

COMBUSTION OF HYTHANE DILUTED WITH CO₂

by

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With increasing concern about energy shortage and environmental protection, improving engine fuel economy and reducing exhaust emissions have become major research topics in combustion and engine development. Hythane (a blend of hydrogen H₂ and natural gas NG) has generated a significant interest as an alternative fuel for the future. This paper describes an experimental study of the effects of CO₂ addition on the stability of a turbulent jet diffusion NG-H₂ flame. The mole fraction of hydrogen (% H₂) in NG-H₂ mixture was varied from 0% to 50%. The equivalence ratio of the hythane/CO₂/air mixture was kept at stoichiometry. The results show that the lift-off height increases with the addition of CO₂ at various % H₂ content in hythane. However, we observe that with 20% H₂, we can obtain a stable flame diluted with 40% CO₂, while for 0% H₂, the flame is blown out above 20% CO₂. This means that the limits of flame blowing out are pushed with the additions of H₂. Moreover, the results show that for %H₂ content in NG-H₂ fuel up to 10%, the addition of CO₂ could produce lifted flame if the % CO₂ is low. At higher % CO₂ dilution, flame would remain attached until blow-out. This is mainly due to the fact that the dilution leads to ejection velocities very high but reactivity of the mixture does not change so the flame tends to stretch.

Keywords: *hythane, hydrogen enrichment of natural gas flames, CO₂ dilution, flame stability*

Introduction

There is growing worldwide interest to move towards a hydrogen-based economy due to many potential advantages of hydrogen fuel over conventional fossil fuels. A part from being environmentally clean, hydrogen represents potentially an unlimited source of energy because it can be produced from abundant sources, it can be readily formed through water electrolysis, as well as from fossil fuels through partial oxidation and reforming. In addition, it has higher energy per unit mass (about 2.6 times that of gasoline), and superior characteristics than hydrocarbon fuels such as wider flammability limits, very low density, higher molecular diffusivity, higher flame speed, and low ignition energy [1, 2].

However, due to its high flammability and low volumetric energy density, many important issues pertaining to hydrogen safety and storage are still being addressed. Numerous efforts are currently underway focusing on the use of hydrogen in various propulsion and energy

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applications, including fuel cells and hydrogen based IC engines [3-5]. In this context, blending hydrogen with hydrocarbon fuels also represents a promising approach to increase the use of hydrogen, and improve the emission and performance of various combustion systems [1, 6, 7].

Hythane (a blend of hydrogen, H₂ and natural gas, NG) is expected to play an important role in future energy production. The researchers are interested only recently on the combustion of natural gas/hydrogen mixture. Only a few studies have been published on this subject. As methane is the main component of natural gas, the literature on the study of methane/hydrogen mixtures will be expanded. The mixture (80% methane/20% hydrogen) is the most studied because it is close to the industrial Hythane.

Auto-ignition times of methane/hydrogen mixture were measured by varying pressure, equivalence ratio and temperature, in rapid compression machine and shock tube [8-10]. Huang *et al.* [8] have measured, in shock tube, the auto-ignition times of two mixtures stoichiometric of methane/(15 or 35 mol%)H₂/air at 16 and 40 atm, and between 1000 and 1300 K. The effect of hydrogen is very visible at high temperature, for example, the auto-ignition delays of the mixture CH₄/35% H₂ at 1300 K and 40 atm are reduced by a factor of 1.5 compared to self-ignition delays of methane. The same trend is observed at 16 atm with a wide effect of hydrogen. In contrast, the promoting effect of hydrogen seems to decrease when increasing pressure or decreasing temperature ($T < 1200$ K). This effect is very light on the ignition of the mixture CH₄/H₂ and similar under these conditions.

Hadj Ali [9] has measured, in shock tube, the auto-ignition times of CH₄/10% H₂ mixtures for equivalence ratios of 0.5 and 1, at 14 atm, and for temperatures between 1250 and 2000 K. An important effect of equivalence ratio for CH₄/10% H₂ mixture is observed at 1625 K but not at 1777 K.

