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Laminar Burning Velocity and Markstein Length of $CH_4/CO_2/Air$ Premixed Flames at Various Equivalence Ratios and CO_2 Concentrations Under Elevated Pressure

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ABSTRACT

Biogas is a renewable fuel predominantly composed of carbon dioxide (CO_2) and methane (CH_4) in varying proportions. The effects of the varying CO_2 proportion need to be clarified for the development of engines. The laminar burning velocity and the burned gas Markstein length of premixed $CH_4/CO_2/Air$ were measured with CO_2 concentration ranging from 0.3 to 0.7 dilution ratios. The equivalence ratio was varied from 0.8 to 1.2, the initial pressure was set at 0.5 *MPa*, and the temperature was set to 298 *K*. The experiment was performed using a high-pressure constant volume combustion chamber. One-dimensional simulation of the flames was conducted using GRI-Mech 3.0. The results showed a reduction in the laminar burning velocity of $CH_4/CO_2/Air$ mixtures with an increase in CO_2 dilution ratio. A non-monotonic relationship was discovered between measured Markstein length and CO_2 dilution ratio with different equivalence ratios. It was found that an increase in the CO_2 dilution increased the response of the flames to stretch. For the lean and stoichiometric flames, the Markstein length was nearly constant with CO_2 dilution of 0–0.5 and decreased with CO_2 dilution of 0.7, suggesting an increase in susceptibility of the flame to the intrinsic flame instability. This was found to be mainly due to an increase in the Zel'dovich number and a decrease in the effective Lewis number with CO_2 dilution. The Markstein length of the rich flame increased with CO_2 dilution as it was more sensitive to CO_2 dilution. Thermo-diffusive effects and pure stretch effects had similar influences on the burning velocity of the rich flames with an increase in stretch rate.

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KEYWORDS
Biogas; methane; carbon dioxide; laminar burning velocity; Markstein length

Introduction

The rise in carbon dioxide (CO_2) concentration in the atmosphere is caused by the anthropogenic activities, including the use of non-renewable fuel (Mondal et al. 2017). The emission of CO_2 is considered as the primary source of global warming (Agahzamin, Mirvakili, Rahimpour 2016; Chen et al. 2016; Marjanović, Milovančević, Mladenović

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2016). Therefore, there is a growing interest and need for alternative carbon-neutral fuels for combustion systems (Anggono et al. 2012).

Biogas is a renewable fuel produced by decomposing biomass using bacterial in anaerobic environment (Kathiraser et al. 2017; Nonaka and Pereira 2016). It mainly consists of methane (CH_4) as the flammable component and CO_2 as a diluent in varying proportions. The composition of biogas depends on the manufacturing method and raw material used (Hosseini and Wahid 2015; Kathiraser et al. 2017; Nonaka and Pereira 2016). Although highly composed of CO_2, the contribution of biogas combustion to greenhouse gases concentration in the atmosphere is relatively small. This is because biogas is derived from organic matters with short carbon cycle that are digested anaerobically (Anggono et al. 2012; Turquand d'Auzay et al. 2019). Those organic matters include agricultural wastes from plants which have contributed to a decrease in CO_2 concentration in the atmosphere (Chen et al. 2016), and possibly neutralize the CO_2 contribution of biogas (Turquand d'Auzay et al. 2019).

There are many benefits of producing and utilizing biogas. Biogas contributes in reducing the environmental odor and wastes minimization (Angelidaki et al. 2011; Heyer et al. 2015) with notable application includes the wastewater treatment (Noyola, Morgan-Sagastume, López-Hernández 2006). It also generates biofertilizer during anaerobic digestion process (Angelidaki et al. 2011) as substitute for the artificial fertilizer. Biogas is a possible replacement of wood for fuel in rural area as overutilization of wood may contribute to deforestation (Zvinavashe et al. 2011). Another advantage of biogas comes from its good resistance to knocking in combustion engines. Such property makes it possible for biogas to be combusted under high compression ratios, which translates to better thermal efficiency (Mokrane, Adouane, Benzaoui. 2018). In terms of economy, the digestion facility for biogas is inexpensive to be constructed and does not require a skilled labor to operate the facility (Anggono 2017). The facility brings economy benefits toward communities in rural area, as the biogas may replace the conventional fuel and fertilizer required by the community (Sasse, Kellner, Kimaro 1991). Furthermore, methods to produce and upgrade biogas quality have been developed to reduce its production cost; notable example is the carbon membranes process suggested by He et al. (2018).

With the overwhelming environmental and economic advantages of biogas, an understanding in its combustion characteristics is critical for its efficient utilization in the combustion systems. Therefore, several studies have examined the combustion characteristics of biogas such as its laminar burning velocity (Anggono 2017). Laminar burning velocity can be used to characterize various premixed flame events including flashback, blow-off, or flame stabilization in the combustion process (Goswani et al. 2014; Okafor et al. 2018). Measurements of the laminar burning velocity are relevant in the improvement of chemical kinetics mechanism. The laminar burning velocity is defined from the basis of an adiabatic planar unstretched flame. Nonetheless, majority of realistic laminar flames involve stretch and/or curvature that consequently lead to a divergence of burning velocities from that of the unstretched flames. Therefore, an approach that relates the stretch rates of the flames with the stretched burning velocity is employed to obtain the laminar burning velocity of unstretched planar flame. (Egolfopoulos et al. 2014).

Another essential combustion parameter, the Markstein length, measures the sensitivity of the stretched laminar burning velocity to the flame stretch rate (Okafor et al. 2018). Markstein length may also be expressed non-dimensionally as Markstein number by normalizing it with the flame thickness (Gu et al. 2000). Albeit the linear correlation between Markstein and Lewis numbers, the Markstein number is the more favorable parameter for modeling the turbulent flame (Bradley et al. 1992), notwithstanding that Lewis number is less complicated to evaluate (Okafor et al. 2019). The behavior of the Markstein number, especially at elevated pressures which are relevant in engines and gas turbine combustors has not been well understood (Okafor, Nagano, Kitagawa 2016).

The current study measured the laminar burning velocity and Markstein length of CH_4 /CO_2/Air mixtures with an attempt to clarify the flame stretch characteristics at elevated pressure using expressions based on the asymptotic analysis. The combustion characteristics of CH_4 are well understood, helping the development of numerical approaches to model the combustion of CH_4. There also exists an extensive volume of reports in the literature on the combustion properties of biogas, mainly investigating the effect of diluents on the burning velocity. Diluents naturally exist in biogas as CO_2 and N_2 (nitrogen). Several studies indicate that an increase in CO_2 concentration leads to a decrease in the laminar burning velocity (Anggono et al. 2013; Askari and Ashjaee 2017; Galmiche et al. 2011; Nonaka and Pereira 2016; Zeng et al. 2018).

