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Effect of hydrogen addition on the laminar flame speed of biogas. Experimental and numerical study

Efecto de la adición de hidrógeno en la velocidad de llama laminar del biogás. Estudio numérico y experimental.

M. J. Acero-Caballero ; L. E. Pacheco-Sandoval ; C.A. Díaz-González.

Abstract—In this study, the laminar flame speed of biogas/H₂ fuel mixtures is measured from pure biogas to biogas with an addition of 15% H₂ in volume by using the method of the inner cone of combustion zone. The laminar flame speed is measured by mean of the geometric parameters of combustion cone capturing images of the flames. The aeration factors covers from stoichiometric conditions to lean conditions. Experimental results have been compared with numerical results obtained with CHEMKIN software, using GRIMECH kinetical mechanism, with good agreement between them and with previous works, showing the reliability of the method used. The trends on laminar flame speed experimental results obtained are explained by mean of numerical results on temperature profile and heat release rate. Hydrogen addition in biogas flames show an important effect, especially in zones near stoichiometric flames, decreasing its effects in lean mixtures combustion.

Index Terms— Biogas-hydrogen; laminar flame speed; experimental and numerical study.

Resumen — En este trabajo, se ha medido la velocidad de llama laminar de las mezclas de combustible biogás/H2, desde biogás puro hasta mezclas de este con una adición de 15% de H2 en volumen, utilizando el método del cono interno de la zona de combustión o método bunsen. La velocidad de la llama laminar se determina por medio de los parámetros geométricos del cono de combustión capturando imágenes de las llamas. Los factores de aireación abarcan desde condiciones estequiométricas hasta condiciones de mezcla pobre. Los resultados experimentales se han comparado con los resultados numéricos obtenidos con el software CHEMKIN, utilizando el mecanismo cinético de GRIMECH, con una buena concordancia entre ellos y con trabajos previos, mostrando la confiabilidad del método utilizado. Las tendencias en los resultados experimentales de la velocidad de la llama laminar se explican por medio de resultados numéricos sobre el perfil de temperatura y la tasa de liberación de calor. La adición de hidrógeno en las llamas de biogás muestra un efecto

importante, especialmente en zonas cercanas a las llamas estequiométricas, disminuyendo sus efectos en la combustión de mezclas pobres.

Palabras claves—Biogás-Hidrógeno; velocidad de llama laminar; estudio numérico y experimental.

I. INTRODUCTION

HE necessary use of combustion technologies to reduce the dependence on fossil fuels and additionally, the emission of greenhouse gases to meet the goals of climate change mitigation [1], [2], have generated a continuous growth in the study of the possible applications of alternative fuels such as biogas generated by anaerobic decomposition of organic substances, synthesis gas produced in the gasification of coal or biomass, liquid biofuels as fuel alcohol obtained different crops such as corn or sugar cane and biodiesel obtained from vegetable oils.

Biogas generated by the anaerobic decomposition of organic substances is a great source of energy, especially in regions or countries where economic growth causes an increasing in the generation of municipal solid waste or organic waste product of industrial or agricultural activities where its decomposition could generate this alternative fuel [3]-[6]. This biogas composed mainly of methane (CH4), carbon dioxide (CO2) and other small amounts of oxygen, nitrogen, volatile compounds and even hydrogen sulphide, is generally cleaned of some of these minor compounds [7] to reduce their harmful effect on combustion systems.

The clean biogas (CH4 and CO2) is a fuel whose combustion properties such as calorific value, flame temperature and deflagration velocity are poor compared to conventional gaseous fuels such as natural gas or LPG [8]-[11], producing that the use of the biogas in combustion systems requires modifications in the design of the burners that avoid the