Recently, Gersen *et al.* [10] have measured the auto-ignition times of CH₄/H₂ mixtures in a rapid compression machine. The auto-ignition times were measured for equivalence ratios of 1.0 and 0.5 at 10 and 69 atm, at temperatures of 950-1060 K after compression and fractions of hydrogen of 0, 10, 20, 50, and 100% in the fuel. For mixtures of hydrogen content less than 20% of the fuel, the effect of hydrogen on the auto-ignition times is low, but the auto-ignition time increases if the hydrogen fraction is above 50%. In addition, at high temperatures, the effect of hydrogen is more pronounced at low temperature and high pressure. Surprisingly, equivalence ratio does not seem to have any effect on the time of auto-ignition, in fact, the same values are found for lean and stoichiometric mixtures of 50% H₂/50% CH₄.

Lift-off and blow-out stability parameters have significant meaning in the diffusion flame modeling and also have strong implication in safety consideration and hazard assessment. Therefore study of effects of various additives on the stability of natural gas/hydrogen jet flames require further investigations. A brief overview is given here on the main results obtained regarding H₂-diluted and CO₂-diluted flames properties.

Choudhuri and Gollahalli [11] performed an experimental investigation on turbulent NG-H₂ jet diffusion flame and observed a continuous reduction in the flame length, a reduction in the soot concentration and emission index of CO (EICO), but an increase in EINO with hydrogen addition.

Ilbas *et al.* [12] studied experimentally the effect of hydrogen enrichment on the natural gas laminar flame properties. They stated that increasing the hydrogen concentrations in mixture causes an increase in the resultant burning velocity and also widening the flammability limits. Kumar and Mishra [13] investigated experimentally a laminar LPG-H₂ jet diffusion flame. They performed that a subsequent reduction in the flame length was observed for the high

hydrogen concentrations. El -Ghafour *et al.* [14] investigated experimentally the effect of hydrogen addition on combustion characteristics of NG-H₂ hybrid fuel turbulent diffusion flame at a fixed Reynolds number (4000). They found that the addition of hydrogen sustains a progressive improvement in flame stability and reduction in flame length for relatively high hydrogen concentrations. On other side, their results shows an increase in NO and CO concentration. Yon and Sautet [15] studied experimentally the influence of hydrogen addition and the equivalence ratio on the structure of a turbulent diffusion oxy-flame of hythane (NG-H₂). They confirmed that, in oxy-fuel burner, hydrogen addition and a lean condition ($\phi < 1$) allows an early combustion, a better flame stability and an amount decrease of fuel consumption.

The effects of CO₂ addition on the burning velocity have been explored to discuss the validity domain of detailed kinetic mechanisms. However, existing studies are limited to highly diluted oxy-fuel mixtures or to air-fuel mixtures in which small CO₂ quantities are added. The chemical effect of CO₂ replacement of N₂ in air on the burning velocity of CH₄ and H₂ flames was numerically investigated. The relative importance of the chemical effect of CO₂ on the burning velocity increases as more CO₂ is added to replace N₂ in air [16].

Mazas *et al.* [17] investigated experimentally and numerically the effect of CO₂ and H₂O (steam) additions on laminar flame speed of oxygen-enriched methane flames at atmospheric pressure. They found that for a same molar fraction of H₂O or CO₂ in the mixture, the flame temperature is lower for CO₂ addition than for H₂O addition, consequently, they showed a slight underestimation of the flame speed of CH₄/O₂ diluted with CO₂ and H₂O, and this difference reduces for higher dilution rates.

Dally *et al.* [18] shown experimentally and numerically that CO₂ addition in a non-premixed methane/air flame reduced flame temperature by decreasing reactant concentration inside the reaction zone.

The influence on soot formation of CO₂ addition in diffusion flames was studied experimentally [19-21] and numerically [22, 23]. It was found that the soot volume fraction were abruptly decreases by CO₂ addition to the co-flow air. This suppression of soot formation resulted from the short residence time in the inception region.

It was shown that CO₂ dilution thermally and chemically limits the formation of soot precursors due to the decrease of H radicals consumed in the reaction $\text{CO}_2 + \text{H} \rightarrow \text{CO} + \text{OH}$.

Guo *et al.* [24] investigated experimentally the effects of different additives to air on the lift-off of a laminar CH₄/air diffusion flame. Results show that the addition of CO₂ causes flame lift-off due to the dilution, thermal and chemical effects, with the dilution effect being the most significant one, followed by the thermal effect. All these effects tend to reduce combustion intensity and cause flame to be lifted. The radiation and transport property effects are negligible.

