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Review

Laminar Burning Velocity of Biogas-Containing Mixtures. A Literature Review

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Abstract: Currently, the use of fossil fuels is very high and existing nature reserves are rapidly depleted. Therefore, researchers are turning their attention to find renewable fuels that have a low impact on the environment, to replace these fossil fuels. Biogas is a low-cost alternative, sustainable, renewable fuel existing worldwide. It can be produced by decomposition of vegetation or waste products of human and animal biological activity. This process is performed by microorganisms (such as methanogens and sulfate-reducing bacteria) by anaerobic digestion. Biogas can serve as a basis for heat and electricity production used for domestic heating and cooking. It can be also used to feed internal combustion engines, gas turbines, fuel cells, or cogeneration systems. In this paper, a comprehensive literature study regarding the laminar burning velocity of biogas-containing mixtures is presented. This study aims to characterize the use of biogas as IC (internal combustion) engine fuel, and to develop efficient safety recommendations and to predict and reduce the risk of fires and accidental explosions caused by biogas.

Keywords: biogas; oxygen addition; hydrogen addition; laminar burning velocity; experimental and computing methods

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1. Introduction

Energy based on fossil fuel consumption is the basis of industrial and economic development. The current fossil fuel utilisation is rapidly depleting the natural reserves. Therefore, researchers are currently turning their attention to find renewable fuels having a low impact on the environment to replace these depleting fossil fuels. The major challenge for scientists and engineers in addition to optimisation of engine combustion, improving fuel economy and lower pollutant emissions, is to find alternative-fuels able to maintain outstanding performance, durability, and reliability of engines at an affordable price. One of the alternative fuels tested and used successfully is biogas.

Biogas is a low-cost alternative, sustainable, renewable fuel existing worldwide produced by microorganisms, such as methanogens and sulfate-reducing bacteria, performing anaerobic respiration. Biogas can refer to gas produced naturally or industrially from decomposition of vegetation or waste products of human and animal biological activity. Because it is a natural by-product, biogas is one of the non-exhaustible supplies in the world. The environmental benefit by producing energy from biogas is deeply appreciated through the reduction of gas emissions that exhibit a greenhouse effect by preventing its release into the atmosphere. Even if the biogas has a low heating value (about 3000–6000 kcal/m^3) compared to natural gas (about 8000–9000 kcal/m^3) or liquefied petroleum gas (about 25,000–28,000 kcal/m^3), its total chemical energy is sufficient to serve as a basis for the production of the heat and electricity that can be further used for domestic heating and cooking, or to feed internal combustion engines, gas turbines, fuel cells, or cogeneration systems [1–5]. A schematic diagram for biogas production and its applications is given in Figure 1.

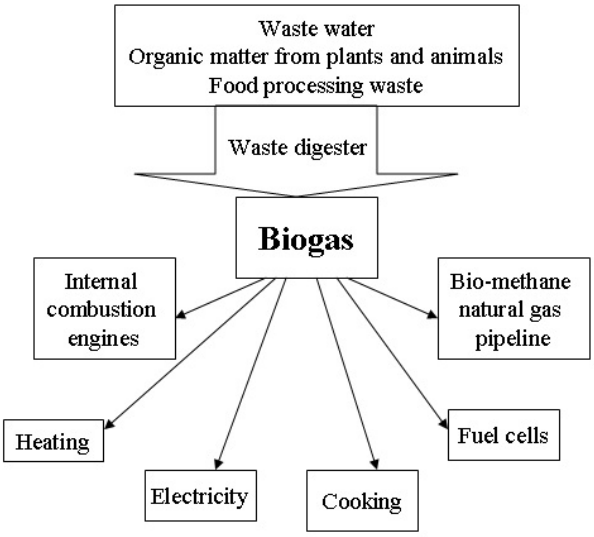


Figure 1. Schematic diagram for biogas production and applications.

