Measurement of Adiabatic burning velocity in Natural Gas like mixtures

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Abstract

Experimental measurements of the adiabatic burning velocities were carried out for natural gas like mixtures burning in air over a range of equivalence ratios at atmospheric pressure. Effect of CO$_2$ dilution upto 60%, N$_2$ dilution upto 40% and 25% enrichment of ethane on burning velocity of methane-air flames were studied. Heat flux method with setup similar to that of de Goey and co workers [9] was used for measurement of burning velocities. Initially experiments were done for methane-air and ethane-air mixtures at various equivalence ratios and the results were in good agreement with published data in the literature. Computations were performed using PREMIX code with GRI 3.0 reaction mechanism for all the mixtures. Predicted flame structures were used to explain effect of N$_2$ and CO$_2$ dilution on burning velocity of methane-air flames. Peak burning velocity for CH$_4$/CO$_2$ -air mixtures occur near to $\phi = 1.0$.

Key words: Adiabatic burning velocity, natural gas, premixed flames

1 Introduction

With the depletion of crude oil reserves and the strengthening of automotive emission legislations, the use of natural gas (NG) as an alternative fuel has been promoted both in combustion engines and power generation. Natural gas contains mainly methane (typically 65-90 % or more by volume) along with
higher hydrocarbons, inert gaseous components like N₂, CO₂, water vapor and trace compounds. The composition of natural gas varies widely from one source to another in terms of the fractions of higher hydrocarbons summarized as "C2+" gases (at present vary from 7 - 16%) and inert gaseous components like N₂, CO₂ (at present 20-25% maximum). This variation of both C2+ and inerts is expected to widen in the future [1]. Hence interest in fuel-flexible gas turbine engines led to research on premixed combustion parameters like laminar burning velocity and ignition delay time. Fuel flexibility can impact several important premixed burner design issues such as flashback, blow off, auto ignition and stability. Laminar burning velocities are important because they are related not only to flashback and blow off issues, but they also play a role in the stability of the flame in the combustor. Laminar burning velocity is also a quantity of fundamental importance because it provides benchmarks for validation of detailed reaction mechanisms. Also in recent years European countries have encouraged utilization of sustainable alternative fuels like biogas in gas turbines [2]. One of the most promising alternative fuel options is biogas (or landfill gas) which contains 45 - 60% CH₄, 40 - 55% CO₂ and traces of H₂S, N₂ etc. This can be derived from waste methanisation, agricultural wastes etc. In Germany the electricity produced based on biogas is 665 MW and likely to increase rapidly in future [3]. The effective utilization of biogas depends on good design of combustion equipments and for this knowledge of burning velocity is essential.

There are two types of methods used for determining laminar velocity: the stationary flame methods and moving flame methods. In the former category of methods, the flame front remains stationary in space: e.g., Bunsen burner method, heat flux method etc. In the latter, the flame moves with respect to some fixed point (i.e., the point of ignition): e.g., soap bubble method, constant volume method, constant pressure method etc.

Botha and Spalding [4] measured the increase in temperature of cooling water flowing through a porous burner and hence the heat loss incurred to stabilize the flame. They obtained adiabatic burning velocity by extrapolating their data to zero heat loss. Practical problems with this method were (a) that the temperature rise was very small leading to larger uncertainties and (b) extrapolation was used to determine burning velocity. Later, de Goey and co-workers [5] introduced heat flux method for measuring adiabatic burning velocity and stabilizing flat flames. The unburnt gas mixture is heated while flowing through the burner plate using hot water jacket. This would compensate for the heat loss necessary for stabilizing the flame. They analyzed how close these flames were to perfect 1D adiabatic flames [6–8]. With some modifications in burner construction, this technique was used for different hydrocarbon-air mixtures and well validated against literature [9,10]. Recently, Hermanns [11,12] used same setup for determining effect of hydrogen addition to the laminar burning velocities of methane-air mixtures. Konnov
and co-workers [13–17] had used a replica of this setup and worked extensively on the determination of adiabatic burning velocities of methane (CH$_4$), ethane (C$_2$H$_6$) and methane-hydrogen mixtures with different dilution ratios of artificial air having carbon dioxide (CO$_2$), nitrogen (N$_2$) or Argon (Ar). Clarke et al. [18] used constant volume technique at zero gravity conditions for determination of burning velocities of natural gas-like compositions (i.e. methane/diluents-air mixtures). Their data are not corrected for the effects of stretch on burning velocity and stretch corrected data of methane-diluent mixtures do not appear to be available in literature.

