DETERMINATION OF LAMINAR BURNING VELOCITY OF METHANE/AIR FLAMES IN SUB ATMOSPHERIC ENVIRONMENTS

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Abstract

The global energy demand enhances the environmental and operational benefits of natural gas as an energy alternative, due to its composition, mainly methane (CH₄), it has low polluting emissions and benefits in energy and combustion systems. In the present work, the laminar burning velocity of methane was determined numerically and experimentally at two pressure conditions, 0.85 atm and 0.98 atm, corresponding to the city of Medellín and Caucasia, respectively, located in Colombia. The environmental conditions were 0.85 atm, 0.98 atm, and 295 ± 1 K. The simulations and experimental measurements were carried out for different equivalence relations. Experimental laminar burning velocities were determined using the burner method and spontaneous chemiluminescence technique, flames were generated using burners with contoured rectangular ports to maintain laminar Reynolds numbers for the equivalence ratios under study and to reduce the effects of stretch and curvature in the direction of the burner's axis. In general, the laminar burning velocity fits well with the numerical results. With the results obtained, a correlation is proposed that relates the laminar burning velocity with the effects of pressure, in the form $S_L = aP^b$, where *a* and *b* are model constants. Sensitivity analysis was performed using the GRI-Mech 3.0 mechanism which showed that the most sensitive reaction was H+O₂ = O+OH (R38). Additionally, it was found that the reactions H+CH₃ (+M) = CH₄ (+M) (R52), 2CH₃ (+M) = C₂H₆ (+M) (R158), and O+CH₃ = H+CH₂O (R10) dominate the consumption of CH₃ which is an important radical in the oxidation of methane, this analysis is carried out for equivalence ratios of 0.8 and 1.0, and atmospheric pressures of 0.85 atm and 0.98 atm.

Keywords: Laminar burning velocity, Methane, Sub atmospheric conditions, Pressure effect.

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1. Introduction

In recent years, due to the increase in world energy demand and trying to satisfy this demand with the least damage to the environment, natural gas (generally 100 % methane is assumed, CH_4) has become a fuel alternative in domestic, commercial, and industrial applications. This has been mainly due to the fact that natural gas produces fewer polluting emissions per unit of energy compared to other fossil fuels. Several studies agree that lean-blend natural gas applications are particularly interesting because they have the potential to reduce gas emissions NO_X and improve thermal efficiencies in combustion equipment.

The laminar burning velocity, S_L , is a characterization parameter of a laminar premix flame since it provides physical-chemical information of the combustible mixture. To define the laminar burning velocity, it is considered in relation to one-dimensional, stable and unstretched flames. Additionally, these considerations are made in most theoretical combustion models, such as the CHEMKIN code, used in this study to determine the theoretical laminar burning velocity [1, 2]. Laminar burning velocity is an important parameter in combustion studies that allows the study and prevention of undesirable phenomena such as extinction, flame detachment, and flashback, and provide important characteristics such as flame stability, heat release rates, and turbulent flame velocities [3, 4]. Determining the value of this property is of utmost importance and is a starting point since several combustion phenomena depend on the laminar burning velocity, such as the structure [5, 6], and turbulent burning velocity [7–9], determine flame instabilities and determine extinction and stabilization phenomena. Also, this parameter is used to prevent explosions and design gas burners [10]. For this reason, much work has been carried out on the laminar burning velocity for methane and other fuels [11], seeking to establish stable and reliable operating ranges [12]. The authors in reference [10] carried out studies of the laminar deflagration rate to develop analytical correlations that allow their calculation and are used in motor simulations. Correlations are generally preferred to detail chemical kinetic models to save computational time. The authors in reference [12] investigated the laminar burning velocity of low calorific blast furnace gas using a constant volume combustion chamber and a CHEMKIN package. Numerical experiments and simulations were carried out using different models and kinetic mechanisms at initial pressures of 0.99-1.97 atm, initial temperatures of 303 to 453 K and equivalence ratios of 0.8 to 1.6. The composition of the fuel was modified considering the variation of the volume fraction in the blast furnace gas with calorific values of 3.27-4.03 MJ/m³. The results show that the laminar burning velocity increase with the decrease of the initial pressure and the increase of the initial temperature, and the Gri-Mech 3.0 mechanism gives very good predictions among the six kinetic mechanisms studied.