The composition of biogas varies depending upon the substrate composition, as well as the conditions within the anaerobic reactor (temperature, pH, and substrate concentration) [6]. Biogas is a mixture mainly consisting of methane and carbon dioxide in different molar fractions, depending on the preparation method. The composition of biogas is approximately 70% CH_4–30% CO_2, with traces of H_2S, H_2, NH_3 and H_2O_{vap}. The most important component of biogas, from the calorific perspective, is methane, CH_4, which is a very flammable fuel. CO_2 and H_2O_{vap} have a diluting effect on the mixture composition and reduce the flammability range. Besides CO_2 and H_2O_{vap}, N_2 from the air contributes to diluting the mixture and therefore can reduce considerably the biogas explosivity. The presence of multiple diluting agents hinders the prediction of the whole flammability envelope since each inert gas has its own inerting power when mixed with methane [7].

Unfortunately, fires and accidental explosions involving biogas could occur [8–11]. Therefore, a strict determination of the explosion parameters of biogas-air mixtures has a great practical interest for mitigation of incidents associated with gaseous explosions and for evaluating safe conditions of running reactors or plants where flammable mixtures are formed.

Scientific contributions regarding biogas explosions are available in the literature [12–19]. Some of them report values of flammability parameters of synthetic biogas (flammability limits, limiting oxygen concentration, maximum explosion pressure and maximum rate of pressure rise) [14,15], while others report laminar burning velocities from measurements or kinetic modelling [17,20–24]. Recent studies regarding synthetic biogas-air mixtures [25,26], CH_4–air–inert mixtures [27–31], and CH_4–nitrous oxide–inert mixtures [32,33] were performed under various initial conditions to obtain the laminar burning velocities. The investigation on the effect of concentration of methane in biogas when it is used as a fuel for a spark ignition engine was reported by Porpatham et al. [34]. Other studies revealed the effect of additives (N_2, Ar, H_2O or CO_2) on CH_4/air, CH_4/CO_2/air and $CH_4/O_2/H_2$/inert flames [35–41]. The attention of other researchers has been directed towards the combustion characteristics of biogas under hydrogen-enriched conditions [24,42–49]. In some experiments, realised in a spark ignition engine, small H_2 amounts were added to biogas to enhance engine performance and reduce pollutant emissions [16,50–53]. The effects of oxygen-enriched air on the operation and performance of a diesel-biogas dual fuel engine were also presented by Cacua et al. [54]. Other studies on biogas combustion in the presence of oxygen-enriched air were presented by Li et al. [42]; Oh and Noh [55]; Cardona and Amell [56]; Navarro-Puyuelo et al. [57]; Striūgas et al. [58]; Wang et al. [59].

Biogas (alone or blended with other fuels) is used as fuel in internal combustion engines, which are the main power source for transport vehicles and commonly used for powering generators of electrical energy [4,5,60]. Therefore, it is necessary to study its combustion properties such as the explosion pressure, severity factor and laminar burning velocity. The last one is the most important parameter that describes the combustion process. Laminar burning velocity represents a unique characteristic of a gaseous mixture with fixed composition, initial temperature and pressure. This parameter adequately describes the essential characteristics of the fuel-oxidizer mixtures regarding thermal diffusivity, exothermicity, and reactivity. On a practical level, this parameter affects the fuel burning rate in internal combustion engines and the engine's performance and emissions. On a fundamental level, the laminar burning velocity is an important target for kinetic mechanism development and validation. Accurate determination of laminar burning velocity is extremely important for the development and validation of kinetic mechanisms for gasoline, diesel surrogate fuels and alternative fuels [60–62].

The state of the art, the challenges of combustion chemistry research, the flammability limits, the impact of carbon dioxide on the laminar burning velocity and stability of the flame for biogas-air mixtures were reviewed by Wang et al. [4] and Pizzuti et al. [60,62]. In the present paper, a comprehensive study of the literature regarding the laminar burning velocity of biogas-containing mixture is presented aiming to characterize the use of biogas as internal combustion engine fuel, of the necessity to develop new safety measures for handling, storage and use of these mixtures and to predict and reduce the risks associated with fires and explosions.