The present work is an experimental and computational investigation into the effect of dilution with N$_2$ and CO$_2$ on adiabatic burning velocity of methane-air mixtures. The effect of addition of C$_2$H$_6$ to methane-air mixtures is also studied in the present work. Heat flux method is used for the experimental study. The setup is validated initially by obtaining laminar burning velocity values for methane-air and ethane-air mixtures and comparing the data with literature. Computations were carried out using the steady one-dimensional laminar premixed flame code, PREMIX, due to Kee et al. [20]. GRI-Mech 3.0 reaction mechanism was used for prediction of adiabatic burning velocity of the mixtures studied in this work.

2 Experimental Setup

The experimental setup used in the present work is similar to the one used by Bosschaart and de Goey [9]. It consists of a burner head mounted on a plenum chamber, mass flow controllers, mixing tube to mix fuel and air and heating and cooling water baths as shown in figure 1a. The brass burner plate (2 mm thick) on which the flame is stabilized is a perforated disc with a hexagonal pattern of 0.5 mm diameter holes with 0.7 mm pitch as shown in figure 1b. It has been shown that this kind of pattern stabilizes a flat flame on the burner [6]. Six K-type thermocouples of 75 µm wire were soldered into selected holes on perforated plate on the upstream side. The thermocouples are positioned at different radii and different circumferential locations to measure the temperature profile on the burner plate. The burner plate was soldered at the burner outlet for good thermal contact.

The upper half of the burner head has a heating jacket and a teflon insulation ring that separates it from the lower half with cooling jacket. The heating jacket is supplied with hot water from a constant temperature bath to keep the temperature of the burner plate constant. During the experiments temperature of hot water flowing through heating jacket was fixed at 343 K. The cooling jacket in lower half of the burner head is supplied with water at room temperature (around 307 K during most of the experiments). The heating
jacket keeps the burner plate at a certain temperature higher than unburnt
gas temperature and due to this gas mixture is heated while flowing through
the burner plate. This would compensate for the heat loss necessary for sta-
bilizing the flame.

When gas velocity is lower than the adiabatic burning velocity, flame is stabi-
lized under sub-adiabatic conditions. This causes the net heat loss ( = [heat
loss - heat gain] > 0) to be greater than zero. Then the center of the burner
plate is hotter than the heating jacket. When gas velocity is higher than the
adiabatic burning velocity ( super-adiabatic conditions), the net heat flux is
lower than zero and the center of burner plate is cooler than the heating jacket.
By varying the flow rate of the gas mixture, an appropriate value of gas veloc-
ity could be found where net heat flux is zero. This will manifest in the form of
uniform radial distribution of temperature. The flow velocity at which the net
heat flux is zero is shown to be adiabatic burning velocity. The temperature
distribution of the burner plate is measured with the thermocouples attached
to it and radial temperature profile of the plate obtained by solving the energy
equation [1].

\[ T_p(r) = T_{center} - \frac{q}{4\lambda x_p}r^2 \]

Here \( T_p \) is the temperature profile across the burner plate, \( T_{center} \) is the plate
center temperature, \( q \) is the net heat flux into the plate, \( \lambda \) is the thermal
conductivity of burner plate, \( x_p \) is the plate thickness and \( r \) is the radial
coordinate.

Fuel(CH\(_4\)/C\(_2\)H\(_6\)), diluent and air cylinders were individually connected to a
40µm filter which also acts as buffer vessel and then to a mass flow controller
(MFC). All the gas lines were connected to a mixing tube for mixing of gases
and then premixed unburnt gas is supplied to the inlet of plenum chamber of
the burner. Mass flow controllers (Alicat Scientific Inc.) used in the present
work have an accuracy of ±0.4% of the reading or ±0.2% of full scale. They
measure the flow rates by measuring the pressure drop across unique laminar
flow restriction.