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problems of flame instability associated to the lower flame speed and the change in the limits of flammability [12]-[14]. One of the ways to improve these combustion properties in to convert biogas to biomethane using different treatments that reduce the CO2 content and its effects [15], [16] as well as other minor compounds such as H2S [7], [17], [18]. However, this treatment process is expensive in terms of equipment and absorbing substances such as amines. The improvement of the combustion properties of the biogas can be achieved by mixing it with another combustible gas whose combustion properties (mainly the flame speed) is greater, so that the resulting mixture has combustion properties that can be compared with those of the gas Natural or LPG, reducing the problems of biogas interchangeability with these fuels [8], [19]. Hydrogen is the combustible gas that has the greatest value of flame speed at standard conditions, in addition to a higher calorific value, a higher flammability range and a higher flame temperature, and therefore, its addition to the biogas will result in an improvement of the combustion properties of this last one. Different studies have concluded that mixing the biogas with other fuels such as H2, LPG or natural gas increases the flame speed of the resulting mixture and therefore the range of flame stability [20]-[24]. Hydrogen has been selected as the biogas additive to improve its combustion properties since it is a renewable fuel, with a zero greenhouse potential and which could additionally be obtained from fossil fuels through reforming processes [25] or even from the same biogas [26]. By means of the realization of experimental measurements compared with computational calculations it is tried to determine the effect of the addition of different proportions of H2 to a biogas simulated in the flame speed. The results are intended to guide the H2 produced from reforming other fossil fuels as an alternative to improve the combustion properties of biogas in atmospheric premix burners.

I. METHODOLOGY

Four experimental assemblies were performed and contrasted with simulations of reactions in the CHEMKIN Software. The mixtures with percentages used in the experimental setup were: a) Pure biogas, b) Biogas with addition of 5% Hydrogen, and c) Biogas with addition of Hydrogen of 15% by volume. The combustion reactions of these same blends have been simulated using the CHEMKIN software mentioned.

A. Simulation of the reactions of the defined mixtures

Simulations of the combustion reactions of the studied mixtures have been performed for different aeration factors, starting with a stoichiometric mixture and increasing these aeration factors as shown in Table I.

Mixture	Composition	Aeration factor n				
Pure biogas	$0.6\text{CH}_4 + 0.4\text{CO}_2$	1	1.1	1.2	1.3	1.4
Biogas + 5% H ₂	$0.57CH_4 + 0.38CO_2 \\ + 0.05H_2$	1	1.1	1.2	1.3	1.4
Biogas + 15% H ₂	$0.51CH_4 + 0.34CO_2 \\ + 0.15H_2$	1	1.1	1.2	1.3	1.4

Table I. Composition of biogas and biogas/H2 mixtures simulated

The aeration factor n for combustion is introduced into the stoichiometric equation of each reaction as given below:

$$A CH_4 + B CO_2 + D H_2 + n(a_t) (O_2 + 3.76 N_2) \rightarrow X CO_2 + Y H_2O + ZN_2 + WO_2$$
(1)

Where n is the aeration factor and a_t is the theoretical air coefficient of the combustion reaction determined by the elementary reaction balances.

The CHEMKIN tool has been used to determine the laminar flame speed of the specified combustible gas mixtures. The chemical kinetics and transport equations for gas phase are solved following the free-propagation flame model to obtain the chemical species concentration profiles, the heat release rate and the temperature profile developed for adiabatic flame starting from the compositions Molars of each of the selected mixtures described above and with initial values of the pressure at which the flame is developed and an approximate temperature profile. For these simulations, the chemical kinetics mechanisms Grimech 2.11 and Grimech 3.0 were used, mechanisms that have been validated for the simulation of biogas flames [27]-[29].

B. Experimental setup

The experimental determination of the deflagration rate of the mixtures studied was performed using the method of the inner cone of the flame, using a premixed atmospheric burner designed for burning natural gas with a nominal power of 1 kW. As it is an induced air burner, it does not require a ventilation system for the supply of combustion air. The gas flow is regulated through a pressure regulating valve and measured with Elster-Handel wet seal meter [0.002 - 0.2 m³/h, accuracy +0.01 dm³]. The pressures and temperatures were measured with manometers and column thermometers. The experimental setup is shown in Fig. 1 and the geometry and dimensions of the burner are shown in Fig. 2 and Table II respectively.

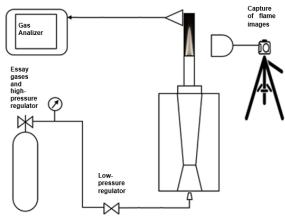


Fig. 1. Experimental setup diagram.