2. General Characteristics of Flames in Gaseous Mixtures. The Laminar Burning Velocity

A flame is the result of a self-sustaining chemical reaction usually made visible by the luminosity of the burning gases. Associated with a flame is the flame front, in which the unburnt gas is heated and converted into products. Whether the flame is stationary or moving in space, the movement of the flame front, which is of finite thickness, is taken as an indication of the progress of the flame [63]. The flames can be laminar or turbulent. The laminar flames have been studied extensively in laboratories as they can provide the most detailed information about combustion chemistry and various physical effects that act upon the flame [64].

The laminar burning velocity is an important intrinsic property of a combustible mixture. It is defined as the speed at which an adiabatic, unstretched, premixed planar flame propagates relative to the unburned mixture [65]. The laminar burning velocity is a physico-chemical property of a premixed fuel-oxidizer mixture, resulting from the combined influence of the mixture diffusivity, exothermicity and reactivity [66,67]. It affects or even determines the burning rate of fuel-air mixtures in practical combustion systems. The laminar burning velocity provides a measure of overall reactivity of fuel-air mixtures and helps in determining the rates of heat release and in testing and validating the detailed reaction mechanisms and simplified kinetic models. Laminar burning velocity is a key parameter that also helps describe various combustion phenomena such as flame stabilisation, flame flash back, flame blow out, and flame extinction [67,68]. The laminar burning velocity as a parameter appears in the modelling of various combustion phenomena and in the development of surrogate fuel models [69], in the prediction of pollutant formation through detailed mechanisms and in the accurate modelling of the combustion of fuel-oxidizer mixture under practical conditions, i.e., in combustion systems such as industrial furnaces, internal combustion engines, gas turbine combustors and rocket engines [67,70,71]. Although the laminar burning velocity is not a measurable quantity and is derived from other observables using different assumptions or theoretical models, this important parameter is used for scaling and modelling of the turbulent combustion flames.

The laminar burning velocity by definition is the velocity of a free flame propagating in the doubly infinite domain configuration and corresponds to the velocity at which the fresh premixed gas makes a planar flame steady. Therefore, this definition makes it suitable for calculations by 1-D computer codes using kinetic models, thermodynamic and transport properties as input parameters and hence leads to the validation of the kinetic schemes. It is, however, unfeasible to perform experiments with planar flames in the doubly infinite domain from both obvious practical and fundamental points of view. The non-quiescent unburned gas mixtures affect the planar flame propagation inside a confined space due to thermal expansion, flame wall interactions, acoustic pressure waves and buoyancy effects [72,73]. One of the main problems in measuring the laminar burning velocity is that a plane flame front can be observed only under special conditions.

3. Experimental Methods for Determining the Laminar Burning Velocity

Researchers from the combustion field have developed many experimental techniques to obtain the laminar burning velocity of fuel-air mixtures. The experimental techniques for laminar burning velocities determination, described in present work, are resumed in Table 1. The experiments realised on biogas-containing mixtures were made using both stationary and non-stationary flames. The stationary flames were studied by (a) burner method using Bunsen burners [38,45,48,74,75] or flat flames burners [24,59,76–78] or (b) the counter-flow twin flames method [79]. The non-stationary flames were studied by the constant volume method using cylindrical [22,23,26,44,46,80–82] or spherical vessels [5,20,21,25,81,83,84].