Several studies have been conducted on S_I under sub atmospheric conditions [13–16]. The authors in reference [15] carried out experimental measurements of adiabatic combustion rate in methane + hydrogen + air flames are presented using the Heat Flux method. The undrawn flames were stabilized in a 0.2 atm to 0.99 atm perforated plate burner. The equivalence ratio ranged from 0.8 to 1.4. Low pressure measurements in CH_4 + air flames made earlier were also accurately reproduced. The pressure dependencies of the combustion rates for the three fuels studied could be approximated by an empirical exponential correlation. The authors in reference [16] examined the propagation of the laminar spherical flame for H_2 -air mixtures. The hydrogen content of the H₂-air mixtures was varied between 4 % by volume and 80 % by volume. The initial pressure was between 0.025 atm and 0.99 atm. The initial temperature was 285-295 K. The laminar burning velocity for a wide range of initial pressures and hydrogen concentration were evaluated for the mixtures. For H₂ content between 40 mol % and 45 mol %, the flame speed was found to be maximum. To characterize the pressure dependence of the laminar burning velocity, the general reaction order was evaluated for different hydrogen contents by changing from the initial pressure. Reaction order less than 2 with a negative pressure dependence was found for lean mixtures. The rest of the mixtures have a positive influence of the pressure on the laminar burning velocity because the reaction order n > 2.

In general, it has been found that the global order of the reaction (*n*) is the parameter responsible for the dependence of S_L on pressure. Furthermore, an analytical expression for the laminar burning velocity as a function of pressure is $S_L \alpha P^{(n-2)/2}$ [17]. This means that pressure decreases the

laminar burning velocity when the general order of the reaction is n < 2, and the reverse is the case when n > 2. The value of n depends on the oxidant mixture of the fuel. The authors in reference [18] carried out measurements of S_L of mixtures of CH₄/air at different pressures. In all the mixtures they studied, n was less than 2; therefore, S_L always decreased as pressure increased. The effect of pressure was greater in the sub atmospheric range, where they obtained a decrease in the value of S_L 27 % when they changed from 0.5 atm to 1 atm in the stoichiometric case. These results are in line with those presented by other authors in the following references [19, 20].

To present the effect of pressure on S_{L_2} some authors have proposed correlations where pressure is directly related to S_L . In the work [21] the authors proposed a relationship between S_L and pressure (P) using the burner method in three equivalence ratios ($\phi = 0.9$, $\phi = 1.0$, $\phi = 1.1$). Their study shows that S_{L_2} decreases as pressure increases and the exponent that best fits all pressure data are -0.5. They proposed an approximate relationship for these data as a function of pressure: $S_L = 0.34 (P/P_0)^{-0.5}$, where P_0 is the reference atmospheric pressure in MPa. Ubbelohde and Koelliker [22] pose the following correlation for S_L as a function of pressure for premixed methane and air flames $S_L = S_{L0} (P / P_0)^{\beta_1}$, where S_{L0} is the reference laminar burning velocity, P is the premix pressure, P_0 is the reference pressure and β_1 is a dimensionless constant of the model. Agnew and Graiff [22] propose the following correlation for S_L as a function of pressure for premix flames of methane and air, $S_L = S_{L0} \left[1 + \beta_2 \log(P / P_0) \right]$, where S_{L0} is the reference laminar burning velocity, P is the premix pressure, P_0 is the reference pressure and β_2 is a dimensionless constant of the model. Other correlations have been raised, not only including the effect of pressure but also the effect of the temperature of the premix, experimental results [23] for methane-air flames describe the behavior of S_L as a function of temperature, pressure, and equivalence ratio from the reference laminar burning velocity $S_L(P^0, T_1^0)$, $S_L(P,T_1) = S_L(P^0, T_1^0) (P / P^0)^{\alpha P} (T_1 / T_1^0)^{\alpha T}$, where P^0, T_1^0 are the reference pressure and temperature, respectively, and αT , and αP are model constants that depend on the equivalence ratio.

