Laminar flame velocity of components of natural gas

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ABSTRACT

Laminar burning velocities are important parameters in many areas of combustion science such as the design of burners or engines and for the prediction of explosions. They play an essential role in the combustion in gas turbines for the optimization of the nozzles and of the combustion chamber. Adiabatic laminar flame velocities are usually investigated in three types of apparatus which are currently available for that type of measurements: constant volume bombs in which the propagation of a flame is initiated by two electrodes and followed by shadowgraphy, counterflow-flame burners with axial velocity profiles determined by Particle Imaging Velocimetry, and flat flame adiabatic burners which consist of a heated burner head mounted on a plenum chamber with the radial temperature distribution measurement made by a series of thermocouples (used in this work). This last method is based on a balance between the heat loss from the flame to the burner required for the flame stabilization and the convective heat flux from the burner surface to the flame front. It was demonstrated that this heat flux method is suitable for the determination of the adiabatic flame temperature and flame burning velocity. The main hydrocarbon in natural gas is methane, with smaller amounts of heavier compounds, mainly species from C2 to C4. New experimental measurements have been performed by the heat flux method using a newly built flat flame adiabatic burner at atmospheric pressure. These measurements of laminar flame speeds are presented for components of natural gas, methane, ethane, propane and n-butane, as well as for binary and tertiary mixtures of these compounds representative of different natural gases available in the world. Results for pure alkanes were compared successfully to the literature. The composition of the investigated air/hydrocarbon mixtures covers a wide range of equivalence ratios, from 0.6 to 2.1 when it is possible to sufficiently stabilize the flame. Empirical correlations have been derived in order to predict accurately the flame velocity of a natural gas containing C_1 up to C_4 alkanes as a function of its composition and the equivalence ratio.

INTRODUCTION

The atmospheric issues and the problem of finite oil reserves have lead to an enhanced interest for the use of natural gas in combustion engines and power generation systems. The main hydrocarbon included in natural gas is methane, with smaller amounts of heavier compounds, mainly species from C_2 to C_4 . The fraction of heavier hydrocarbons can vary widely depending on the source of natural gas, e.g. from 2 to 17 % by volume [1].

The laminar flame velocities of methane/air, ethane/air and propane/air mixtures have already been investigated in the three types of apparatus which are currently available for that type of measurement : constant volume bombs (e.g. [2-7]) in which the propagation of a flame is initiated by two electrodes and followed by shadowgraphy, counteflow -flame burners (e.g. [8-14]) with axial velocity profiles determined by Particle Imaging Velocimetry, and flat flame adiabatic burners (e.g. [16][17][18][19][20]) which consist of a burner head mounted on a plenum chamber with the radial temperature distribution measurement made by a series of thermocouples (used in this work). There are much less data available for n-butane : Warnatz [21] reported some measurements in 1984, since then only two experimental studies have been published by Davis and Law (counterflow flame burner) [10] and by Bosschaart and De Goey (flat flame adiabatic burner) [19].

While the influence of the content in C_2 to C_4 compounds can be of importance for the reactivity of natural gas, only a few papers dealt with the laminar flame velocity of mixtures containing these species : in 2004, Liao [22] reported some measurements concerning the laminar flame speed of a Chinese natural gas (96.2% CH₄, 1.1% C₂H₆, 0.1% C₃H₈, 2.5% CO_2 and 0.1% others). In 2006, Huang [23] studied the same gas, then enriched with hydrogen (from 0 % to 100 %). In the present work, a new flat flame adiabatic burner was validated in the case of methane flame and measurements were performed for gaseous alkanes and mixtures representative of selected natural gases.