Min *et al.* [25] focused on the effect of CO₂ addition to the air on the transition from an attached flame to a lifted flame issued from a coaxial non-premixed methane-air jet. They found experimentally that the CO₂ is the best destabilize among the diluents, because all the three effects (dilution > thermal > chemistry) contribute to induce loss of flame stability (CO₂ has the strongest ability to break flame stability, followed by N₂, then by Ar).

From the presented brief analysis it is clear that, despite numerous investigations of the effects of various additives on the stability of hydrogen jet flames, further investigations are required. Therefore, the present study aims to determine experimentally fundamental combustion characteristics of a turbulent jet diffusion NG-H₂ flames diluted with CO₂.

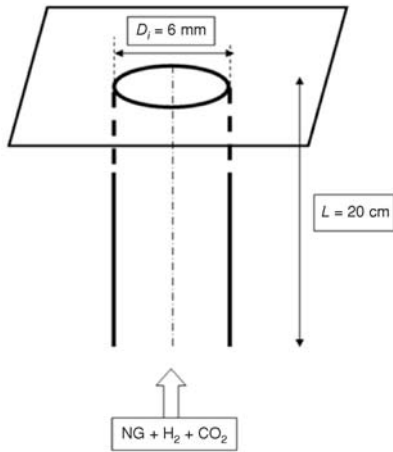


Figure 1. Sketch of the burner

Experimental set-up and measurement techniques

Experimental set-up

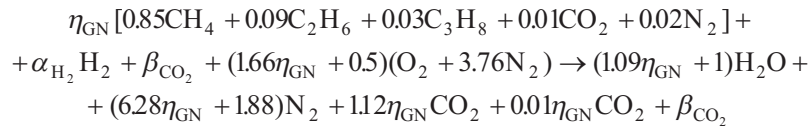
Figure 1 presents a sketch of the experimental apparatus used in the study. The experimental device consists of a 15 kW power burner powered by natural gas added to hydrogen and CO₂. A cylindrical burner, of internal diameter 6 mm, brings the mixture CH₄/H₂/CO₂ into the ambient air.

The natural gas used has a mass density of 0.83 kg/m³ and a volume composition of 85% CH₄, 9% C₂H₆, 3% C₃H₈, 2% N₂, 1% CO₂, and traces of higher hydrocarbon species.

The following table presents the characteristics of gas used.

	Natural gas	Hydrogen
Inferior calorific value PCI [kJkg ⁻¹]	45·10 ³	119·10 ³
Flammability limits in air [vol.%]	5-15	4-74.15
Laminar flame speed [cms ⁻¹]	43.4	170

The chemical equation for combustion of hythane diluted with CO₂ in air is:



with

80% < η_{GN} < 100%: percentage of natural gas in the fuel (hythane),

0 < α_{H_2} < 20%: percentage of hydrogen in the fuel (hythane), and

0 < β_{CO_2} < 50%: percentage of CO₂ in the mixture (GN + H₂ + CO₂).

The regulation of natural gas flow rate is done by a mass flow regulator and the carbon dioxide and hydrogen flow rates are regulated by sonic nozzles and pressure regulator.

The study has been carried out according to the percentage of hydrogen (% H₂ between 0 and 20%) in the fuel mixture (NG + H₂) in order to conserve the flame power of 15 kW. The fuel volumetric flow rate is then $Q_{\text{fuel}} = Q_{\text{vGN}} + Q_{\text{vH}_2}$. Then CO₂ is introduced in the fuel mixture to study the effect of CO₂ dilution. The percentage of CO₂ (% CO₂) in the mixture (NG + H₂ + CO₂) varies from 0% (no dilution) to 50%. The CO₂ flow rate is Q_{vCO_2} and the total flow rate is $Q_{\text{tot}} = Q_{\text{vfuel}} + Q_{\text{vCO}_2}$.

Table I summarizes the different parameters of the experiment according to the percentage of H₂ in the fuel mixture and the percentage of CO₂ in the total jet, such as the volumetric flow rates of the natural gas, hydrogen and CO₂, the exit jet bulk velocity and the jet Reynolds number.