The present study seeks to determine the laminar burning velocity for two pressure conditions 0.85 atm and 0.98 atm, providing reliable theoretical, experimental, and numerical data for these pressure conditions. Also, an analysis of the effect of sub atmospheric pressure on the laminar burning velocity is included, through sensitivity analysis and considering the reactions that are most important in this effect, and subsequently, propose a correlation for the pressure conditions and the equivalence ratio studied.

2. Materials and methods

2. 1. Experimental methodology

To measure and experimentally determine the laminar burning velocity of methane, the burner method was implemented. The flames were generated using burners with contoured rectangular ports to maintain the laminar Reynolds numbers for the equivalence relationships under study and to reduce the effects of stretching and curvature in the direction of the burner axis [24]. This method and burner design allow to obtain triangular flames with straight edges and a uniform exit velocity profile in the burner, in **Fig. 1** it is possible to see the procedure to obtain the images by the burner method.



Fig. 1. Profile of the experimental flames obtained by the burner method for methane at 0.98 atm, 295 K, and $\phi = 1.0$: *a* – Burner method; *b* – ICCD Image

An ICCD camera (PI-MAX; Princeton Instrument) was used to measure chemiluminescence. To capture the light emission of CH (CH*) radicals, the lens was equipped with an interference filter with a central wavelength of 430 nm. The maximum mean width (FWHM) was 11.02 nm and the minimum transmissivity was 45 %. In chemiluminescence measurements, CH* images were taken 25 images in each condition and signal/noise ratios were less than 10 % of maximum intensity. The complete experimental setup used to obtain the images is shown in **Fig. 2**. More details on the experimental setup can be found in previous studies [24, 25].



Fig. 2. Experimental setup

The experiments were carried out at atmospheric conditions of 295 ± 1 K, the average relative humidity of 65 ± 5 %, and an atmospheric pressure of 0.85 atm and 0.98 atm corresponding to the city of Medellín and Caucasia, respectively, located in Colombia. The equivalence ratios in this study ranged from 0.8 to 1. To prepare the methane/air mixtures and the equivalence ratios of the present study, rotameters calibrated specifically for each gas were used, and the total error is estimated to be less than 2 %. Error analysis was used to determine the errors in the laminar burning velocity measurements and was based on the measurement errors of the average velocity of the unburned gases at the burner outlet, the angles of the flames (**Fig. 3**), the area of the burner port, and air and fuel flow.

After determining the angle of the flame and knowing the velocity of the unburned gases at the burner outlet, U, the relation of (1) is used to determine the laminar burning velocity and supported in **Fig. 3**.

$$S_L = U\sin\theta. \tag{1}$$

Where S_L is the laminar burning velocity of the premix, U is the average speed of the mixture at the burner port, and θ is the angle of the flame front of the premix.



Fig. 3. Flame profile and burner method

2.2. Numerical calculation

The Premix subroutine of the Chemkin Pro software [24, 25] used to calculate the laminar burning velocity, requires an initial assumption by the user of the species profiles (reactants, products, intermediate species) and the temperature that will serve as a starting point to begin troubleshooting. It depends on the correct assumption of these profiles that the program reaches a final solution, as well as that it is quickly. Within the calculation of incorporates the Thermo diffusive effect, the multicomponent transport calculation for the transport properties of the mechanism species.

The maximum number of points or nodes that the mesh must-have in the final solution must be around 1000 so that the temperature of the products agrees with the adiabatic flame temperature. The value of 1200 is set in case the entered GRAD and CURV values are so low that the solution requires more than 1200 points to satisfy the entered GRAD and CURV values. However, as it is known that a solution with 1000 points is sufficiently precise, the program will stop the simulation at 1200 points and show the solution corresponding to that number of points, even though the mesh does not satisfy the GRAD and CURV values.

The magnitude of the initial mesh is defined in a starting point corresponding to 0 cm and has an endpoint of 1 cm, however, to refine the mesh and give a correct prediction this mesh must be refined, this procedure is carried out by expanding the mesh domain until reaching a domain from -2.0 cm to 10.0 cm and the GRAD and CURV are defined to obtain a greater number of nodes in the solution. To expand the domain, continuations are used where the mesh is gradually refined, in this study 3 continuations were carried out, which correspond to those presented in the **Table 1**.