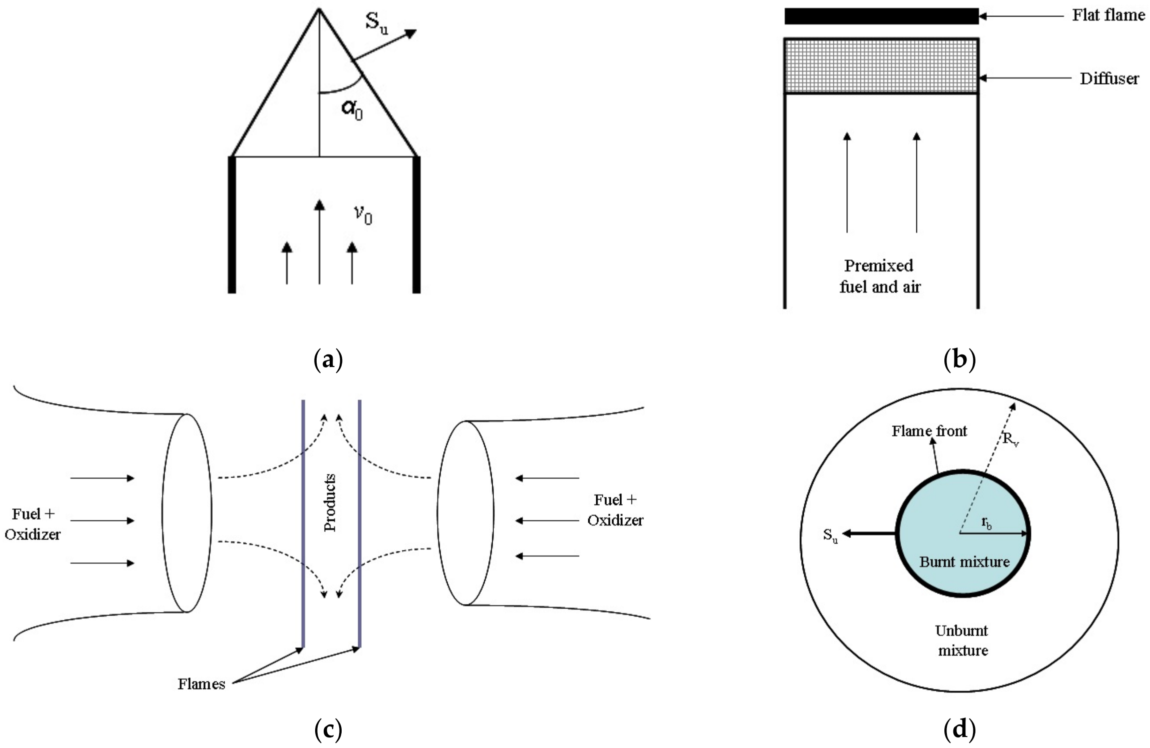
Table 1. Experimental techniques for laminar burning velocities determination described in present work.

Flame Type	Experimental Method		Flame Monitoring	Initial Conditions
Stationary	Burner method	Bunsen burner	Schlieren photography	Various pressures and compositions
		Flat flame burner	Schlieren photography	Over the whole flammability range
	Counter-flow twin flames		Dopler velocimetry particle Imaging velocimetry	Various pressures and temperatures
Non-stationary	Spherical flames	Constant volume method	Schlieren photography Pressure transducers Ionization gauges High speed camera	Various compositions, pressures and temperatures
		Constant pressure method	Schlieren photography Pressure transducers Ionization gauges High speed camera	Various compositions and temperatures

The Bunsen burner method is a simple method to measure the laminar burning velocity for a conical flame [85]. The method offers the possibility of obtaining the laminar burning velocities over various initial compositions and pressures. This method involves calculating the laminar burning velocities from photographs or tracings considering the volume rate of flow through the section of the flame surface being constant. A simple relation to obtain the laminar burning velocity, S_u, is:

$$S_u = v_0 \cdot sin\left(\frac{\alpha_0}{2}\right) \tag{1}$$

where v_0 represents the gas flow velocity in the central area of the burner tube and α_0 represents the angle of the cone formed by the front of the flame. However, the errors made in measuring of the laminar burning velocity are large, and the correct assessment of either the area of the flame front or the angle α_0 is difficult. In addition, it has been realised [86,87] that Bunsen flames are affected by different factors such as flame instability, stretch, curvature and heat loss. A schematic representation of the Bunsen burner is presented in Figure 2a.



(**a**) (**b**)

(**c**) (**d**)

Figure 2. Schematic representation of different experimental methods: (**a**) Bunsen burner; (**b**) Flat flame burner; (**c**) Counter-flow twin flames; (**d**) Closed vessel method.