Study of OH radical emission

The structure of the lift-off height is studied by collection of OH* radical emission on main band (0-0) to at 306.4 nm. The wavelength range of OH* emission is located between 280

Table 1. Gas flow rates, exit velocities of the jet (natural gas, H₂, and CO₂), jet Reynolds number and Schmidt number according to the percentage of H₂ in the fuel mixture and the percentage of CO₂ in the total jet

Jet flame mixture												
Fuel mixture												
%NG [fuel]	%H ₂ [fuel]	Q _{vNG} [lmin ⁻¹]	Q _{vH₂} [lmin ⁻¹]	Q _{vfuel} [lmin ⁻¹]	%CO ₂	Q _{vCO₂}	Q _{tot} [lmin ⁻¹]	Exit velocity U ₀ [ms ⁻¹]	Jet Reynolds number	Jet Schmidt number	%H ₂ [total mixture]	%NG [total mixture]
100	0	24.09	0	24.09	0	0	24	14.2	6312	0.8	0	100
					10	2.6	26.7	15.7	9005	0.86	0	90
					20	6	30.1	17.7	10741	0.91	0	80
					30	10.3	34.4	20.3	12968	1.04	0	70
					40	16	40.1	23.6	15937	1.11	0	60
					50	24	48.2	28.4	20095	1.25	0	50
95	5	23.73	1.25	24.98	0	0	24.9	14.7	7518	0.75	5	95
					10	2.7	27.7	16.3	8959	0.78	4.5	85.5
					20	6.2	31.2	18.4	10769	0.83	4	76
					30	10.7	35.7	21	13118	0.9	3.5	66.5
					40	16.6	41.6	24.5	16298	1	3	57
					50	24.9	49.9	29.4	20848	1.13	2.5	47.5
90	10	23.34	2.59	25.94	0	0	25.9	15.3	7422	0.71	10	90
					10	2.8	28.8	16.9	8914	0.73	9	81
					20	6.4	32.4	19.1	10804	0.77	8	72
					30	11.1	37	21.8	13289	0.83	7	63
					40	17.3	43.2	25.4	16712	0.92	6	54
					50	25.9	51.8	30.5	21733	0.95	5	45
85	15	22.92	4.046	26.97	0	0	26.9	15.9	7322	0.67	15	85
					10	2.9	29.9	17.6	8868	0.69	13.5	76.5
					20	6.7	33.7	19.8	10847	0.73	12	68
					30	11.5	38.5	22.7	13486	0.79	10.5	59.5
					40	17.9	44.9	26.5	17192	0.84	9	51
					50	26.9	53.9	31.8	22784	0.9	0.075	42.5
80	20	22.47	5.61	28.09	0	0	28.1	16.5	7205	0.65	0.2	80
					10	3.1	31.2	18.4	8822	0.68	0.18	72
					20	7	35.1	20.7	10899	0.7	0.16	64
					30	12	40.1	23.6	13713	0.76	0.14	56
					40	18.7	46.8	27.6	17753	0.8	0.12	48
					50	28.1	56.1	33.1	24052	0.86	0.1	40

and 310 nm, which implies the filtration of incident radiation of flame to collect the radical emission. The results coming from this technique enable to visualize the lift-off height as well as the flame front fluctuations and the flame length.

In order to visualize the OH* radical, the recording set-up is made up of an intensified CCD camera (512×512 pixel², 14 bit dynamic range, FlameStar Lavision), an objective (UV-Nikkor, Nikon) with a focal of 45 mm, and a glass filter (UG11, Melles Griot) with 3 mm thickness to filter out the flame radiation (transmittance coefficient greater than 0.1 between 275 and 375 nm). To obtain a reasonable and workable signal, it is possible to control the camera gain, fixed to 250, as well as the duration of the intensification door, varying according to the configuration from 100 to 250 μ s. Indeed the OH* production at the bottom of flame deeply depends on the configuration and involves to modify these parameters according to the configuration. From the recording of 500 instantaneous images, a statistical processing enables to obtain the mean position of the lift-off height, allowing the characterization of the stability criterion.

To determine the lift-off position, it is necessary to binarize the images and then to detect the contour.