EXPERIMENTAL SETUP

Originally developed by de Goey and co-workers [15], the heat flux method was used in this study to measure laminar burning velocity. While other methods for the measurement of burning velocities such as bombs, counter-flow burners or burners with a stagnation plate need to correct the stretch effect by an extrapolation, heat flux method is the only one allowing a direct measurement. The main uncertainty remains then in the gas speed through the burner, i.e. in the gas flow control. The burner is presented in Figure 1. The apparatus consists of a burner head mounted on a plenum chamber. The burner head is a thin perforated plate made of brass of 30 mm diameter which is used to stabilize the flame. Each small hole of the plate has a 0.5 mm diameter and the pitch between the holes is 0.7 mm. Eight type K thermocouples of 0.5 mm diameter are soldered into the plate surface and are positioned at different distances and angles from the center to the periphery of the burner. The edge of the burner plate is heated at 353 K with water to keep the temperature of the burner plate constant and to heat up the mixture when it flows through the plate at a higher temperature than the unburned gas mixture. Indeed, the plenum chamber has a separate cooling system supplied with water at a temperature of 298 K. Thus, the heat gain of the unburned gas mixture can compensate for the heat loss necessary for stabilizing the flame, knowing that the measurement of the heat loss or gain is performed with the thermocouples.



Figure 1: Scheme of the burner

If the gas velocity is lower than the adiabatic flame burning velocity, the sum of the heat loss and heat gain is higher than zero. Then, the center of the burner plate is hotter than the periphery, and the flame is stabilized under sub adiabatic conditions. On the other hand, if the unburned gas velocity is higher than the adiabatic burning velocity, the center of the burner plate is cooler than the periphery and the flame is stabilized under super adiabatic conditions. Thus, when the temperature profile is flat, it means that no heat is lost or gained by the flame so that the flame becomes adiabatic. By changing the flow rate of the gas welocity to nullify the net heat flux so that the radial temperature distribution in the burner plate is uniform and equal to the temperature of the heating jacket. The flow rate at which the net heat flux is zero corresponds to the adiabatic flame burning velocity.

The complete experimental setup also includes a mixing panel which is assembled to supply gases from gas arrivals to the plenum chamber. Gas flows are regulated by Bronkhorst High-Tech mass flow controllers (MFC). As previously mentioned, two thermostatic water baths provide the water to the heating and cooling jackets of the burner. Measurement of the temperature distribution is based on differences between each pair of neighbor thermocouples connected as an electrical circuit of two wires and the burner plate. Gases were delivered by Messer and Air Liquide.

As the adiabatic laminar flame speed is found when the net heat loss is zero, the error is only dependent on a few factors: the error in the mass flow controllers (around 0.5%) which can lead to a maximum error of 2% in the equivalence ratio in the worst case, the error in the reading of thermocouples which could lead to an error of 0.5 to 1 cm/s in the laminar flame speed. The statistical error obtained by repeating measurements was below 2 cm/s for the higher burning velocities. The errors increase for low burning velocities close to the flammability limits and the measurements become difficult because of the flame instability.

RESULTS

The apparatus described above was used to study the laminar flame velocity of four hydrocarbons which are present in natural gas: methane, ethane, propane and n-butane. Experiments were performed at 298 K, at atmospheric pressure and using synthetic air (79 % N₂ and 21 % O₂). Then, three different surrogate mixtures were chosen to represent natural gases from various origins because of their low content in other species (higher hydrocarbons, CO₂, sulfur compounds...) and because they are different enough regarding their methane proportion (from 82% up to 90%).

Laminar flame velocity of pure compounds

Methane, ethane, propane and butane are all gaseous at 298 K and atmospheric pressure. Methane is the gas the laminar flame velocity of which was the most studied ([4][6][11][12][16][17][20]) since it is a reference gas and the major compound in natural gases. The velocities of ethane ([3][8][9][14][18][20]) and propane ([2][3][5][8][9][14][19]) were slightly less studied, particularly with the heat flux method, although they are relatively important for natural gases flame speeds prediction. Finally, the velocity of butane has been less studied than those of the three previous compounds ([10][13][19][21]), insofar butane is much less present in natural gases or in negligible quantities. Results concerning these four pure compounds are of great importance for the study of mixtures of these compounds representative of natural gases.

Methane

Figure 2 presents the evolution of the experimental laminar burning velocity of methane/air flames with equivalence ratio.