The flat flame burner is obtained by mounting a "diffuser" to the ordinary burner that creates a constant profile of the gas flow rate, thus obtaining a flat flame. This method is used at low gas flow rates allowing to measure small values of the laminar burning velocities that occur in mixtures with the composition close to the explosion limit [88]. An interesting feature of this method is the appearance of an intense heat transfer between the flame and burner, which makes the explosive mixture suffer an appreciable preheating. Thermostating the upper part of the burner affords to maintain the unburned gas at the desired temperature and offers the possibility to achieve reproducible conditions for the production of these stationary flames. In addition, the introduction of the burner in an enclosure filled with inert gas allows measurements of normal combustion rates at different initial pressures and thus offers a considerable extension of this method. A schematic representation of the flat flame burner is presented in Figure 2b.

In the Figure 2c a diagram of the counter-flow twin flames method is presented. This method consists of two symmetric (twin) and planar flames generated on either side of a stagnation plane, at an offset distance, with the same chemical composition. This method has been used in the past in the laboratories being the most sensitive method regarding the changes in the flow characteristics [89–91]. The axial velocity profile is determined using different optical techniques such as laser Doppler velocimetry or particle image velocimetry. The advantage of this method is that the influence of stretch on the flame speed can be quantified and extracted by using the procedure proposed by Wu and Law [90]. However, it is difficult to use this method at pressures above 5 atm [64].

Another method used for determining the laminar burning velocity of biogas-air mixtures is the constant volume method, which measures the flame radius and/or pressure rise in the explosion vessel. At the early stage of flame propagation, the temperature gradients in both unburned and burned gas are small and can be neglected. Additionally, the flame remains spherical and the heat losses do not influence its development. For a given mixture, the flame speed can be obtained from a single test over a wide range of temperatures and pressures. Although the method has many advantages, it is found that the stretch effect is proportional to the Markstein length and inversely proportional to the flame size. Furthermore, for mixtures with Lewis numbers greatly deviating from unity, the stretch effect on flame speed is significant [92]. This method is suitable for laminar burning velocity measurements at high pressures and temperatures, which are close to relevant conditions from internal combustion engines and gas turbines [93,94]. To capture the images with the spherical expanding flames, the Schlieren measurement technique can be applied or a high-speed camera can be coupled to the experimental set-up. Records of the pressure and flame radius are used along with adequate correction procedures meant to consider the flame stretch [73,95]. The laminar burning velocity, S_u, can be obtained from pressure-time records by using specific equations. One of them is that which uses the cubic law coefficient of pressure rise (k) in the early stage of the flame propagation in a closed vessel by assuming isothermal compression of unburned gas ahead the flame front [96]:

$$S_u = R_v \left(\frac{k}{\Delta p_{\max}} \right)^{\frac{1}{3}} \left(\frac{p_0}{p_{\max}} \right)^{\frac{2}{3}} \tag{2}$$

In Equation (2) R_v is the radius of the explosion vessel; $\Delta p_{\max}$ is the peak pressure rise of the explosion at the initial pressure p_0; and $p_{\max} = \Delta p_{\max} + p_0$.

A diagram of the closed vessel method is drawn in the Figure 2d.

A comparison of experimental methods for determining the laminar burning velocity described in the present paper is given in Table 2.

Table 2. Comparison of experimental methods for determining the laminar burning velocity described in the present work.

Method	Advantage	Disadvantage
Bunsen burner [85]	- a simple method to measure the laminar burning velocity for a conical flame; - laminar burning velocities obtained over various initial compositions and pressures.	- the measuring of laminar burning velocity is affected by large errors; - flames are affected by flame instability, stretch, curvature and heat loss; - lack of uniformity of the burning velocity over the flame surface; - unburnt gas temperature profile is difficult to establish.
Flat flame burner [88]	- a simple method to measure the laminar burning velocity for a flat flame; - provides a close approximation to the ideal one-dimensional flat flame; - laminar burning velocities obtained over various initial pressures and compositions.	- is limited to measure low burning velocities; - flames are affected by stretch; - the exact position of the flame edge is uncertain; - heat transferred between the flame and burner can preheat the mixture.