The binarization threshold permits to eliminate the picture noise but requires precautions to not affect the image signal. Then the lift-off position is defined as the nearest point from the burner where the OH* signal appears. The mean image is calculated in each point from 500 instantaneous images. The averaged binary image can also be obtained. Figure 2 illustrates an example of image statistical processing of the OH* emission in the lift-off zone.

Results and discussion

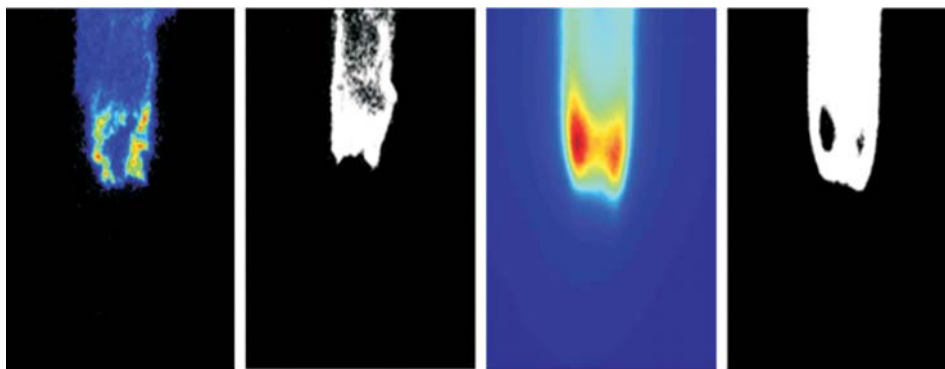


Figure 2. Statistical processing of the OH* emission in the lift-off zone; (a) instantaneous image; (b) instantaneous binary image, (c) average image of 500 instantaneous images, and (d) binary image of the average (for color image see journal web-site)

Flame lift-off position and flame fluctuations

Study of the flame lift-off position

The study of the flame lift-off position is carried out by collecting the OH* radical emission. Figure 3 shows an example of the average image from 500 instantaneous images. From the average OH* field, the flame lift-off position is studied according to hydrogen volume fraction and the dilution of CO₂.

Figure 4 illustrates the variation of the lift-off height of the flame with the % CO₂ added at various % H₂ content in the hythane. From this figure it can be seen that the lift-off height increases with the addition of CO₂. The lift-off height passes from 0 mm to 90 mm when 20% of CO₂ is added in stoichiometric condition and is hooked to the burner when adding the hydrogen.

This is due to the fact that the addition of CO₂ causes the flame lift-off through the following three paths: dilution effect because of reduction in the concentration of the reactive species, the drop in turn causes a reduction in the reaction rate, while decreasing the the flame velocity which weakens the flame stability; thermal effect because of the change in flame temperature, CO₂ addition implies a drop of local flame temperature; and the direct chemical effect because of the participation of the additive in chemical reactions related to soot formation and oxidation. Reaction CO₂ + CH → HCO + CO was found to be responsible for the chemical effects of carbon dioxide addition, Liu *et al.* [22] investigated numerically the chemical effects of carbon dioxide addition on both the fuel side and the oxidizer side of a diffusion flame. They performed that reactions CO₂ + H → CO + OH and CO₂ + CH → HCO + CO were found to be responsible for the chemical effects of CO₂ addition. The chemical effects of CO₂ addition on the fuel side are small but become significant when introduced on the oxidizer side.

Guo and Smallwood [23] paid particular attention to soot formation in an ethylene/air flame. For this purpose, some simulations were carried out with CO₂, whereas other calculations were made with a chemically inert species (FCO₂), with the same physical properties as CO₂. Comparison between results obtained with CO₂ and FCO₂ indicated the following reactions: CO₂ + H → CO + OH in the air stream, and CO₂ + CH → HCO + CO in the fuel stream were enhanced by adding CO₂. The dilution effect is most significant, followed by the thermal effect, relatively; the chemical effects of CO₂ addition on the fuel side are small.

As soon as one H₂ is added, the flame hangs on the burner whatever the dilution of CO₂, in fact the addition of hydrogen promotes mixing and allows the attachment to burner rim due to its high molecular diffusivity and its high reactivity. The high molecular diffusivity of hydrogen increases the mixing between fuels and air. Thanks to the hydrogen addition, the species react more quickly in the mixture. Moreover it can be noted that with 20% H₂, the configurations are stable even when the jets are diluted with CO₂ concentration of 30% while for 0% H₂ flame is blown over 20% CO₂. This means that the limits of flame blowing are pushed with the addition of H₂. Indeed the addition of hydrogen increases the burning rate and the difference in density in the jet and allows the attachment of the rim of the burner flame.