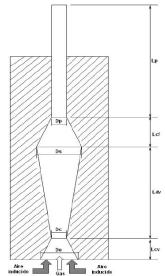


Fig. 2. Geometry of the premix burner

Dimension	Value [mm]	
Venturi inlet diameter – De	33	
Venturi throat diameter – Dc	16	
Venturi oultet diameter – Ds	34	
Port diameter – Dp	17	
Length of the converging zone – Lcv	38	
Length of the diverging zone – Ldv	132	
Length of the final convergent zone – Lcf	44	
Port length – Lp	15	

Table II. Dimensions of the premix burner

For the combustion analysis, the GAS analyzer MADUR-Photon, owned by the GIRES Research group of the UNAB was used. The CO_2 and O_2 concentration was measured to determine the aeration factor in each experimental test using the following equation:

$$n = \frac{\binom{A/C}_{used}}{\binom{A/C}_{theo}}.$$
(2)

Where:

n Is the aeration factor used in the combustion.

 $(A/C)_{used}$ Is the air-fuel ratio used in the combustion.

 $\binom{A}{C}_{theo}$ Is the theorical air-fuel ratio for each fuel mixture studied.

The aeration factors obtained in the experimental tests are shown in Table 3.

Fuel mixtures	Aeration factors experimentally obtained n						
Pure biogas	0.992	1.09	1.21	1.28	1.374		
Biogas + 5% H ₂	1.01	1.10	1.22	1.31	1.40		
Biogas + 15% H ₂	-	1.11	1.23	1.33	1.42		

Table 3. Aeration factors experimentally obtained for fuel mixtures

For pure biogas and its mixture with 5% hydrogen, aeration factors were obtained from the stoichiometric mixture but for

the biogas mixture with 15% hydrogen, a stable flame could not be obtained for an aeration factor near the stoichiometric mixture. In general, poor flames were obtained because the design of the burner used is for this type of flames. For each aeration factor obtained, image captures of the stabilized flame were taken. A professional Canon EOS t3i camera with 18-55 mm lens has been used to obtain the desired quality in the flame image. This image has been observed in treatment imaging software to determine the geometric parameters of the flame cone as the internal angle and height of the flame. Fig. 3 shows the geometric parameters determined for each image taken to determine the laminar deflagration rate of the combustible mixtures.

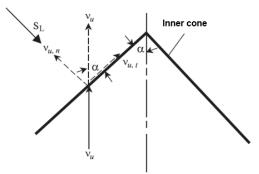


Fig. 3. Geometrical parameters of the premix flame

Finally, the flame speed has been determined by mean of following equation:

$$S_L = V_u \cos \alpha \dots (3)$$

Where:

 S_L It is the laminar flame speed of the fuel mixture.

 V_u It is the average exit velocity of the air-fuel mixture at the burner port.

 α Is the inner angle of the combustion cone.

II. RESULTS AND DISCUSSION

Now, the experimental results and the results of the simulations obtained in the estimation of the laminar deflagration velocity of the studied mixtures are presented and discussed.

A. Experimental laminar flame speed.

According to the experimental methodology presented, the images of the flame for the different combustible mixtures for each established aeration factor have been captured. The flame images of pure biogas, biogas with 5% addition of hydrogen and biogas with 15% hydrogen addition blends are shown in Fig. 4-6.

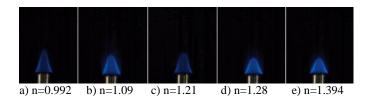


Fig. 3. Flame for pure Biogas with different aeration factors

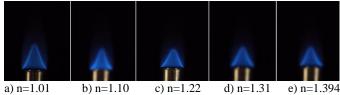


Fig. 4. Flame for pure Biogas + 5% H_2 with different aeration factors

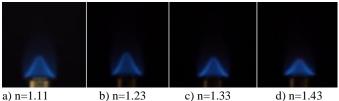


Fig. 5. Flame for pure Biogas + 15% H₂ with different aeration factors

In the captured images, there is a slight decrease in the height of the combustion cone delimiting the reaction zone as the flame aeration increases. It is observed that biogas flames with addition of hydrogen, present a geometry with the typical triangular profile of flames of fuels containing hydrogen [30]. The results of the laminar flame speed experimentally obtained are shown in Fig. 7. It is observed how the addition of hydrogen to the biogas produces an increase in its laminar deflagration rate. This increase is more notable, according to the trend observed, in aeration factors close to the stoichiometric flame. In poor flames obtained in the experiment, the effect of hydrogen is lower.