Table 2. *Cont.*

Method	Advantage	Disadvantage
Counter-flow twin flames [89–91]	- the most sensitive method regarding the changes in the flow characteristics; - two symmetric (twin) and planar flames with the same chemical composition are used; - velocity profile can be determined using optical techniques (Doppler velocimetry or particle image velocimetry); - flame stretch is quantified and extracted using a dedicated procedure.	- the method is difficult to use at pressures above 5 atm; - the uncertainty in laminar burning velocity due to thermocouple measurements depends on the rate of heat production in the flame.
Closed vessel method [93–96]	- measurements of flame radius and/or the pressure rise variation in time; - vessels with different shapes and sizes; - laminar burning velocities obtained over various initial compositions, pressures and temperatures; - laminar burning velocity can be obtained from a single test.	- equipments involved are relatively complex and expensive; - correction procedures for flame stretch are necessary; - laminar burning velocity is obtained using specific equations which involve many parameters; - radiative loss to the vessel walls and convective loss to the spark electrodes occurs.

More details regarding the accuracy, uncertainties, strengths and weaknesses of these methods can be found in the review papers of Egolfopoulos et al. [64], Konnov et al. [73] and Faghih and Chen [93].

4. Computing Methods

The laminar burning velocities of biogas-air flames can be obtained from the computation on one-dimensional freely propagating flames using different dedicated programs and various mechanisms, at various initial conditions (initial temperature and pressure, fuel concentration, diluent concentration). These numerical calculations are required to validate the experimental results. Computing programs try simulating the reaction pathways using one or more kinetic mechanisms. There are some programs for kinetic modelling of the combustion, many of them being suitable for methane combustion.

For biogas-containing mixtures, several authors used the CHEMKIN package together with GRI-Mech mechanism (version 2.11 or 3.0) [22–24,26,41,45,56,74–76,81,97] while other authors used USC Mech II mechanism [22,79]; San Diego mechanism [23,24]; C1-C3 mechanism [56] or Le Cong mechanism [77].

Goodwin et al. [98] used the CANTERA code and three chemical kinetic mechanisms: GRI-Mech 3.0, USC Mech II and San Diego mechanism.

Another set of papers reports laminar burning velocities obtained from simulations performed using the REGATH package [39] or COSILAB package [29,99] with GRI 3.0 chemical scheme.

The GRI Mechanism (versions 2.11 or 3.0) is an optimized mechanism designed to model natural gas combustion, which includes NO formation and reburn chemistry.

This mechanism provides sound basic kinetics that also furnish the best combined modeling predictability of basic combustion properties. The GRI-Mech 3.0 mechanism [100] can be adjusted to methane combustion at various initial pressures (between 0.1 and 10 bar) and is based on 53 species (including argon) and 325 chemical reactions.

The USC Mech II mechanism considers 111 species and 784 chemical reactions relevant to C1–C4 hydrocarbons and syngas oxidation at high temperature [101]. It applies to a wide variety of combustion scenarios, incorporating the recent thermodynamic, kinetic, and species transport updates relevant to high-temperature oxidation of hydrogen, carbon tmonoxide, and C1–C4 hydrocarbons.

The San Diego mechanism is optimised for autoignition and diffusive flames of different hydrocarbons and considers 37 species and 177 chemical reactions [102]. It applies to various experimental conditions: temperatures between 1000 K and 3000 K; pressures up to 10 atm; equivalence ratios between 0.5 and 2.0.

Qin et al. [103] developed the C1-C3 mechanism for combusting light hydrocarbons such as CH_4, C_2H_4, C_2H_6, C_3H_4, C_3H_6 and C_3H_8 optimising the rate parameters of 258 reactions. This detailed chemical reaction mechanism is conceptually structured hierarchically with H_2 and CO chemistry at the base, supplemented by elementary reactions of larger chemical species.

The Le Cong mechanism consists of 18 species and 66 reactions, as described by Le Cong et al. [104,105]. This mechanism is used to simulate the one-dimensional freely propagating flame, determining the premixed, laminar flame speed of the gaseous mixtures. It is designed for modelling the oxidation of hydrogen, CO, methane, methanol, formaldehyde, and natural gas over a wide range of conditions, including jet-stirred reactor, flame, shock tube, and plug flow reactor.