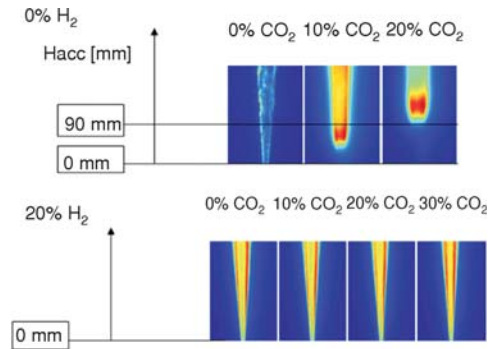


Figure 3. Images of OH chemiluminescence on the base of the flame (for color image see journal web-site)

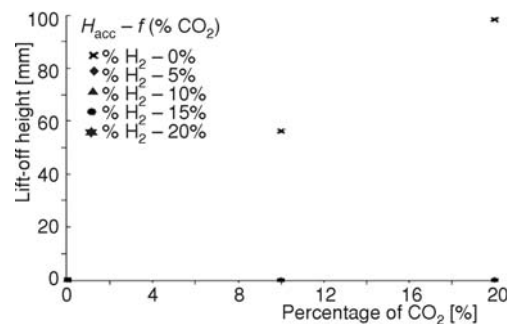


Figure 4. Flame lift-off height against CO₂ concentrations at various percentages of H₂

Fluctuations in the bottom region of the flame

The knowledge of the fluctuations of the lift-off heights permits to understand the stability phenomenon of flame lift-off position due to the wide flammability limits of hydrogen.

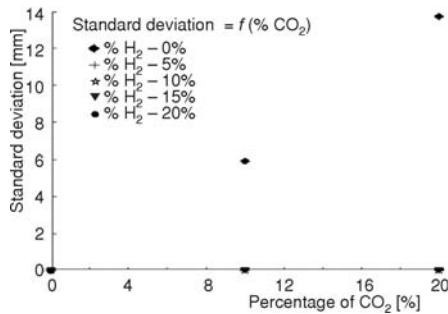


Figure 5. RMS of the flame lift-off height according to hydrogen volume fraction and CO₂ dilution

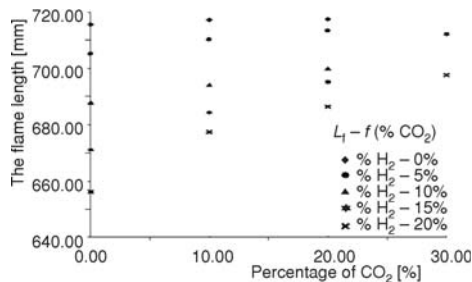


Figure 6. The height of the flame diluted with CO₂ at various % H₂

combustion rate and thereby reduces the overall flame length. In addition, the hydrogen low molecular weight and rapid radical molecular diffusion causes an increase in the flame width, which is conversely proportional to the flame length. For example in the configuration when 0% of CO₂ is added to (NG/H₂), the normalized visible flame length (L_f/D_i) passes from 123 to 113 when 20% of H₂ is added to NG/H₂).

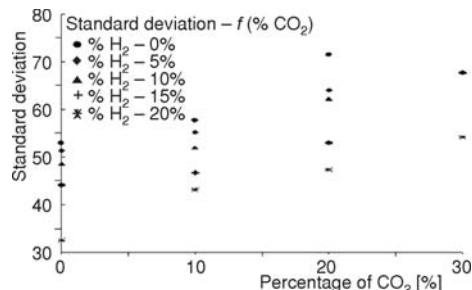


Figure 7. RMS of the flame height according to hydrogen volume fraction and CO₂ dilution

From the RMS of the lift-off heights according to the hydrogen volume fraction and the CO₂ dilution (fig. 5), it is possible to deduce the evolution of the combustion stability in the base of flame. Figure 5 shows that fluctuations of the lift-off heights decrease with the growth of hydrogen fraction. The addition of hydrogen within natural gas permits to ameliorate the flame stability due to the extension of the flammability limit and the high reactivity of hydrogen. Figure 5 reveals that the diluted combustion with CO₂ increases the fluctuation of the lift-off heights. For example, the RMS passes from 0 mm to 14 mm when 20% of CO₂ is added at stoichiometric conditions.