Table 1

Continuations

Davamatar	Continuation		
r ar ameter	1	2	3
Starting Axial Point	-0.5	-2	-2
Ending Axial Point	5	8	10
Adaptive Grid Control Based on Solution Gradient (GRAD)	0.8	0.7	0.009
Adaptive Grid Control Based on Solution Curvature (CURV)	0.8	0.7	0.018

The numerical results were performed in the Premix subroutine of the Chemkin Pro [26, 27] using for the simulations the GRI-Mech 3.0 mechanism [28] detailed reaction mechanism with 325 reactions and 53 species, which has been extensively verified and optimized for the representation of flames and ignition of natural gas.

3. Results and discussion

3. 1. Laminar burning velocity and validation study

Laminar burning velocity was determined numerically and experimentally for the atmospheric conditions corresponding to the city of Medellín (0.85 atm, 295 K) and for the city of Caucasia (0.98 atm, 295 K).

In **Fig. 4** it is observed that both numerical and experimental results for the pressure of 0.98 atm are always lower than for pressure of 0.85 atm. The mechanism, for pressure of 0.98 atm, has a very good fit between the experimental and numerical results. In general, for the pressure of 0.85 atm, there is a good fit, with a maximum difference of 6 % between the numerical and experimental values, this maximum value corresponds to the equivalence relation of $\phi = 0.8$. For an equivalence relation of $\phi = 1.0$. The following results are obtained, for the pressure of 0.85 atm the laminar burning velocity is 40.04 cm/s while for the pressure of 0.98 atm the laminar burning velocity is 36.53 cm/s, which corresponds to a decrease of approximately 9 % of the laminar burning velocity corresponding to the pressure of 0.98 atm concerning the pressure of 0.85 atm.

For the conditions of 0.98 atm and an equivalence ratio of $\phi = 0.8$, it was not possible to establish a value for the laminar burning velocity because the detachment phenomenon was present, making it impossible to have a stable flame, for which it was decided to use a value reported in the work [29]. These laminar burning velocity values will be used later in other analyzes and calculations. Additionally, the results presented by other authors [3, 4, 30] were included in **Fig. 4**.



Fig. 4. Laminar burning velocity of methane at atmospheric conditions of 0.85 atm, 0.98 atm and 1 atm

According to the experimental values presented in **Fig. 4**, the range of values is like those reported by other authors, validating the results presented in the present study.

3. 2. Sensibility analysis

A sensitivity analysis of the most important reactions on the laminar burning velocity was carried out to establish which species and reactions generate the decrease in the laminar burning velocity with pressure increases.

In this sensitivity analysis, the 15 most influential reactions were taken for atmospheric pressure corresponding to 0.98 atm and 0.85 atm.

Fig. 5, **6** show the results for methane at conditions of 0.85 atm and 0.98 atm. The sensitivity analysis explains the behavior of the formation of intermediate chemical species, which is an important factor in the sensitivity of the laminar burning velocity.

According to the results of the sensitivity analysis of the kinetic parameters, the recombination reactions (R35, R43, R45, R52, R53, R55, R98, R158) have the greatest negative effect on the laminar burning velocity. And the reactions that surround the radical HO₂ have a negative effect too (R35: $H+O_2+H_2O = HO_2+H_2O$ and R45: $H+HO_2 = O_2+H_2$). The branching reaction $H+O_2 = O+OH$ (R38) has the largest positive value of sensitivity and was higher for the pressure corresponding to 0.98 atm and a slightly lower value for the pressure of 0.85 atm. Similar analyzes are reported by Dong and others [31] for the combustion of methane. On the other hand, the recombination reaction $H+CH_3$ (+M) = CH₄ (+M) (R52) negatively affects the reactivity of methane for both pressure conditions.

According to the sensitivity analysis, the decrease in the laminar burning velocity with the pressure increases is associated with the increase in the radical production HO_2 . The molar fraction

of the radical was higher when the pressure is 0.98 atm. Similar behavior was observed for lean and stoichiometric conditions. The radical HO₂ is a very inactive vehicle compared to other radicals, causing the overall reaction rate to decrease.