Being the gas for which the largest number of experimental studies concerning its laminar flame velocity is available. methane is then the most appropriate pure compound to validate our newly built experimental setup. Note that it is the hydrocarbon the laminar flame speed of which is the lowest with a maximum of about 38 cm/s for an equivalence ratio of 1.1. Each experiment has been repeated three times in order to test the reproducibility of the experiments. The overall accuracy of the burning velocity measurements was estimated thanks to a statistic law with 97.5% confidence level. With methane, it was possible to measure a laminar flame speed in an equivalence ratio range from 0.7 to 1.7. The results obtained with our apparatus are in good agreement with experimental data from the literature as shown in Figure 2, particularly in lean mixtures. When the mixture is rich (above 1.2), there is a more significant difference with recent literature data. Although one might think that the real equivalent ratio in flat flames, especially in rich mixtures, may be affected by entrainment of the ambient air, it had been tested and reported that it was not the case [17][18].



Figure 2: Measurement of the laminar burning velocity as a function of the equivalence ratio for methane/air mixtures. (Solid symbols = Heat Flux Method ; Open symbols = CounterFlow Method; Crosses = Combustion Bomb Method)

Ethane

Contrary to methane, ethane has the highest laminar flame speed with a maximum of 43.5 cm/s at an equivalence ratio of 1.1. It was possible to perform experiments over a wider range of equivalence ratios (from 0.6 to 2.1), as it was much easier to stabilize a flame with ethane than one with methane. The results are plotted in Figure 3. For ethane, there are also many results in the literature as previously mentioned and the results obtained in this work are in good agreement with them, although our measurements are slightly higher (between 3 and 7% approximately) close to the maximum. This consideration can be explained by the fact that it was necessary to use a different mass flow controller which is less accurate than the previous one (operating range of 28 nL/min instead of 3 nL/min) to obtain data at equivalence ratios from 1.0 to 1.25.



Figure 3: Measurement of the laminar burning velocity as a function of the equivalence ratio for ethane/air mixtures. (Solid symbols = Heat Flux Method ; Open symbols = CounterFlow Method ; Crosses = Combustion Bomb Method)

Propane

Measurements of the laminar flame speeds of propane were investigated too. The maximum obtained at an equivalence ratio of 1.1 is around 42 cm/s, and the propane profile is located between those of methane and ethane. Like in the ethane case, it was possible to cover a wide range of equivalence ratios, from 0.6 to 2.0. Experimental points from this work are in good agreement with those from the literature as shown in Figure 4, and it seems even to be that our measurements are in the closest agreement with the literature data.



Figure 4: Measurement of the laminar burning velocity as a function of the equivalence ratio for propane/air mixtures. (Solid symbols = Heat Flux Method ; Open symbols = CounterFlow Method ; Crosses = Combustion Bomb Method)

n-Butane

N-butane is the last pure compound which was studied in this work. As in other experiments presented burning velocity is the average of three measurements and the potential error is calculated thanks to a Student statistic law with 97.5% confidence level. The maximum is around 40 cm/s, just between methane and propane. Contrary to the other alkanes previously considered, there are only a few experimental data available in the literature, as previously mentioned. Results obtained in this work are broadly in agreement with the few points from the literature as shown in Figure 5. Note that there is a little gap between the point which is at an equivalence ratio of 1.15 and the one at 1.20. Indeed, there was a change of mass flow controller because of its scale, and this change is more visible for butane than for ethane or propane for them it was also necessary to use two MFCs.



Figure 5: Measurement of the laminar burning velocity as a function of the equivalence ratio for butane/air mixtures. (Solid symbols = Heat Flux Method ; Open symbols = CounterFlow Method ; Crosses = Combustion Bomb Method)

Natural Gas surrogates

The study of pure compounds has provided useful data to approach the study of hydrocarbons mixtures. According to Table 1, the composition of natural gases depends on its

Table 1: Composition of different natural gases (% volume) [1]

origin. As an example, natural gas from North Sea is mainly composed of methane (95.7%) and ethane (3.55%) whereas the natural gas from Abu Dhabi contains 82.07% of methane, 15.86% of ethane and 1.89% of propane. Some natural gases not only contain hydrocarbons but also species such as nitrogen, carbon dioxide and sulfur compounds. It is important therefore to study mixtures and especially binary and tertiary mixtures. In this work, we only studied the influence of the amount of ethane and propane along with methane on the laminar flame speed.

First, natural-gas like mixtures were investigated. Then, the study has been carried out with methane-ethane mixtures considering the variation of the proportion of ethane compared with methane at different equivalence ratios (from 0.7 to 1.5).