Nonaka and Pereira (2016) examined the laminar burning velocity of biogas through heat flux method at 0.10 *MPa* and CO_2 concentration up to 50%. Furthermore, the study complimented the experimental results with flame modeling of four different mechanisms: GRI-Mech 3.0 (Smith et al. 2000.), San Diego (University of California at San Diego 2011), Konnov (Coppens, De Ruyck, Konnov 2007), and USC Mech II (Wang et al. 2007). They reported the decrease of burning velocity ranging from 6.9% to 50.2% due to dilution of CO_2 with concentration extending from 10% to 50% for stoichiometric mixtures (Nonaka and Pereira 2016). On the other hand, Zeng et al. (2018) investigated the laminar burning velocity of landfill gas with various CO_2 concentrations of 23.5%–35.5% and fixed N_2 concentration of 17%. The experiment was performed under atmospheric and elevated initial pressures with equivalence ratio ranging from lean to rich mixtures. They conducted the study experimentally by employing Schlieren technique in a constant volume combustion chamber. Their study found a decrease of laminar burning velocity with an increase of CO_2 proportion from 23.5% to 35.5% in the mixture.

Anggono et al. (2013) compared the laminar burning velocity of biogas (containing N_2 and CO_2) with pure CH_4 for various equivalence ratios of 0.5–1.4 in a spherical combustion bomb. The measurement was conducted under atmospheric temperature and the initial pressure was set for 0.05 *MPa* and 0.1 *MPa*. Their results indicated a decrease in laminar burning velocity with the presence of CO_2 and N_2. Askari and Ashjaee (2017) conducted their study using landfill gas with CO_2 mole fraction of 0.3–0.5, under various equivalence ratios and pressures ranging from 0.1 to 0.5 *MPa*. The study was conducted experimentally using steel cylindrical combustion chamber and numerically under steady laminar one-dimensional flame with GRI-Mech 3.0 (Smith et al. 2000.) and UBC 2.1 (Huang et al. 2006) mechanisms. Askari and Ashjaee (2017) concluded that the additional fraction of CO_2 in the mixture decreases the adiabatic flame temperatures and consequently, the burning velocity for all level of equivalence ratios and pressures examined.

Furthermore, Galmiche et al. (2011) assessed the laminar burning velocity of CH_4/Air with diluent ratio of CO_2 up to 20% at high temperature of 393 *K* and atmospheric pressure (0.1 *MPa*). The temperature of 393 *K* was set to investigate the effect of water vaporization temperature during combustion. The study was performed by utilizing constant volume cylindrical stainlesssteel combustion chamber using spherical expanding flames technique and flame modeling of GRI-Mech 3.0 (Smith et al. 2000.). The results of the study carried by Galmiche et al. (2011) suggested a consistency between measurement and the flame modeling results; it corroborates the trend that increasing the CO_2 proportion in the mixture decreases the laminar burning velocity. Some studies reported that, compared to different diluents or inhibitors such as water (H_2O) and N_2, CO_2 has a more pronounced effect on the flame speed of the fuel mixture (Galmiche et al. 2011; Halter et al. 2009; Zhang et al. 2016). Halter et al. (2009) suggested that, compared to other diluents, the substantial decrease of the laminar burning velocity as a result of CO_2 dilution is caused by the dynamic of CO_2 dissociation and the heat capacity related to the dilution ratio.

On the other hand, the effects of diluent on the flame stretch response of biogas has not received much attention; notably at elevated pressures as high as 0.5 *MPa*. Most studies conducted their measurements under atmospheric initial pressure. Halter et al. (2009) investigated the impact of N_2 dilution on the burned gas Markstein length of CH_4/Air at 0.1 *MPa* and found an increase of burned gas Markstein length as a result of an increase in the dilution ratio. Khan et al. (2017) also studied the Markstein length of $CH_4/O_2/CO_2$ mixture at atmospheric condition and reported an increase of burned gas Markstein length with an increase in CO_2 proportion in the mixture. According to Halter et al. (2009), the increase in the Markstein length with an increase in the dilution ratio is due to a decrease in the Lewis number. Askari and Ashjaee (2017) determined the Markstein length of landfill gas at elevated pressure and their results shows that the variation of the Markstein number with pressure may depend on the CO_2 dilution ratio and equivalence ratio. Interestingly, their results show an increase in Markstein length with an increase in pressure for the mixtures with CO_2 dilution ratio of 0.5; which is contrary to the trend in the Markstein length of methane-hydrogen-air flames (Okafor, Nagano, Kitagawa 2016) and methane-ammonia-air flames (Okafor et al. 2019). Furthermore, the Lewis number alone may not sufficiently explain the trends in the Markstein length, which is known to incorporate the influence of, in addition to the Lewis number, the preheat zone thickness, the Zel'dovich number and the thermal expansion coefficient. Okafor, Nagano, and Kitagawa (2016), and Okafor et al. (2019) suggested that at elevated pressures, the influence of the Zel'dovich number on the response of the flame to stretch is significant. This phenomenon has not been investigated in CO_2 diluted CH_4 flames.

In this study, the combustion parameters of biogas were investigated for different equivalence ratios and CO_2 concentrations under initial pressure of 0.5 *MPa*. Biogas characteristics with initial pressure of 0.5 *MPa* have not been comprehensively investigated, even though it reflects the realistic condition of an engine (Duynslaegher, Jeanmart, Vandooren 2010). The CO_2 concentration observed in this study reached as high as 70% CO_2 concentration, indicating a highly diluted gas mixture. This level of CO_2 concentration has not been studied previously. This degree of dilution ratio is investigated due to the different range of CH_4 and CO_2 concentration between biogases from various sources (Kathiraser et al. 2017; Nonaka and Pereira 2016), with possibility for some of them having larger composition of CO_2 rather than CH_4 (Hosseini and Wahid 2015). Therefore, clarification for high range of CO_2 concentration under elevated initial pressure in relation to combustion characteristics needs to be done.

The unstretched laminar burning velocity and burned gas Markstein length of CH_4/CO_2/Air flames were meticulously measured in this study under large range of CO_2 concentrations and equivalence ratios. The elaborate kinetic mechanism has been implemented to appropriately simulate the laminar burning velocity for various CO_2 concentrations of the flames. The Markstein length was obtained and its alterations through changes of equivalence ratios and the CO_2 concentrations were analyzed.

Experimental and numerical procedures

The experiment was done by utilizing cylindrically shaped combustion chamber under fixed volume at the Institute of Fluid Science, Tohoku University (Okafor et al. 2018, 2019). The chamber's internal diameter was 270 mm and its length was 410 mm. The mixtures were ignited by spark electrodes in the middle of the chamber. The spark electrodes' radius was 0.75 mm while the spark gap was 2 mm . Flame propagation could be monitored through visual panels made of quartz glass. The diameter of the visual panel was 60 mm, which is small enough compared to the volume of the chamber. Therefore, the pressure rise during flame observation was almost constant (Okafor et al. 2018). A schematic diagram for this experimental apparatus is shown in Figure 1.

The flame was captured using high-speed camera (Photron, FASTCAM Mini AX100). The lens (Nikon, AF Micro-Nikkon 200 mm f/4-D IF-E-D) had approximately 0.2 mm/pixel spatial resolution of the direct image. The memory recorder and high-speed camera captured the ignition timing of the mixture simultaneously.