Study of the flame length

The flame length, L_f , defined as the height of the conical reaction zone was measured by collecting the OH* radical emission. Figures 6 and 7 illustrate, respectively, the variation of the flame length and his standard deviation with the % CO₂ (vol.) added. From fig. 6, it can be seen that the flame length decreases as the hydrogen content increases in the NG-H₂ hybrid fuel. This can be attributed to the increase in the radical pool that accompanies the H₂ addition, such H and OH radicals, which consequently enhances the combustion rate and thereby reduces the overall flame length. In addition, the hydrogen low molecular weight and rapid radical molecular diffusion causes an increase in the flame width, which is conversely proportional to the flame length. For example in the configuration when 0% of CO₂ is added to (NG/H₂), the normalized visible flame length (L_f/D_i) passes from 123 to 113 when 20% of H₂ is added to NG/H₂).

This result depicts a good agreement with the available data in the existing literature [11] for NG-H₂ hybrid fuel. They confirmed a reduction in the NG-H₂ turbulent flame length with hydrogen addition, the normalized visible flame length (L_f/D_i) decreases as hydrogen content increases in the mixture; it passes from 230 to 200 when 20% of H₂ is added to NG/H₂.

Also, it can be noticed from fig. 6 that the flame length increases with the mole fraction of CO₂ added to NG-H₂ hybrid fuel for that same % H₂ content in fuel. For % H₂ content in NG-H₂ hy-

brid fuel up to 10% CO₂ additions could produce lifted flame if the CO₂ concentration is high. At higher CO₂ concentration, flame would remain attached until blow-out. This is mainly due to the fact that the addition of CO₂ causes the elongation of the flame through thermal effect because of the decrease in temperature which leads a reduction in the reaction rate and thus decreases the turbulent flame speed which increases the length of the flame, and dilution effect by reducing the reactivity of the mixture and thus the turbulent flame speed as well the elongation of the flame.

From fig. 7, it can be seen that fluctuations of the flame length decrease with the growth of hydrogen fraction. The addition of hydrogen favors the fluctuation reduction. Also, it can be noticed from fig. 7 that the fluctuations of the flame length increases with the mole fraction of CO₂ added to NG-H₂ hybrid fuel for that same % H₂ content in fuel. For example, in the configuration where 5% of hydrogen is added to the natural gas, the RMS passes from 51 mm for 0% of CO₂, to 64 mm when the percentage of CO₂ is 20%.

Conclusions

This study focused on the combustion of hythane diluted with CO₂, the jet being composed of a mixture in variable proportions of natural gas, hydrogen, and CO₂. This experiment was performed on a cylindrical burner generating a 15 kW turbulent flame in ambient air.

This experimental study consisted mainly of studying the flame lift-off position and the structure of the flame according to the addition of hydrogen in the hybrid fuel and especially the effect of dilution with CO₂. By direct OH* imaging, the overall behavior of the flame has been investigated.

- The technique of chemiluminescence of OH* centered on the initial zone of the flame was used to assess the evolution of attachment heights based on the percentage of hydrogen and CO₂. The study showed the improvement of flame stability with the addition of hydrogen in natural gas. Indeed, experimental measurements have shown that the addition of hydrogen increases the fuel jet velocity as well as the molecular diffusivity, promotes mixing of reactants and ameliorates the flame stability.
- With the OH* chemiluminescence, the study of fluctuations in the bottom region of the flame shows an increase of the lift-off height stability according to the two parameters. The reasons are the same as the previous.
- The second part also by collection of the emission of the OH* radical centered on the final zone of the flame was used to assess changes in the average length of the flame based on the percentage of hydrogen and CO₂ dilution. The study showed that dilution tends to grow the length of the flame and that the addition of hydrogen increases the reactivity of the mixture and therefore the turbulent flame speed which decreases the length of the flame.
- With the chemiluminescence, the study of fluctuations of the base of the flame shows an increase of the fluctuations of the length of the flame according to the two parameters. The reasons are the same as the previous.

Acknowledgment

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