Boundary layers exist near the edge of the burner, due to the influence of the
environment. In these regions flame will not be flat. Therefore the thermocou-
plies are located in an area where these boundary layer effects are negligible,
and this area is termed as 1-D area. The magnitude of this area is determined
by measuring burnt gas temperature profiles at an axial distance of 3 mm
above the burner plate. These measurements were done using a silica coated B-
Type thermocouple with 200 µm wire diameter mounted on traversing equip-
ment, such that the thermocouple was traversed in the radial direction. Figure
2 shows the plot of burnt gas temperature with the radial distance. The tem-
peratures obtained from thermocouples were corrected for radiation using the
method described by Shaddix [19]. From this plot, the 1-D area is found to
be in a radius of < 12 mm. It can also be seen that radiation corrected burnt
temperature is 2160 K (with uncertainty of ±50 K) which is close to equi-
librium adiabatic temperature (2231 K) [8] of methane-air mixture at $\phi = 1$.
This shows that the flame is nearly adiabatic.

In order to experimentally obtain the adiabatic burning velocity of a fuel/oxidizer
mixture at a particular equivalence ratio, first a range of velocities is deter-
mined using burning velocity values predicted computationally. For this range
of velocities, fuel and oxidizer flow rates are calculated.

Keeping the equivalence ratio fixed, the velocities of fuel and oxidizer are
increased and at each combination of fuel and oxidizer velocity, temperature
profile across the burner plate is measured using the thermocouples. Once the
measurements are taken, a least square parabolic curve is fitted to the points
to obtain an equation of the form of the following.

$$ T_p(r) = T_{center} + \alpha^2 r^2 $$

(2)

Here $\alpha^2$ is a constant and proportional to heat flux $q$ in equation (1). As the
velocities are increased the temperatures reduce and also the curve becomes
flatter. Fitting the profile given in equation 2 gives us the value of $T_{center}$ and
$\alpha^2$. $\alpha^2$ is then plotted against the corresponding total mixture velocity. Figure
3 shows such a plot for an equivalence ratio of 0.9. A second degree polynomial
fit is made through the points in the plot. The velocity at which $\alpha$ becomes
zero gives the value of adiabatic burning velocity at that equivalence ratio[9].

3 Uncertainty Analysis

Uncertainty analysis was carried out using method given by de Goey and co-
workers [9,12]. After calibration uncertainty of an MFC is 0.4% of reading +
0.2% of full scale and uncertainty in temperature measurements using K-Type
thermocouples is ±1 K. Maximum uncertainty in measured burning velocity
is ±0.8 cm/s. Relative accuracy of equivalence ratio was found to be 1.5%.

4 Computational details

Premixed flat flames stabilized on flat burners resemble steady one-dimensional
adiabatic freely propagating flames in the doubly infinite domain. The agree-
ment of the vertical temperature profile of stabilized adiabatic flame measured by using CARS thermometry [8], compared to corresponding free flame was found to be very good near the flame. The only deviation observed was upstream of the flame where the burner plate is present in the former since the burner plate is at relatively lower temperatures (400-450K). Hence the flame structure is not significantly affected by the presence of burner plate and is identical to that of the freely propagating flame.

Therefore, burning velocity computations for comparison with experimental data are performed on freely propagating steady adiabatic flames. The PREMIX code [20] was used for the computations. The transport properties from Sandia National Laboratories [21,22] were used. Multi-component diffusion and thermal diffusion options were taken into account. The upwind differencing for convective terms was used and hence numerical diffusion which is introduced using this scheme, is minimized by adaptive refining of the computational mesh. It is observed that by using adaptive grid parameters GRAD = 0.02 and CURV = 0.1, the burning velocity obtained is grid independent.

Hermanns [12] has compared various chemical reaction mechanisms available in literature for prediction of laminar burning velocity of methane-air mixtures and found that GRI-Mech 3.0 predicts burning velocities well within uncertainty limits of experiments for a wide range of equivalence ratios. GRI-Mech 3.0 [23] is an optimized mechanism designed to model natural gas combustion, including NO formation and reburn chemistry. Its optimization was primarily limited to temperature range of 1000 to 2500 K, pressures from 10 Torr to 10 atm, and equivalence ratio from 0.1 to 5 for premixed systems.