Dry air premixed with CH_4 with 99.9% of purity was used as the fuel. Dilution of CO_2 was done by using CO_2 with 99.995% purity. Since biogas only contains a minuscule quantity of N_2 and other components (approximately 3%), the proportions of the remaining components are negligible. Therefore, the compounds of the biogas examined in this study were mainly CH_4 and CO_2 (Anggono et al. 2012; Effendi et al. 2005; Kathiraser et al. 2017). The CO_2

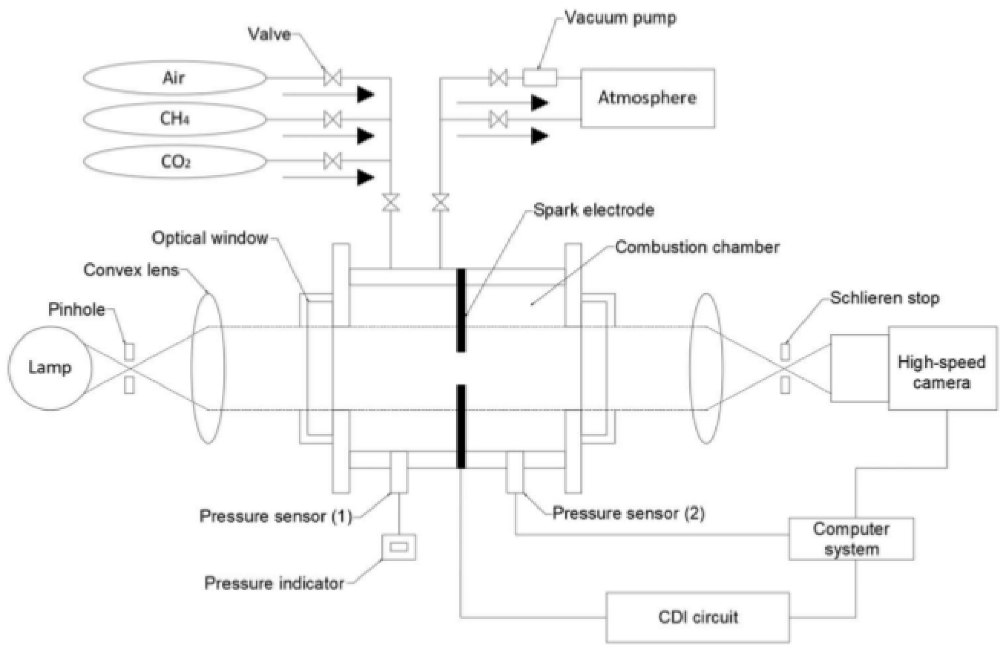

Figure 1. Schematic of the experimental apparatus.

dilution ratio within the fuel, Z_{CO2}, which is considered as the CO_2 fraction in the CH_4/CO_2 mixture, is expressed in Eq. (1).

$$Z_{CO_2} = \frac{[CO_2]}{[CH_4] + [CO_2]} \tag{1}$$

Here $[CO_2]$ stands for mole concentration of CO_2 and $[CH_4]$ stands for the mole concentration of CH_4. Biogas commonly has Zco_2 value between 0.3 and 0.5 (Anggono et al. 2013); however, Zco_2 may reach higher than 0.5 (Hosseini and Wahid 2015). For this study, Zco_2 value was ranged from 0.3 to 0.7 in order to understand the flame characteristics of biogas with various range of CO_2 concentration. Initial pressure, P_i, was set for 0.5 MPa. The initial pressure was measured using GE UNIK 500 silicon pressure sensor with enhanced accuracy of 0.02% FS (full scale) (Okafor et al. 2019). The reason to investigate at this level of pressure is to simulate the elevated pressure condition relevant in SI engines (Duynslaegher, Jeanmart, Vandooren 2010) and therefore yielding a more realistic result compared to the atmospheric initial pressure. The pressure history in the present combustor has been reported elsewhere for the case of spherical CH_4/air flames (Okafor et al. 2018). It was shown that the pressure does not vary appreciably during the period of recording of the flame propagation in the chamber; therefore, the present measurements are considered to have been completed at constant pressure and temperature. Equivalence ratio, ϕ, was varied from lean mixtures (ϕ = 0.8) to rich mixtures (ϕ = 1.2) and the initial mixtures temperature were set to a constant 298 K while being kept within range of ±3 K. All equipment used to perform the experiment were calibrated beforehand to ensure the accuracy of the measurement. The experimental measurements were performed for at least five replications to estimate the uncertainty and to ensure the repeatability of the measurements. The average value of the measurements and the estimated uncertainty based on the standard error of the mean are presented.

Speed of the propagated flame, S_n, was assessed through measuring the evolution of flame radius over time, as depicted in Eq. (2); where r_{sch} is the flame radius obtained from the Schlieren image and t is the time. The development of flame radius in relation with time was evaluated from the obtained Schlieren images, as seen in Figure 2 for flame with Z_{CO2} = 0.5 for all equivalence ratios investigated in this study.

$$S_n = \frac{\mathrm{d}r_{sch}}{\mathrm{d}t} \tag{2}$$

S_n correlates with the flame stretch rate, ε, as depicted in Eq. (3). A_f in Eq. (3) indicates the area of the front spherical flame.

$$\varepsilon = \frac{1}{A_f}\frac{\mathrm{d}A_f}{\mathrm{d}t} = \frac{2}{r_{sch}}\frac{\mathrm{d}r_{sch}}{\mathrm{d}t} \tag{3}$$

Numerous correlations between S_n and ε have been suggested which allow the exclusion of the stretch influence on the stretched flame speed through extrapolation of S_n to zero stretch rate (Egolfopoulos et al. 2014). Unstretched flame speed, S_s, and Markstein length, L_b, were assessed by employing the linear relationship between speed of the propagated flame, S_n, and flame stretch rate, ε, as depicted in Eq. (4). The same relationship is also depicted graphically in Figure 3 for all

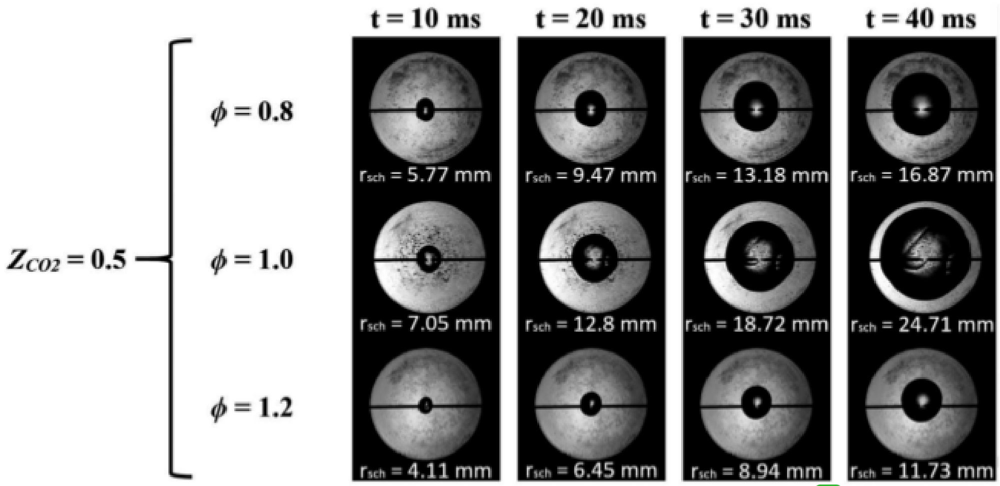

Figure 2. Evolution of the flame radius over time for CH_4/CO_2/Air mixture under initial pressure, P_i, of 0.5 *MPa* for CO_2 dilution ratio, Z_{CO2}, of 0.5 and various equivalence ratios.