The highest fraction of the radical HO₂ with higher pressure can be associated with the reactions R35 ($H+O_2+H_2O = HO_2+H_2O$) and R45 ($H+HO_2 = O_2+H_2$) for a dose of 1.0, and the reactions R35 ($H+O_2+H_2O = HO_2+H_2O$), R36 ($H+O_2+N_2 = HO_2+N_2$), R45 ($H+HO_2 = O_2+H_2$), R168 ($HCO+O_2 = HO_2+CO$) for an equivalence ratio of 0.8, particularly the first one, which improves with increasing pressure and takes a more dominant role in the reaction process [30].

The rate of production of the radical was calculated HO₂ total reaction rate (**Fig. 7**). As can be seen, the production of HO₂ was higher at atmospheric pressure, and reactions R35 and R45 had a great influence on this increase, confirming the above.



Fig. 7. Total production rate of HO₂: $a - \phi = 1.0$; $b - \phi = 0.8$

Consequently, more H radicals are consumed by both reactions at a lower pressure, reducing the availability of this radical. This directly affects the reaction rate of the reaction R38 (H+O₂ = O+OH), which has the most positive effect on laminar burning velocity according to sensitivity analysis. In this way, the net effect is a reduction in the laminar burning velocity due to the lower availability of H radicals to produce OH through the R38 reaction. Instead, the generation of OH is carried out by the following reactions:

$$H_2O_2 + M \rightarrow 2OH + M(-R85),$$
$$HO_2 + H_2O \rightarrow OH + H_2O_2(-R89).$$

The decrease in the laminar burning velocity with increasing pressure is due to the higher production of species HO_2 at sea level atmospheric conditions, which increases the consumption of radicals *H*. This changes the reaction path of radical production OH, leading to a slower net reaction process.

Furthermore, according to the study carried out by [19], the species CH_3 has a significant effect on the laminar burning velocity. Fig. 8, 9 represent the flows as a function of the

pressure of the following reactions R52 (H+CH₃(+M) = CH₄(+M)), R158 (2CH₃(+M) = C₂H₆(+M)), and R10 (O+CH₃ = H+CH₂O), and the profile of the species is traced on a secondary axis CH₃.

Reactions R52, R158, and R10 dominate the consumption of CH_3 which is an important radical in the oxidation of methane, this analysis is carried out for equivalence of 0.8 and 1.0, and atmospheric pressures of 0.85 atm and 0.98 atm, and in general the following conclusions are reached:

- the R10 reaction that leads to higher consumption of CH₃ for $\phi = 1.0$ for the pressures of 0.85 atm and 0.98 atm has a considerable decrease in consumption for a $\phi = 0.8$;

- the reaction rate of R158 decreases in the same order of R52 since fewer radicals CH₃ are produced in lean mixture;

- the evolution of R10 remains qualitatively the same for the two pressure conditions, however, the rate of consumption of the radical CH₃ through reaction R158 and R52 for a $\phi = 0.8$ to $\phi = 1.0$ and the same pressure condition decreases considerably;

- the molar fraction of CH₃ increases slightly for a condition of 0.85 atm compared to 0.98 atm for $\phi = 1.0$ and this same behavior is followed for $\phi = 0.8$;

– when comparing for the same pressure and for a dose of $\phi = 1.0$ and $\phi = 0.8$ the mole fraction of the radical CH₃ is approximately 1.5 times for a $\phi = 1.0$ regarding $\phi = 0.8$ for both pressure conditions.



Fig. 8. Molar fraction of CH₃ and consumption rate of CH₃ for reactions R52 (H+CH₃(+M) = CH₄(+M)), R158 (2CH₃(+M) = C₂H₆(+M)), and R10 (O+CH₃ = H+CH₂O) for $\phi = 1.0$: a - 0.85 at; b - 0.98 atm



Fig. 9. Molar fraction of CH₃ and consumption rate of CH₃ for reactions R52 (H+CH₃(+M) = CH₄(+M)), R158 (2CH₃(+M) = C₂H₆(+M)), and R10 (O+CH₃ = H+CH₂O) for $\phi = 0.8$: a - 0.85 atm; b - 0.98 atm

3. 3. Correlation for laminar burning velocity

An analysis was carried out for different pressures corresponding to heights above sea level from 0 masl to 3000 masl equivalent to a pressure range of 1 atm to 0.71 atm respectively, for the laminar burning velocity of methane for a range of equivalence ratios between 0.8 to 1.2 with the detailed mechanism GRI-Mech 3.0 [26, 27], where the results presented in **Fig. 10** were obtained, it should be clarified that the results presented for equivalence ratios of 1.1 and 1.2 were included in the study to give a wider range in **Fig. 10** presentation.