5 Validation of experimental setup

In this section, initial experiments done on methane-air and ethane-air to validate the experimental facility built are presented. There is literature abundant available on unstretched laminar burning velocity of methane and ethane using different techniques for validation of the setup.

5.1 Methane-air

Methane-air is the most widely used mixture to demonstrate the validity of any new experimental approach to determine laminar burning velocity due to availability of many experimental results using various techniques. In the recent years, stretch corrected results have converged to very narrow range of scatter [10].
The present experimental results of adiabatic burning velocities of methane-air are plotted in figure 4. Present experiments were performed at 1 bar pressure and unburnt gas temperature of 307 K, while in literature data were obtained at 295-298K. Figure 4 includes data obtained from stretch free flat flames using heat flux method [10,12,13].

The present experiments had an unburnt gas temperature 9K higher than that used in literature, which typically leads to 1-2 cm/s increase in burning velocity. This deviation is demonstrated in the figure 4 by the difference between simulations done at 298 K and 307 K. Present experimental data was corrected at each equivalence ratio for unburnt gas temperature using the corresponding difference between computed data at 298K and 307 K (\( S_{uncorrected} = S_{exp} - [S_{307} - S_{298}]_{GRI} \)) so that comparison with literature become easier. It can be seen that the comparison with data from literature heat flux method is very good over the entire range of equivalence ratio.

5.2 Ethane-air

Measured adiabatic burning velocity values for ethane-air mixtures are compared with computational results from PREMIX using GRI 3.0 and experimental data from literature in figure 5. Experimental results of unstretched burning velocity for ethane-air available from literature were obtained using heat flux method [10,14] and transitional near zero stretch counterflow flames [24]. The present experiments were done at 307 K and 1 bar pressure. Here again the effect of difference in unburnt gas temperature is corrected using difference in computed burning velocities at 307 K and 298 K with PREMIX at the corresponding equivalence ratios. The present experimental results are found to be in good agreement with literature. Also, the predicted adiabatic burning velocities with GRI 3.0 mechanism are in very good agreement with experiments.

6 Results and Discussion

The present work focuses on the effect of dilution with CO\(_2\) and N\(_2\) on the adiabatic burning velocity of methane-air mixtures, and on the enhancement effect due to addition of C\(_2\)H\(_6\). Computationally obtained flame structures are used to reason out the behaviour of the above mixtures. All the additions and dilutions in the present work are expressed as a fraction of fuel mixture. For example, 40% dilution with CO\(_2\) means that the fuel is a gas mixture containing 60% methane and 40% CO\(_2\) by volume. The air used is 21% O\(_2\) and 79% N\(_2\) and the relative proportion of air in the fuel-air mixture is defined
by the equivalence ratio.

Measured adiabatic burning velocity of methane-air with CO₂ dilution in fuel is plotted in figure 6. Dilution with CO₂ is varied upto 60% in fuel. The recent results available in literature for CO₂-diluted methane burning in air were due to Clarke et al. [18] using constant volume method. For 20% dilution present results are in very good agreement with clarke et al with an unburnt gas temperature difference of 7 K. In constant volume method, burning velocity at 1 bar and 300 K were obtained by extrapolation and also stretch rate values are reported to be about 50-100 l/s. It should be noted that these results of Clarke et al [18] are not stretch corrected.

With 20% CO₂ dilution, adiabatic burning velocity shows a maximum reduction of 29%, while with 40% CO₂ dilution reduction is upto 45%. Reduction of burning velocity is less in lean side than at stoichiometric and rich side. Also peak burning velocity occurs at near-stoichiometric mixture for 20%, 40% and 60% CO₂ dilution. Predicted burning velocities shown in figure 6 are in good agreement with present measurements in the whole range of equivalence ratio and dilution degrees. Predictions also reproduce the trend of experiments with peak burning velocity at $\phi = 1.0$. This can be explained by gradual peak shift of equilibrium adiabatic flame temperature from $\phi \approx 1.05$ to $\phi \approx 1.0$ with CO₂ of 0% to 60% in methane-air mixtures as shown in figure 7. The equilibrium adiabatic flame temperatures were calculated using STANJAN [25].