level of equivalence ratios and CO_2 concentrations investigated in this study, except for $\phi = 1.2$ and $Z_{CO2} = 0.7$ as no flame propagated under this condition. Markstein length, L_b, was defined as the negative slope of the linear relationship depicted in Eq. (4) and the associated figure (Clavin 1985; Hu et al. 2009). According to Eq. (4), the unstretched flame speed, S_s, is defined as the speed of the propagated flame, S_n, at flame stretch rate, $\varepsilon = 0$ through extrapolation of the stretched flame speed to the point of infinite flame radius, i.e. zero stretch rate. The method described has been utilized by several research teams to evaluate the unstretched flame speed, S_s, and Markstein length, L_b (Miao et al. 2009; Tahtouh, Halter, Mounaïm-Rousselle 2009; Wu et al. 2009).

$$S_s - S_n = L_b \varepsilon \tag{4}$$

The effective Lewis numbers in the present study were near unity, supporting the use of Eq. (4) for evaluating the unstretched flame speed and burned gas Markstein length. Chen (2011) stated that the linear relationship, as shown in Eq. (4), is appropriate for evaluating the unstretched flame speed of mixtures with near-unity Lewis number. The results from linear relationship were compared with the results from non-linear relationship suggested by Kelley and Law (2009), as shown in Eq. (5). Example for the comparison between the extrapolations of linear and non-linear relationships is shown in Figure 4. It was found that both relationships have comparable results and give the same trend of the laminar burning velocity and Markstein length with the addition of CO_2 dilution ratio. However, the magnitude of the Markstein length measured using the linear method did not agree with that measured using the non-linear method for $\phi = 1.2$ and $Z_{CO2} = 0.5$, and $\phi = 0.8$ and $Z_{CO2} = 0.7$, probably due to a relatively strong response of the flame to stretch effect at these conditions as discussed further in the subsequent section.

$$\left(\frac{S_n}{S_s}\right)^2 \ln\left(\frac{S_n}{S_s}\right)^2 = -2\frac{L_b \varepsilon}{S_s} \tag{5}$$

Evaluation of the unstretched laminar burning velocity, S_L, was conducted by using relationship between the unstretched flame speed, S_s, the density of unburnt mixture, ρ_u,

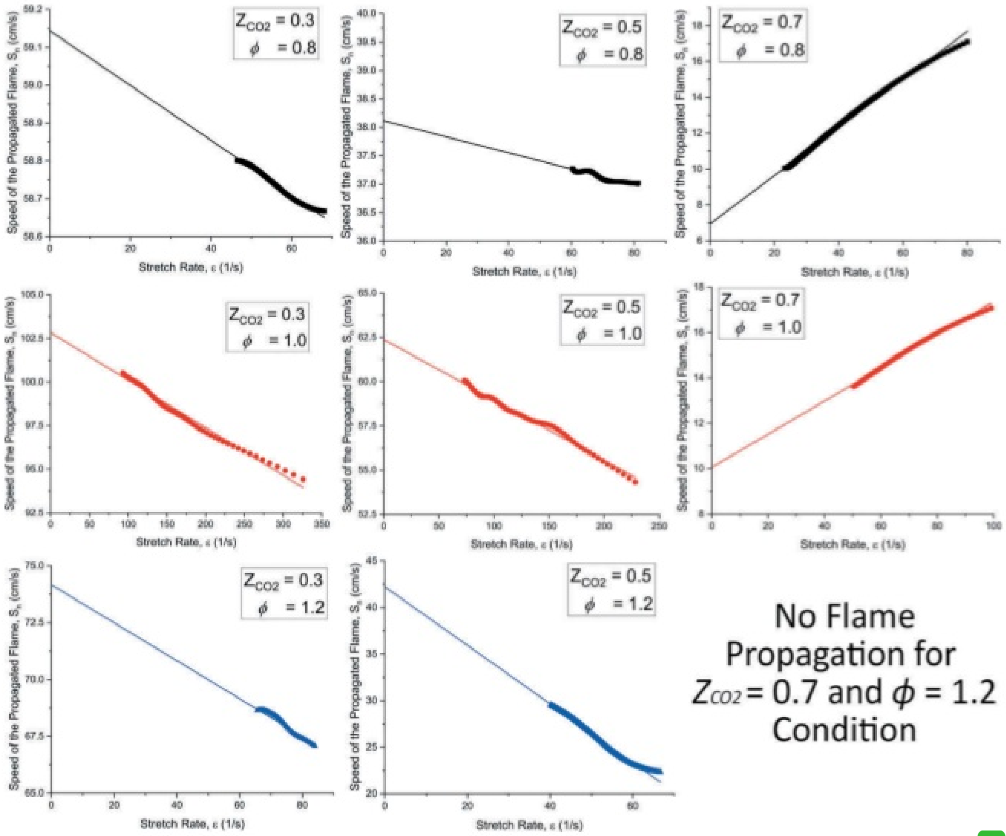

Figure 3. Speed of the propagated flame, S_n, as a function of stretch rate, ε, of CH_4/CO_2/Air under initial pressure, P_i, of 0.5 *MPa* for various equivalence ratios, ϕ, and CO_2 dilution ratio, Z_{CO2}

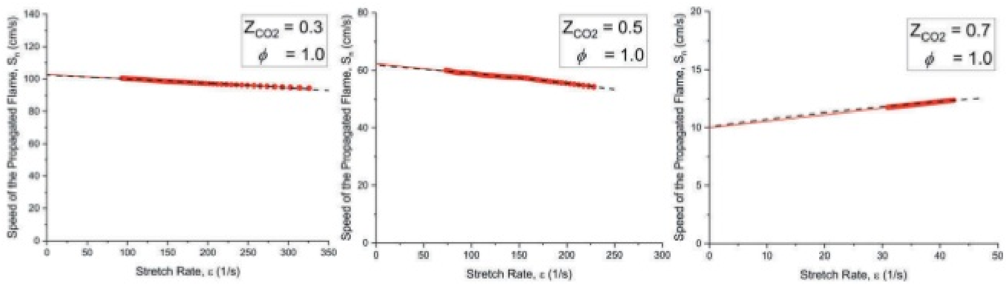

Figure 4. Comparison of the linear extrapolation (solid line) and non-linear extrapolation (dashed line) for speed of the propagated flame, S_n, as a function of stretch rate, ε, of CH_4/CO_2/Air under initial pressure, P_i, of 0.5 *MPa* for equivalence ratio, ϕ, of 1.0 and various CO_2 dilution ratios, Z_{CO2}.

and the density of burnt gas, ρ_b, shown in Eq. (6). The densities were calculated using GASEQ software (Morley 2005). Similar equipment to capture the flame radii and the technique used to evaluate them have been used previously to measure the laminar burning velocity and the Markstein length of the flame from various fuel mixtures (Hayakawa et al. 2015; Ichikawa et al. 2015; Okafor et al. 2018, 2019).

$$S_L = \frac{\rho_b}{\rho_u} S_s \quad (6)$$

Numerical investigations were performed to assess the unstretched laminar burning velocity, S_L, of the mixture. The laminar burning velocity is assessed by utilizing a laminar freely propagating one-dimensional flame model of CHEMKIN-PRO with reaction model based on GRI-Mech 3.0 (Smith et al. 2000.). In numerically evaluating the unstretched laminar burning velocity, S_L, the initial temperature of the mixture was set to 298 K.