Natural gas-like mixtures

For the present work, we chose three surrogate mixtures with compositions as close as possible to those of three representative natural gases : Pittsburgh, Abu Dhabi and Indonesia (shaded columns in Table 1). These natural gases were represented by the following mixtures: 85% CH₄ and 15% C₂H₆ for Pittsburg, 82% CH₄, 16% C₂H₆ and 2% C₃H₈ for Abu Dhabi, and 90% CH₄, 6% C₂H₆ and 4% C₃H₈ for Indonesia.

Figure 6 presents the results obtained for the three natural gas surrogates. These experimental data show that the profiles for the three surrogates are lying in between those of methane and ethane. Note that the methane proportion seems to determine the laminar flame speed. Indeed, the proportion of propane is not of great importance as the difference between Pittsburg and Abu Dhabi gases is not significant. While the flame velocity profiles stand between those of pure methane and ethane, they are much closer to the methane profile.

-	Frigg	Lacq	Urengoï	Hassi R'Mel	Abu Dhabi	Indonesia	Matheson	Groningue	Abu Madhi	Pittsburgh
	(North Sea)	(France)	(Russia)	(Algeria)	(UAE)	Indonesia	(Ontario)	(Netherlands)	(Egypt)	(USA)
CH₄	95.7	69.2	98	83.5	82.07	89.91	96.62	81.3	92.8	85
C ₂ H ₆	3.55	3.5	-	7.9	15.86	5.44	2.32	2.9	4.1	14
C ₃ H ₈	0.04	1.1	-	2.1	1.89	3.16	0.54	0.4	1.2	-
i-C₄H ₁₀	0.04	0.7	-	1	-	1	-	0.2		
n-C ₄ H ₁₀	0.01				0.06	0.75	0.12		-	-
i-C₅H ₁₂	-	-	-	-	-	0.03	-	-	-	-
CO ₂	0.3	9.3	0.8	0.2	-	-	-	0.9	0.7	-
N ₂	0.4	0.6	1.2	5.3	0.05	0.04	0.3	14.3	0.4	1
Others (sulfur compounds,)	-	15.3	-	-	-	-	-	-	-	-



Figure 6: Measurement of the laminar burning velocity as a function of the equivalence ratio for natural gas/air mixtures. Natural gas from Indonesia (90% CH₄, 6% C₂H₆, 4% C₃H₈); Natural gas from Pittsburgh (85% CH₄, 15% C₂H₆); Natural gas from Abu Dhabi (82% CH₄, 16% C₂H₆, 2% C₃H₈).

Methane-ethane mixtures

In order to complete the study of hydrocarbons mixtures, methane-ethane mixtures were investigated and experiments were performed by considering the variation in the proportion of ethane relative to methane in the mixture. Figure 7 presents variations of the laminar flame velocities with the mole fraction of CH₄ in the binary mixture from 0 to 100 % for equivalence ratios ranging from 0.7 to 1.5. For all equivalence ratios, these variations are close to linear at least until 50% of CH₄, and then the decrease of the flame velocity with the increase of the CH₄ content is more pronounced, particularly for rich mixtures. For three equivalence ratios (1.0, 1.1 and 1.2), it was necessary to use another mass flow controller for oxygen because of the operating range of the apparatus in order to realize the entire profile; a slight step-shift of the curves is observed.



Figure 7: Measurement of the laminar burning velocity as a function of methane proportion relative to ethane for methaneethane/air mixtures.

CORRELATIONS

By considering the previous experimental results, it was possible to achieve a correlation for the laminar burning velocities of hydrocarbons flames, not only for the pure compounds, but also for the mixtures. In the pure compounds case, the following correlation, which was proposed by Gülder [23] for the laminar burning velocity of methane flames, was used in this work:

$$S_L = ZW\varphi^{\eta}e^{-\xi(\varphi-\sigma)^2} \tag{1}$$

This correlation has been fitted with the measurements made in this work. The values of the parameters are listed in Table 2. Z is a parameter that is equal to 1 for pure compounds but if the fuel is a mixture, it depends on its composition.