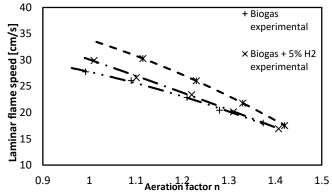


Fig. 6. Experimental laminar flame speed at different biogas/ H_2 mixtures varying the aeration factor.

i. Comparison with computational models

Experimental results have been compared with computational results obtained with CHEMKIN, as was mentioned in the methodology. Comparison between experimental values of pure biogas laminar flame speed with values obtained

numerically for different aeration factors is shown in Fig. 8. In Figs. 9 and 10 are shown the results of comparison between experimental and numerical values obtained for mixtures of biogas with 5% and 15% hydrogen addition respectively.

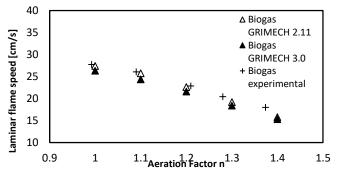


Fig. 7. Laminar flame speed of experimental and simulated pure Biogas with variation of aeration factor.

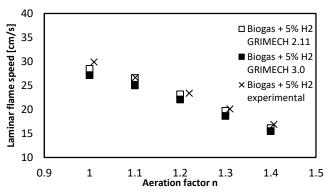


Fig. 9. Laminar flame speed of experimental and simulated of biogas/H2 mixture with 5% H₂ with variation of aeration factor.

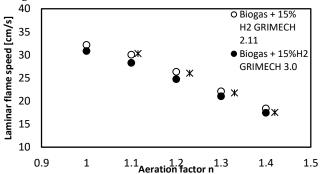


Fig. 10. Laminar flame speed of experimental and simulated of biogas/H₂ mixture with 15% H₂ with variation of aeration factor.

Values experimentally obtained shown a correspondence with numerical values and equally with reported values for biogas flames [15], [19], [21], [22]. It is also observed a slight difference between the values obtained for the kinetic mechanisms used, this may be due to the difference between the number of reactions included in each mechanism (277 in GRIMECH 2.11 and 325 in GRIMECH 3.0) and the number of species included in these mechanisms (49 in GRIMECH 2.11 and 53 in GRIMECH 3.0). The good agreement between the experimental results and the numerical results shows that the error in the laminar flame speed measurements (associated with

image analysis and gas flow measurement) is acceptable and that this method is therefore appropriate for this purpose.

In stoichiometric flames the maximum value of the laminar flame speed for the biogas/ H_2 mixtures is reached, with values ranging from 27.7 m/s for pure biogas and 29.9 m/s for biogas with 5% H_2 . For biogas mixture with 15% H_2 , it was not possible to obtain the deflagration rate in stoichiometric flame due to flame instability. For aeration factors of n=1.1, the values obtained were 26.07 m/s, 26.65 m/s and 30.28 m/s for pure biogas, a mixture with 5% H_2 and a mixture with 15% H_2 respectively , where the effect of hydrogen in the increase of the biogas flame speed is appreciated.

Observing the flame structure obtained in the different simulations, we can relate the observed increase in the rate of deflagration with the effect of hydrogen on the kinetics of the reaction. Fig.s 11-13 show the temperature profiles for stoichiometric flames (n = 1) and poor flames (n = 1.2 and n = 1.4).

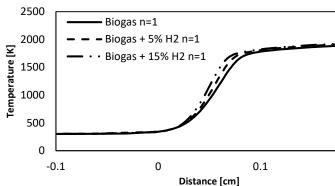


Fig. 11. Temperature profile of the reaction zone for biogas/ H_2 mixtures with n = 1.

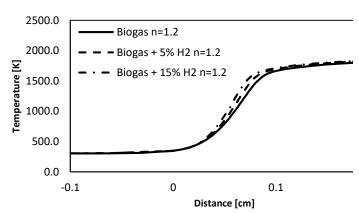


Fig. 12. Temperature profile of the reaction zone for biogas/ H_2 mixtures with n = 1.2

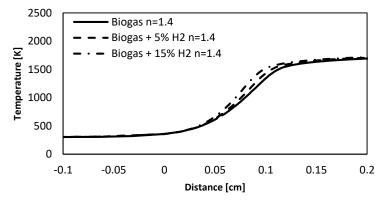


Fig. 13. Temperature profile of the reaction zone for biogas/ H_2 mixtures with n=1.4

As can be seen, the addition of hydrogen to the biogas generates an acceleration of the combustion reaction. This being reflected in higher temperature gradients in the reaction zone and resulting in higher rates of heat release, which ultimately cause less thickness of the reaction zone, which is indicative of the increase in the rate of deflagration [31], [32]. In Figs. 14-16, it can be observed the effect of hydrogen addition in heat release rate of combustion reactions.