In this study, buoyancy effect is observed during the measurement of the flame speed; particularly for flames under high dilution ratio (Z_{CO2} = 0.7) as those flames have small value of laminar burning velocity. For this study, flame under Z_{CO2} = 0.7 and ϕ = 0.8 has the lowest laminar burning velocity value and hence it is the most susceptible to buoyancy effect (Okafor et al. 2019). Flame radii of this flame under Z_{CO2} = 0.7 and ϕ = 0.8 are shown in Figure 5 for the smallest and largest radii used to measure the flame speed. From the figure, it can be observed that, while buoyancy effect promoted a displacement of the flame in the combustion chamber, the shape of the flame did not appreciably change within the range of flame radius used in measuring the laminar burning velocity and the Markstein length. Therefore, buoyancy effects on the flame did not significantly affect the measurements in the present study.

Results and discussion

Laminar burning velocity

Figure 6 displays the experimental and numerical values of the unstretched laminar burning velocity for CH_4/CO_2/Air under various levels of CO_2 dilution ratio, Z_{CO2}, with 0.8, 1.0, and 1.2 equivalence ratio, ϕ. The solid line displays the experimental results and the dashed line displays the numerical results of the unstretched laminar burning velocity. Data for Z_{CO2} = 0 (methane/air flames) for this study are adopted from Okafor et al. (2019). The experimental data for laminar burning velocity with equivalence ratio of 1.2 and CO_2 dilution ratio of 0.7 is

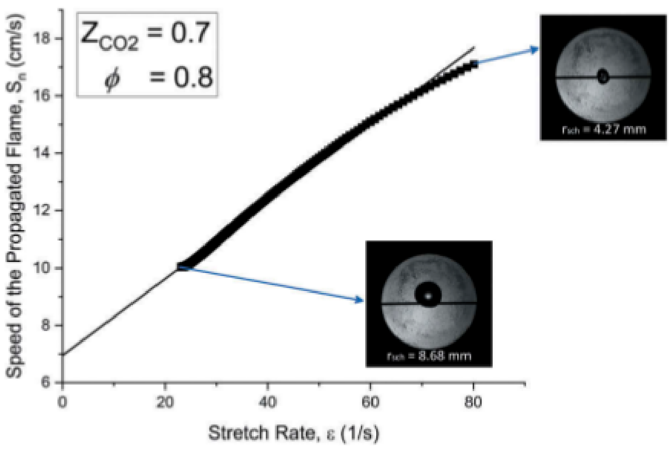

Figure 5. The flame radii of Z_{CO2} = 0.7 and ϕ = 0.8 for the smallest and largest radii used to measure the flame speed.

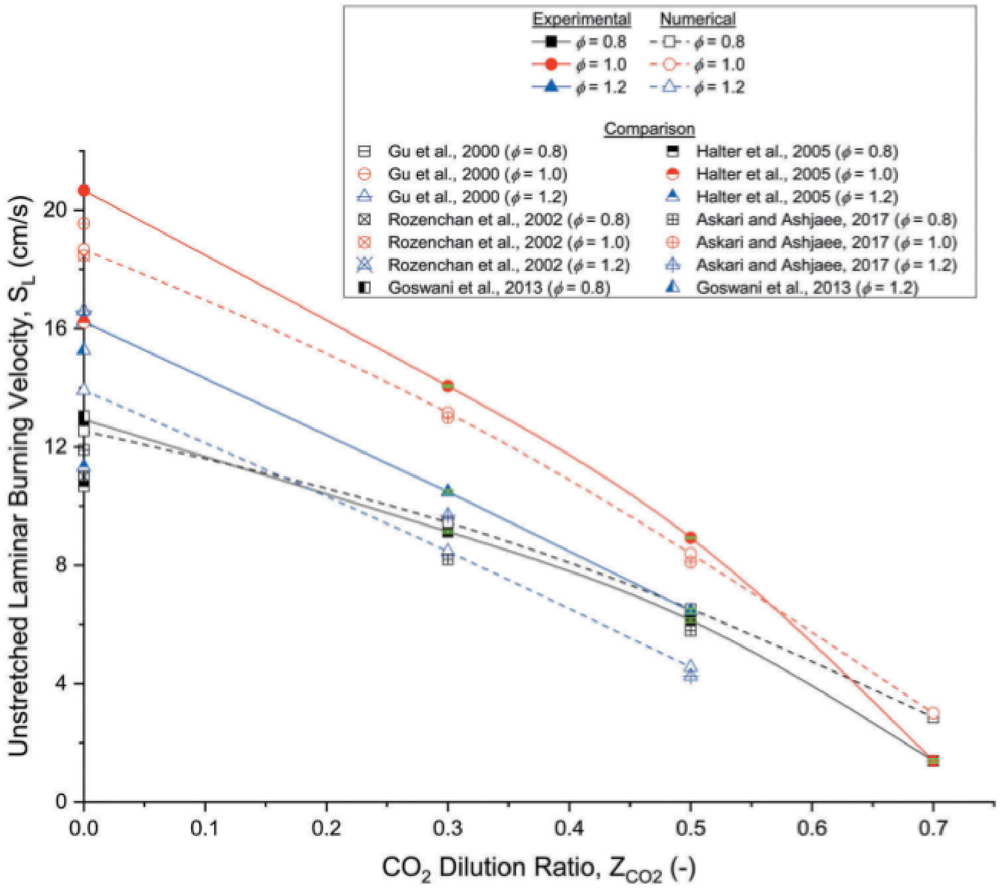

Figure 3 Unstretched laminar burning velocity, S_L, of CH_4/CO_2/Air mixture as a function of CO_2 dilution ratio, Z_{CO2}, at equivalence ratio, ϕ, of 0.8 (lean), 1.0 (stoichiometry) and 1.2 (rich) under initial pressure, P_i, of 0.5 *MPa*. Data for $Z_{CO2} = 0$ (methane/air flames) are adopted from Okafor et al. (2019). The green error bars represent the standard error of the mean in the present measurements.

not available since no flame propagated at this condition. The results showed that the numerical measurements barely differ from the experimental measurements, indicating good accuracy of the numerical results. Furthermore, results from previous studies (Askari and Ashjaee 2017; Goswani et al. 2013; Gu et al. 2000; Halter et al. 2005; Rozenchan et al. 2002) are provided in the figure and they show that the results of this study are consistent with them. The results of this study suggested that, as the CO_2 dilution ratio increases, the unstretched laminar burning velocity decreases. CO_2, an inhibitor, decreases the flame temperature (Askari and Ashjaee 2017), and axiomatically the unstretched laminar burning velocity by decreasing the amount of heat produced during fuel oxidation process (Anggono et al. 2013, 2012; Askari and Ashjaee 2017). Furthermore, CO_2 also decreases the proportion of the reactive fuel oxidation components in the mixture, diminishing the chemical reactions rate of the bimolecular reactions (Anggono et al. 2013, 2012). Zeng et al. (2018) suggested that the lower laminar burning velocity in CO_2 diluted mixture could be associated with the lower calorific value of the mixture compared to that of pure CH_4.