Table 2: Parameters obtained for the correlation (1)

Parameter	Methane	Ethane	Propane	Butane
Z	1	1	1	1
W (cm/s)	38.638	43.1041	42.9296	41.8775
η	-0.15	0.1163	-0.3099	-0.5973
ξ	6.2706	4.5959	5.2477	5.4277
σ	1.1	1.1	1.1	1.1

Correlations were derived covering a range of equivalence ratios between 0.6 (0.7 for methane) and 1.6. Indeed, they were not conclusive using the wider range of equivalence ratios covered by the experiments. The calculated results and the measured ones do not agree for equivalence ratios above 1.6. Despite this limitation, there is a very good agreement between calculated results and experimental data. This agreement is a little less convincing in the butane case around stoichiometry.

Mixtures

In the mixtures case, it was not possible to use the same correlation as for pure compounds because it does not take into account the proportion of another compound into methane. In fact, the Z parameter has to be modified to include the quantity of another gas in methane. Moreover, it was also necessary to insert an additional term in the exponent so as to simulate the shift of the maximum of the laminar flame speed dependence with the additional gas concentration. Thus, the following correlation was used as proposed by Coppens et al. [24] for the mixtures of methane with another gas :

$$S_L = (1 + \gamma \alpha^{\mathrm{T}}) W \varphi^{\eta} e^{-\xi (\varphi - \sigma - \Omega \alpha)^2}$$
(2)

Like for pure compounds, measurements from this work were used to fit this correlation. The parameters obtained for methane/ethane and methane /propane blends are listed in Table 3.

Table 3: Parameters obtained for the correlation (2) for mixtures

Parameter	$CH_4 + C_2H_6$	CH ₄ + C ₃ H ₈		
γ	0.2869	0.2759		
τ	0.5826	0.7901		
W (cm/s)	38.6385	38.6385		
η	-0.15	-0.15		
ξ	6.2706	6.2706		
σ	1.1	1.1		
Ω	-0.0182	-0.0514		

Results are in good agreement with experimental values, with a deviation below 2 cm/s for equivalence ratios between 0.7 and 1.5, even if they are not as conclusive as those obtained for pure compounds. Indeed, they are in very good agreement for lean and rich mixtures, but the curve is beyond experimental results near stoichiometry. Note that calculations were performed using experimental values for a range of equivalence ratios between 0.8 and 1.3.

From the correlations for binary mixtures, it was possible to combine the results to propose for a tertiary mixture the following correlation :

 $S_L = (1 + \gamma_1 \alpha_1^{\tau_1})(1 + \gamma_2 \alpha_2^{\tau_2})W\varphi^{\eta}e^{-\xi(\varphi - \sigma - \Omega_1 \alpha_1 - \Omega_2 \alpha_2)^2}$ (3) with α_1 and α_2 the proportion of ethane and propane respectively in methane, $\gamma_1, \tau_1, \Omega_1$ the parameters calculated for a methane-ethane mixture, and $\gamma_2, \tau_2, \Omega_2$ the parameters calculated for a methane-propane mixture. The proposed correlation has been fitted with the measurements made in this work for natural gas-like mixtures. Results are presented in Figure 8.



Figure 8: Correlation of the laminar burning velocity as a function of the equivalence ratio for natural gas-like/air mixtures. Points : experiments, lines : correlation. Natural gas from Pittsburgh (85% CH₄, 15% C₂H₆), Abu Dhabi (82% CH₄, 16% C₂H₆, 2% C₃H₈) and Indonesia (90% CH₄, 6% C₂H₆, 4% C₃H₈)

Correlation results are in good agreement with the experimental ones. As for methane-ethane mixtures, the agreement is better for lean and rich regions but less satisfactory near stoichiometry. Indeed, it's not surprising to observe the same behavior in the two cases insofar as

correlation for natural gas-like mixtures is based on correlation for methane-ethane and methane-propane mixtures.

CONCLUSION

The heat flux method has been used for measuring adiabatic laminar burning velocities of pure alkanes from methane up to n-butane. It appeared that methane has the lowest burning velocity, ethane the highest, and that other linear alkanes lies in-between. Results are in very good agreement with the literature data. Binary mixtures and blends representative of different natural gases have been also investigated. Empirical correlations have been deduced from the experimental results which allow the evaluation of the flame speed of natural gases at 298 K under 1 atm from their chemical compositions.

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