Adiabatic burning velocity measurements in the mixtures of methane-air with 20% and 40% N₂ dilution in fuel are presented in figure 8. Constant volume results of Clarke et al. [18] for 20% N₂ in fuel is also presented in the same figure along with modeling results. Computational results are also seen to be in good agreement with experiments, except that on the rich side predictions are lower than the measurements.

Figure 9 compares the relative effect of CO₂ and N₂ dilution by upto 50% of fuel volume on the burning velocity methane-air at $\phi = 1.0$. The reduction in burning velocity of methane-air is more with CO₂ than with N₂ dilution of the fuel. This behaviour of burning velocity reduction can be attributed to the higher specific heat of CO₂ than that of N₂.

Measurements and predictions of adiabatic burning velocities were in reasonably good agreement for lean to moderately rich mixtures (i.e., upto $\phi = 1.2$), for different concentrations of the diluents. Therefore, the detailed flame structure from predictions were studied to gain a better insight of the effect of diluents on laminar burning velocities. Flame structures of freely propagating methane-air flame with 50 % N₂ and CO₂ dilution in fuel are presented in figure 10 for $\phi = 1.0$. Figures 10a and 10b provide profiles of temperature and mole fractions of stable species (CH₄, H₂O, O₂). Figures 10c and 10d provide
profiles of radical species (H, OH, O and CH₃) concentrations as a function of distance through the flame. Comparison of the temperature distributions in Figures 10a and 10b indicates the reduction of final burnt gas temperature from 2007 K to 1930 K. This behaviour is solely due to the CO₂ having larger specific heat of than that of N₂. For the present stoichiometric flames, the radical OH has the largest maximum concentrations in the flames, with H having somewhat smaller maximum concentrations, roughly 10-30% lower than OH. O and CH₃ have smaller concentrations, about less than half of OH concentration. The maximum concentration of OH, H and O in flames with N₂ dilution (figure 10c) are higher than those in flames with CO₂ dilution (figure 10d). Existing literature [26] indicates that a strong correlation exists between the burning velocity and maximum concentrations of H and OH radicals in reaction zone of premixed H₂/O₂/N₂ flames. Based on such a finding, study of figures 10c and 10d shows that the burning velocity can be expected to be higher for the case of dilution with nitrogen. Peak OH and H radical concentrations in 50% N₂ diluted CH₄-air flame are higher by 25% and 58% respectively over those in 50% CO₂ diluted CH₄-air flame.

Experiments with methane enriched with 25% ethane as fuel were performed and these results along with predictions were reported in figure 11. The agreement of predictions with experiments is very good except at very rich mixtures. The effect of enrichment of burning velocity is very small on the lean side while in stoichiometric to rich mixtures, burning velocity increases considerably due to addition of C₂H₆.

7 Conclusions

The heat flux method was used to determine adiabatic burning velocities in methane-diluent-air mixtures and methane-ethane-air mixtures at 1 bar and 307 K. Initially experimental measurements of adiabatic burning velocities in methane-air and ethane-air were found to be in good agreement with recent literature data (especially with those determined using heat flux method).

(1) Laminar burning velocity data for CO₂ and N₂ dilution of CH₄-air mixtures are presented. Burning velocity of methane-air reduces more with CO₂ than with N₂ dilution of the fuel.

(2) The peak burning velocity for CH₄/CO₂-air mixtures occur near to φ=1.0 for 20%, 40%, 60% by volume of CO₂. The gradual shift of adiabatic flame temperature from φ ≈ 1.05 to φ ≈ 1.0 with CO₂ dilution of 0% to 60% in methane-air mixtures is responsible for the shift in the equivalence ratio corresponding to peak burning velocity.

(3) Predicted flame structures showed that peak concentrations of OH, H and O radicals in reaction zone of flame are related to adiabatic burning.
velocities for CO$_2$/N$_2$ diluted methane-air mixtures.

4) Enrichment of 25% ethane to methane-air has very little effect on burning velocity for lean mixtures and considerable effect on rich mixtures.

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