As for the effect of equivalence ratio, the change from $\phi = 0.8$ to $\phi = 1.0$ increased the unstretched laminar burning velocity, and from $\phi = 1.0$ to $\phi = 1.2$, the unstretched laminar burning velocity decreased. The increase of unstretched laminar burning velocity from $\phi = 0.8$ to $\phi = 1.0$ is the result of the ideal proportion of air and fuel mixture in the $\phi = 1.0$ (stoichiometry) condition to thoroughly burn the biogas fuel (Anggono et al. 2013, 2012). On the other hand, the unstretched laminar burning velocity decreases from $\phi = 1.0$ to $\phi = 1.2$ as a result of increasing CO_2 mole fraction in higher equivalence ratio (Anggono et al. 2013). CO_2 adversely affects the propagation of flame, decreasing the flame radius and supposedly the flame speed. It is also observed from the figure, the increase of CO_2 dilution ratio decreases the difference of the unstretched laminar burning velocity value between equivalence ratios investigated.

Markstein length

Figure 7 shows the measured Markstein length, L_b, of CH_4/CO_2/Air flames at the initial pressure, P_i, of 0.5 *MPa* for various levels of CO_2 dilution ratio, Z_{CO2}, and equivalence ratio, ϕ. Data for $Z_{CO2} = 0$ (CH_4/air flames) for this study are adopted from Okafor et al. (2019). The results of this study were compared with previous measurements (Askari and Ashjaee 2017; Gu et al. 2000; Halter et al. 2005; Rozenchan et al. 2002) and were found to be consistent. The results from Askari and Ashjaee (2017) for $\phi = 1.2$ shows similar trend with the measurement conducted in this study, although the results differ in magnitude. There are not many studies available to compare the results as most studies were conducted under atmospheric initial pressure and with low or even no CO_2 dilution. The experimental results suggested, for equivalence ratio of 0.8 and 1.0, the Markstein length was nearly constant as CO_2 dilution ratio increased from 0 to 0.5. It decreased and became negative as the CO_2 dilution ratio increased to 0.7. A negative Markstein length

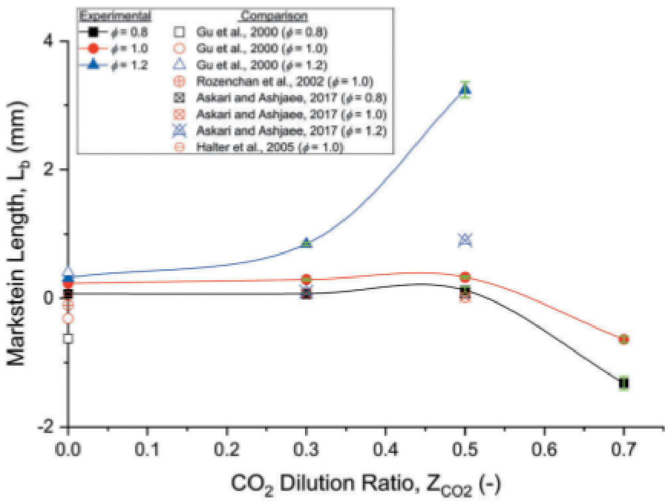

Figure 7. Markstein length, L_b, of CH_4/CO_2/Air mixture as a function of CO_2 dilution ratio, Z_{CO2}, at equivalence ratio, ϕ, of 0.8 (lean), 1.0 (stoichiometry) and 1.2 (rich) under initial pressure, P_i, of 0.5 *MPa*. Data for $Z_{CO2} = 0$ (methane/air flames) are adopted from Okafor et al. (2019). The green error bars represent the standard error of the mean in the present measurements.

indicates an increase in the laminar burning velocity with an increase in the flame stretch rate. For equivalence ratio of 1.2, the Markstein length, which was positive for all values of Z_{CO2}, was more sensitive to the change of CO_2 dilution ratio than those of the lean and stoichiometric flames. Under equivalence ratio of 1.2, the flame also showed a significant increase of Markstein length with the increase of the CO_2 proportion in the fuel mixture. These observations indicate that an increase in CO_2 concentration in the mixture increased the response of the laminar burning velocity to flames stretch rate. The result also suggests that for the stoichiometric and lean flames, an increase in the dilution ratio may increase the susceptibility of the flames to the intrinsic flame instability. On the other hand, thermo-diffusive effects may increasingly suppress the intrinsic flame instability in the rich flames as the dilution ratio increases. The non-monotonic relationship of Markstein length with respond to CO_2 dilution ratio across equivalence ratios found in this study has not been observed previously.

To evaluate and verify the non-monotonic relationship found in this study, the trend in the Markstein length was further studied. An analytical expression based on the asymptotic analysis was used to evaluate it. The analytical expression correlates the Markstein length with the preheat zone thickness, δ_l, effective Lewis number, Le_{eff}, thermal expansion coefficient, σ, and Zel'dovich number, Ze (Clavin 1985) as given in Eq. (7).

$$L_b = \delta_l\left[f_1(\sigma) + Ze\left(Le_{eff} - 1\right)f_2(\sigma)\right] \tag{7}$$

Here, $f_1(\sigma)$ and $f_2(\sigma)$ are function of σ as given in Eqs. (8) and (9). Both have positive value for realistic value of σ (Okafor et al. 2018).

$$f_1(\sigma) = \frac{2}{\sqrt{\sigma} + 1} \tag{8}$$

$$f_2(\sigma) = \frac{2}{\sigma - 1}\left[\sqrt{\sigma} - 1 - \ln\frac{\sqrt{\sigma} + 1}{2}\right] \tag{9}$$

Equation (7) indicates that the laminar burning velocity may still vary with flame stretch rate (i.e. $L_b \neq 0$) when there is no thermo-diffusive effects ($Le_{eff} = 1$) owing to the effect of pure stretch, which is expressed by the first term of Eq. (7). An increase in pure stretch always tends to lead to a decrease in the laminar burning velocity of positively stretched flames. On the other hand, thermo-diffusive effects, expressed by the second term of Eq. (7), may contribute to a decrease or an increase in the laminar burning velocity depending on the value of the effective Lewis number.

