ is proposed, in which the scatter plot of $S_T/S_L$ data as a function of $u'/S_L$ for N₂ and CO₂ diluted flames can be compressed and well approximated.

As a final remark, the large decrease in values of $S_T$ and peak CH*/C₂* light intensities due to the radiation losses for rich CH₄ flames can be understood from the structure of premixed flames, in which the rate-determining reaction in the inner layer is rather sensitive to the presence of H radicals and the depletion of H radicals is much faster in rich CH₄/air flames than in lean CH₄/air flames.
Acknowledgements

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References


TABLE 1
Some properties of CH$_4$/air flames with diluents

<table>
<thead>
<tr>
<th>Properties</th>
<th>$S_L$ (cm/s)</th>
<th>$T_{ad}$ (K)</th>
<th>$C_{p,\text{react}}^a$</th>
<th>$C_{p,\text{prod}}^a$</th>
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</thead>
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<tr>
<td>Lean $\phi = 0.7$</td>
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<td></td>
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<tr>
<td>CH$_4$(100%)</td>
<td>17</td>
<td>1833</td>
<td>29.58</td>
<td>35.94</td>
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<tr>
<td>CH$_4$/N$_2$(44/56%)</td>
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<td>1720</td>
<td>29.55</td>
<td>35.31</td>
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<tr>
<td>CH$_4$/CO$_2$(59/41%)</td>
<td>10</td>
<td>1729</td>
<td>29.92</td>
<td>36.39</td>
</tr>
<tr>
<td>Rich $\phi = 1.4$</td>
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<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>CH$_4$(100%)</td>
<td>15</td>
<td>1976</td>
<td>29.95</td>
<td>35.19</td>
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<tr>
<td>CH$_4$/N$_2$(59/41%)</td>
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<td>1848</td>
<td>29.88</td>
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<td>CH$_4$/CO$_2$(70/30%)</td>
<td>10</td>
<td>1844</td>
<td>30.32</td>
<td>36.19</td>
</tr>
</tbody>
</table>

$^a$units: kJ/kmol-K, $T_{\text{react}} = 300$K, $T_{\text{prod}} = 1000$K
Table 1. Some properties of CH$_4$/air flames with diluents.

Fig. 1. (a) Schematic diagrams of the cruciform burner with arrangements for flame speeds, pressure, and chemiluminescence intensities measurements. (b) A typical example of the time evolution of voltage signals obtained from a pair of ion-probe sensors using measurement (I) for $S_T$ determination.

Fig. 2. (a) Emission intensities as a function of wavelength for both lean ($\phi = 0.9$) and rich ($\phi = 1.4$) laminar CH$_4$/air flames measured by Kojima et al. [25]. (b) The present light intensities measured by the PM tube without optical filters (all wavelength) plotted against the time duration for both laminar ($f = 0$ Hz; $u'/S_L = 0$) and turbulent ($f = 10$ Hz; $u'/S_L = 4.62$) lean CH$_4$/CO$_2$/air flames, where $S_L = 0.1$ m/s.

Fig. 3. Turbulent flame speeds of CH$_4$-air mixtures at $\phi = 1$ measured by a pair of ion-probe sensors at four different measuring positions (see Fig. 1). The annexes show typical measuring uncertainties plotted as the error bars in the main figure.

Fig. 4. The time evolution of pressure changes inside the cruciform burner during a run with different fan frequencies ranging from 0 to 100 Hz for CH$_4$-air mixtures at $\phi = 1$. The middle small figure is the pressure-voltage correlation for the present pressure transducer.

Fig. 5. The effect of radiation heat loss on $S_T/S_L$ as a function of $u'/S_L$ for CH$_4$/air flames with diluents. Each case includes previous data without diluents [1]. Also plotted are the limiting boundaries for global quenching of these CH$_4$/diluent/air flames [10].

Fig. 6. Same as Fig. 5, but showing the diminishing percentage difference of $S_T/S_L$ between CO$_2$ and N$_2$ diluted flames at various $u'/S_L$ for both lean and rich cases, where all diluted flames have nearly the same $S_L$ ($\approx 0.1$ m/s).

Fig. 7. The chemiluminescence intensities for turbulent propagating CH$_4$/diluent/air flames at both $\phi = 0.7$ (a) and 1.4 (b), where $S_L = 0.1$ m/s and $u'/S_L = 4.62$ for both cases. For comparison, the peak values of all these chemiluminescence data are shifted to 0.5 s, and these data around 0.5 s are best fitted by solid lines.

Fig. 8. Peak CH* and C$_2$* chemiluminescence intensities respectively for lean (a) and rich (b) CH$_4$/diluent/air flames of the same $S_L = 0.1$ m/s as a function of $u'/S_L$. Also plotted are the mean data of turbulent burning rates without error bars from Fig. 5 for comparison.

Fig. 9. A general correlation of turbulent velocities as a function of the turbulent Damköhler number for the present turbulent propagating premixed CH$_4$/diluent/air flames with consideration of radiation heat loss. The annex is previous data for pure CH$_4$/air flames from [1].
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