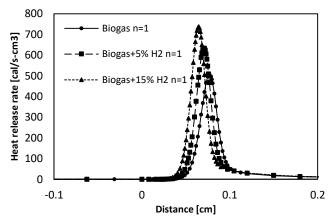


Fig. 14. Heat release rate for biogas/ H_2 mixtures with n=1

For stoichiometric mixtures, pure biogas presents a value of this maximum heat release rate of 508 cal/s-cm³. Addition of 5% of hydrogen causes a maximum rate of heat release of 634 cal/s-cm³ and an addition of 15% of hydrogen generates a maximum rate of heat release of 738 cal/s-cm³. This increase in this parameter is reflected in the flame speed.

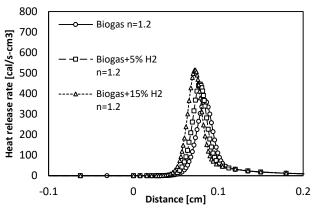


Fig. 15. Heat release rate for biogas/ H_2 mixtures with n = 1.2

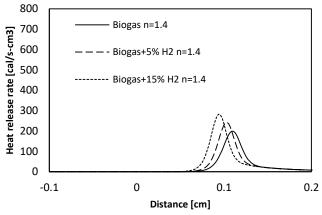


Fig. 16. Heat release rate for biogas/ H_2 mixtures with n = 1.4

For the biogas/H₂ mixtures with aeration factor n = 1.2, it can be seen that the maximum value of the heat release rate for pure biogas is 366 cal/s-cm³, increasing to 446 cal/s-cm³ for mixing with 5% H₂ and 516 cal/s-cm³ for mixing with 15% H₂. In this case, the increase in the rate of heat release presents a less noticeable increase than in the stoichiometric flame, as it is also observed in the rate of deflagration. Finally, for lean flames (n = 1.4), the values of the heat release rate have values from 199 cal/s-cm³ for pure biogas and 243 cal/s-cm³ and 283 cal/s-cm³ for Mixtures with 5% H₂ and 15% H₂ respectively. As noted, the increase in the rate of heat release in these lean mixtures is less noticeable in absolute terms but similar in percentages (45% for stoichiometric mixture, 40% and 42% for poor mixtures).

III. CONCLUSIONS

In this work, the velocity of laminar flame speed of biogas and biogas/ H_2 mixtures have been studied under normal conditions. The experimental determination was based on the method of the height of the internal cone of the flame by the determination of geometric parameters of the flame with its image. The experimental results have been compared with numerical simulations with good correspondence between them and with previous works. The influence of the addition of hydrogen and the aeration rate on the laminar deflagration velocity of the mixtures have been studied.

The main results allow us to conclude that the laminar flame speed of the biogas under normal conditions and aeration conditions of n = 1.1 reaches a value of 26.07 m/s. The addition of hydrogen allows increasing this velocity to values of 30.28 m/s with a 15% addition of H_2 , implying an increase of up to 16% of this parameter. However, under lean conditions, the increase in laminar flame speed between pure biogas and the addition of 15% H_2 only reaches approximately 5% according to results presented. The increase in the laminar flame speed can be divided into two zones, one close to stoichiometric mixtures where a significant increase is observed and another one of lean mixtures where this increase is less noticeable.

The addition of H_2 to the biogas generates an increase in the kinetics of the reaction. This is reflected in the higher temperature gradients in the reaction zone of mixtures with higher H_2 content and also higher rates of release of heat in the reaction zone. This show a different behavior to the laminar flame speed (where there is a relative increase), since in mixtures close to the stoichiometric one has an increase in this rate of up to 45% according to the results obtained and of 40% and 42% in poor mixtures.

IV. ACKNOWLEDGMENTS

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