The Lewis number gives a qualitative measure of thermo-diffusive effects on the laminar burning velocity. For mixtures with stoichiometry far from unity, the Lewis number is usually defined base on the deficient reactant. In the present study, the effective Lewis number of the mixtures was calculated using the Lewis number of the fuel and the oxidizer as given in Eqs. (10) and (11) (Bechtold and Matalon 2001). For mixtures with equivalence ratios far from unity, the Lewis number of the deficient reactant is weighted more than that of the excess reactant in Eq. (10). On the other hand, the effective Lewis number of a mixture with $\phi = 1.0$ is defined as the average of the Lewis number of the fuel and oxidizer.

$$Le_{eff} = 1 + \frac{(Le_{O_2} - 1) + (Le_{CH_4} - 1)A}{1 + A} \quad (10)$$

$$A = 1 + Ze\,(\varphi - 1) \quad (11)$$

The Lewis number of the fuel, Le_{CH_4}, or oxidizer, Le_{O_2}, was defined as the thermal diffusivity of the mixture divided by the molecular diffusivity of the fuel or oxidizer, respectively. These properties were obtained at the initial temperature and pressure for each condition in this study. Φ is the ratio of unburned mixture's mass excess-to-deficient reactants in regards with their ratio for $\phi = 1.0$. The Zel'dovich number, Ze, is a dimensionless activation energy of the chemical reaction, which conveys the sensitivity of the laminar burning flux to the variation in the peak flame temperature. The Zel'dovich number can be approximated using Eq. (12) (Muller, Bollig, Peters 1997).

$$Ze = 4\frac{T_b - T_u}{T_b - T^0} \quad (12)$$

where T_u and T_b are the mixture temperature and the adiabatic flame temperature, respectively, and T^0 is the inner layer temperature. In this study, following Muller, Bollig, and Peters (1997), T^0 was evaluated as the peak of the gradient in the temperature profile. Similar method is verified by

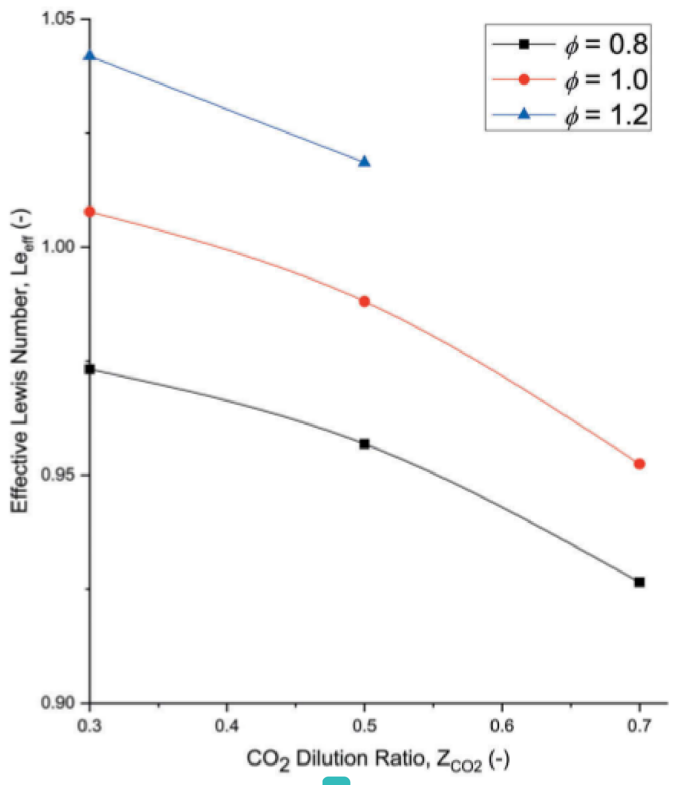

Figure 8. Effective Lewis number, Le_{eff}, of CH_4/CO_2/Air mixture as a function of CO_2 dilution ratio, Z_{CO2}, at equivalence ratio, ϕ, of 0.8 (lean), 1.0 (stoichiometry) and 1.2 (rich) under initial pressure, P_i, of 0.5 *MPa*.

Göttgens, Mauss, and Peters (1992) and has been used in a study conducted by Middleton et al. (2012) to evaluate the inner layer temperature.

Figure 8 shows that the effective Lewis number of the mixtures decreases with an increase in the CO_2 dilution ratio. For the lean flame, the effective Lewis number was less than unity for all values of CO_2 concentration. The effective Lewis number of the stoichiometric flame became less than unity as CO_2 dilution ratio increased to 0.5. For the rich flame, the effective Lewis numbers was larger than unity at all studied conditions. An effective Lewis number less than unity indicates that the effects of molecular diffusion into the flame is more dominant than that of thermal diffusion from the flame thereby promoting an increase in flame temperature and, consequently, the laminar burning velocity of the positively stretched flame fronts (Addabbo, Bechtold, Matalon 2002). The effects of this imbalance in the diffusion of heat and molecules may also promote the development of cellular instability on the flame front of these flames with effective Lewis number less than unity. According to Eq. (7), the Markstein length may be negative when the effective Lewis number is less than unity, in agreement with the trend in the measured L_b of the lean and stoichiometric mixtures. On the other hand, thermo-diffusive effects may subdue the development of cellular flame instability in flames with Lewis number larger than unity, owing to the dominance of heat diffusion from the flame over molecular diffusion into the flame. For these flames, the laminar burning velocity may always decrease with an increase in positive flame stretch rate, i.e. the Markstein length may always be positive. This corresponds to the trend in the measured L_b of the rich flame.

It is important to note however, that the effective Lewis number alone does not fully explain the trends in the measured Markstein length because some of the mixtures with effective Lewis number less than unity still have a positive Markstein length. As given in Eq. (7), the Zel'dovich number may promote or inhibit thermo-diffusive effects because it may express the sensitivity of the laminar burning velocity to the variation in flame temperature due to thermo-diffusive effects. Figure 9 shows that the Zel'dovich number increased with CO_2 dilution ratio. It can be understood that for the flames with effective Lewis number less than unity, the Markstein length becomes negative when the Zel'dovich number is sufficiently large such as seen for the lean and stoichiometric flames with dilution ratios of 0.7.

Figure 10 shows that the preheat zone thickness, δ_l, increased with an increase in CO_2 dilution ratio. Here, the preheat zone thickness was evaluated through Eq. (13) (Okafor, Nagano, Kitagawa 2016) in which c_p is defined as the specific heat of CH_4/CO_2/Air under constant pressure and λ is defined as the thermal conductivity of CH_4/CO_2/Air. Numerical results of the S_L were used to calculate the preheat zone thickness.

$$\delta_l = \frac{\lambda}{(\rho_u c_p S_L)} \tag{13}$$

According to Eq. (7), an increase in the preheat zone thickness leads to an increase in the absolute value of the Markstein length, which corresponds to the trends in the measured Markstein length.

Figure 11 shows that the thermal expansion coefficient, σ, decreased with an increase in CO_2 dilution ratio. Thermal expansion coefficient is the ratio of the density of unburnt mixture, ρ_u, and the density of burnt gas, ρ_b. The effect of the decrease in the thermal expansion coefficient may contribute to an increase in pure stretch effects according to Eq.