For each curve, an exponential approximation was carried out with higher values for the approximation value (R^2) of 0.9995, and a relationship like the one shown in (2) was obtained:

$$S_L = aP^b, (2)$$

where *a* and *b* are constants, and *P* is the pressure, the values of the constants are presented in Table 2.

According to these results, the aforementioned is reaffirmed, as the pressure increases, the laminar burning velocity decreases.

The laminar burning velocity was determined numerically and experimentally for atmospheric conditions corresponding to 0.85 atm and 0.98 atm, and a temperature of 295 ± 1 K, providing reliable theoretical, experimental, and numerical data for these pressure conditions.



Fig. 10. Laminar burning velocity of methane as a function of pressure for different equivalence ratios

Table 2	
Values for the constants a and b	5

values for the constants a and b		
Equivalence relation	<i>a</i> *	b
0.8	26.221	-0.378
0.9	32.538	-0.346
1.0	36.517	-0.325
1.1	37.008	-0.323
1.2	32.668	-0.365

*Note:** – *units of* $a = \left[cm/s(atm)^b \right]$, b = [dimensionless]

In general, the effect of pressure on the laminar burning velocity was studied, it was found that for the pressure of 0.85 atm, S_L increases compared to pressure at 0.98 atm, with the increase in S_L it is expected that the stability of the flame improves, decreasing the tendency to blow off compared to flames at 0.98 atm. Additionally, it is expected that the specific power of the flame will increase and consequently the release of heat per unit volume, constituting, for example, a valuable advantage for the design of thermal equipment. However, it is necessary to carry out future studies that include lower ranges of sub atmospheric pressure, finding an experimental limitation, to achieve wider ranges of sub atmospheric pressure. In **Fig. 10** a linear increase in laminar deflagration velocity with pressure is observed, however, it should be studied in greater detail as it can be a useful tool to determine theoretical correlations of laminar deflagration velocity with pressure.

From these results, reliable ranges of operation of combustion equipment operating at heights above sea level corresponding to atmospheric pressures of 0.85 atm and 0.98 atm were determined, these results are important when it is necessary to design atmospheric burners for different industrial processes. and domestic, in addition these results are a fundamental basis for future studies on turbulent deflagration speeds in sub atmospheric conditions. These results also provide a reliable data base for future work on combustion engines, where to perform numerical simulations, it is essential to know values of laminar deflagration velocity. Knowing the behavior of the laminar flame speed, as a function of the unburnt mixture strength, temperature, and pressure, is essential for an efficient and reliable simulation of the combustion process that occurs in

a spark-ignition engine. As future work it is important to analyze more sub atmospheric pressure conditions and have a greater base for the different applications in combustion equipment.

4. Conclusions

1. The sensitivity analysis showed that the reaction that has the greatest influence on the laminar burning velocity is the chain branching reaction $H+O_2=O+OH$ (R38), this reaction has become one of the main elemental reactions in combustion chemistry and the most reactive.

2. The analysis of the effect of pressure on the laminar burning velocity showed that the radical with the greatest influence is the radical CH_3 .

3. The molar fraction of CH₃ increases slightly for a condition of 0.85 atm compared to 0.98 atm for $\phi = 1.0$ and this same behavior is followed for $\phi = 0.8$. When comparing for the same pressure and for an equivalence ratio of $\phi = 1.0$ and $\phi = 0.8$ the mole fraction of the radical CH₃ is about 1.5 times for a $\phi = 1.0$ regarding $\phi = 0.8$ for both pressure conditions.

4. An empirical correlation was proposed for the laminar burning velocity and the pressure effects in the proposed equivalence ratios, the correlation has the form $S_L = aP^b$, where *a* and *b* are model constants and depend on the corresponding equivalence ratio between 0.8 and 1.2.

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