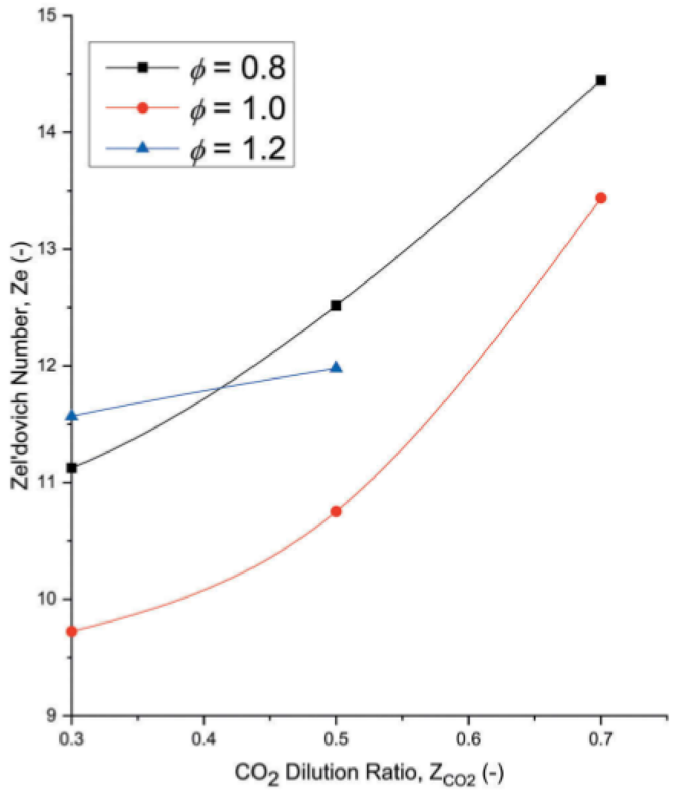

Figure 9. Zel'dovich number, Z_e, of CH_4/CO_2/Air as a function of CO_2 dilution ratio, Z_{CO2}, at equivalence ratio, ϕ, of 0.8 (lean), 1.0 (stoichiometry) and 1.2 (rich) under initial pressure, P_i, of 0.5 *MPa*.

(8). The higher sensitivity of the Markstein length of the rich flame to CO_2 dilution in comparison to that of the other flames is because pure stretch effects and thermo-diffusive effects contribute to an increase in the Markstein length of the rich flame. On the other hand, pure stretch effects and thermo-diffusive effects have opposing influences on the value of Markstein length of the lean and stoichiometric flames which have effective Lewis number less than unity.

Conclusion

The unstretched laminar burning velocity and burned gas Markstein length of biogas (CH_4/CO_2/Air) were studied under elevated initial pressure of 0.5 *MPa*, for various equivalence ratios of 0.8–1.2, and CO_2 concentration up to 70%. The unstretched laminar burning velocity of the mixtures decreased with an increase in CO_2 concentration. The Markstein length increased with CO_2 dilution for the rich flame. The Markstein length was nearly constant for CO_2 dilution of 0–0.5 and decreased with CO_2 dilution of 0.7 for the lean and stoichiometric flames, indicating a non-monotonic relationship. Similar trend has not been observed previously. An analytical expression was used to verify the non-monotonic relationship. It was found that the response of the flame to stretch increased with an increase in the CO_2 concentration owing to an

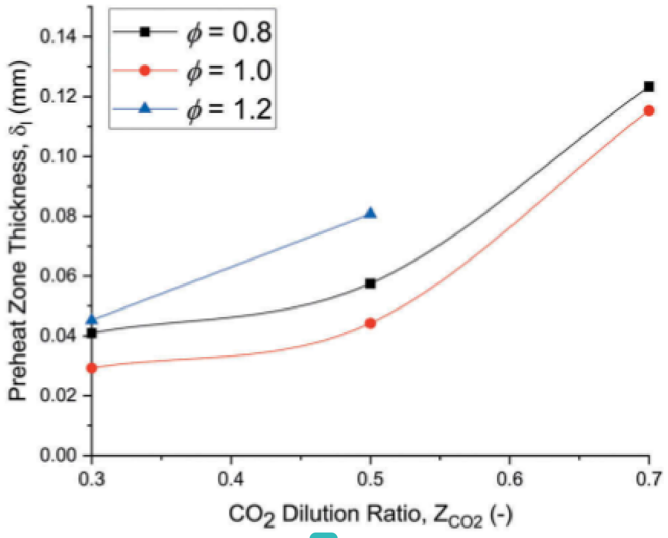

Figure 10. Preheat zone thickness, δ_l, of CH_4/CO_2/Air mixture as a function of CO_2 dilution ratio, Z_{CO2}, at equivalence ratio, ϕ, of 0.8 (lean), 1.0 (stoichiometry) and 1.2 (rich) under initial pressure, P_i, of 0.5 *MPa*.

increase in the preheat zone thickness, the Zel'dovich number and a decrease in the effective Lewis number. For the rich flame, the response of the laminar burning velocity to flame stretch rate increased more rapidly than the case of the lean and stoichiometric flames because thermo-diffusive effects and pure stretch effects had similar influences on the burning velocity of the rich flames unlike in the lean or stoichiometric flames.

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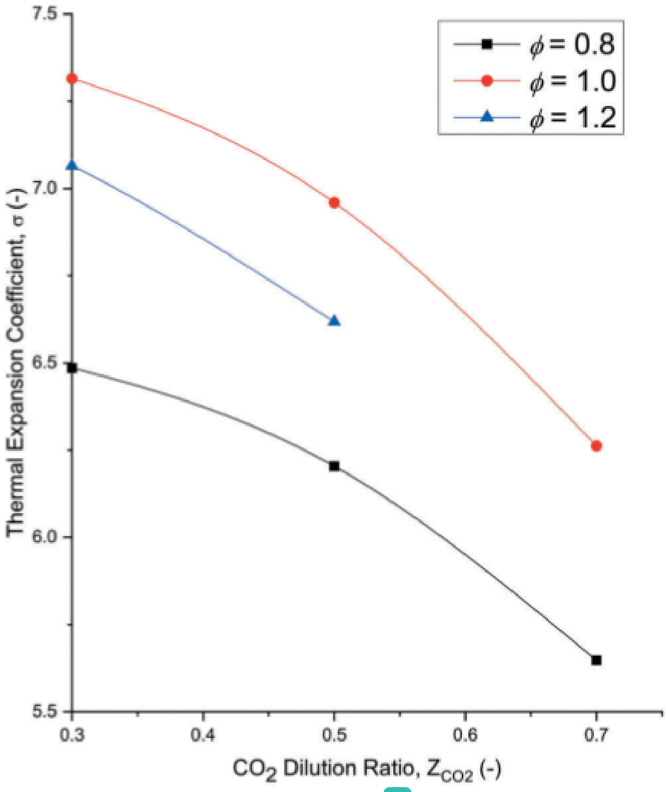

Figure 11. Thermal expansion coefficient, σ, of CH_4/CO_2/Air mixture as a function of CO_2 dilution ratio, Z_{CO2}, at equivalence ratio, ϕ, of 0.8 (lean), 1.0 (stoichiometry) and 1.2 (rich) under initial pressure, P_i, of 0.5 *MPa*.

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