The Konnov mechanism used in [78] consists of 127 species and 1027 reactions. It is a detailed reaction mechanism, which accurately describes pyrolysis, ignition, oxidation and detonation of many small hydrocarbons and nitrogen-containing fuels reacting with different oxidizers over a wide range of conditions typical for combustion processes.

5. Initial Conditions of Studied Biogas-Air Mixtures

Whether the studies reported in the literature on laminar burning velocity of biogas-air mixtures were conducted experimentally or by numerical modelling, these were performed at various initial pressures (ranging from 0.5 to 18 bar), temperatures (between 295 and 661 K), and compositions (equivalence ratios ranging from 0.4 to 1.6 and CO_2 dilution between 0 and 70%).

All of the aforementioned conditions for obtaining the laminar burning velocity of biogas-air blends, described in the present work, are summarised in Table 3.

Table 3. Initial conditions for obtaining the laminar burning velocities of biogas-air and synthetic biogas (CH_4/CO_2)-air flames described in the present work.

Author	Equivalence Ratio, φ	Initial Pressure, p_0	Initial Temperature, T_0	$[CO_2]$ %
Anggono et al. [20,21,26,84]	0.5–1.4	0.5; 1; 3 bar	298 K	30.6
Pituzzi et al. [23]	0.7–1.1	1–5 bar	298 K	35–55
Yadav et al. [24]	0.7–1.4	1 bar	298 K	5–50
Hinton and Stone [25]	0.7–1.4	1–18 bar	380–660 K	0–40
Mitu et al. [29]	0.6–1.3	0.5–2 bar	298 K	0–17.5
Cohé et al. [38]	0.6	1–9 bar	298 K	0–40
Patino et al. [39]	0.7–1.3	1 bar	298 K	0–50
Ren et al. [41]	0.8–1.2	1 bar	398 K	0–40
Acero-Caballero et al. [43,45]	1.0–1.1	1 bar	298 K	34–40
Suhaimi et al. [44]	0.4–0.9	1 bar	300 K	50
Wei et al. [46]	0.9–1.3	1 bar	298 K	40–60
Nurmukan et al. [48]	0.8–1.3	1 bar	298–440 K	30–40
Quintino et al. [49]	0.8–1.0	1 bar	298 K	0–20

Table 3. *Cont.*

Author	Equivalence Ratio, φ	Initial Pressure, p_0	Initial Temperature, T_0	$[CO_2]$ %
Cardona and Amell [56]	0.6–1.5	0.85 bar	295 K	34
Hu et al. [74,75]	0.6–1.4	1 bar	300; 400; 543 K	25; 35
Kishore et al. [76]	0.8–1.3	1 bar	307 K	0–60
Chan et al. [77]	0.8–1.4	1 bar	298 K	0–15
Nonaka and Pereira [78]	0.7–1.4	1 bar	298 K	0–50
Park et al. [79]	0.75–1.25	1; 2; 4 bar	298 K	25; 45
Halter et al. [80]	1.0	1 bar	300 K	0–20
Xie et al. [81]	0.4–1.6	1; 2; 3 bar	300 K	0–60
Bai et al. [82]	0.8–1.2	0.5–6.9 bar	298–661 K	0–60
Stone et al. [83]	0.6–1.4	0.5–10.4 bar	295–454 K	0–60
Zahedi and Yousefi [97]	0.7–1.3	1–5 bar	298 K	0–20
Boushaki et al. [99]	0.7–1.3	1 bar	298 K	0–50
Zhen et al. [106]	0.8–1.2	1 bar	298 K	40–60
Qin et al. [107]	0.65–0.75	1 bar	300 K	0–50

6. Discussions

6.1. *Laminar Burning Velocities of Biogas-Air Flames*

When compared to methane, biogas has narrower flammability limits, a lower heating value and lower burning velocity, mainly due to the presence of a high amount of carbon dioxide. Moreover, biogas is resistant to self-ignition due to its higher auto-ignition temperature [34]. Therefore, biogas presents certain problems related to flame instability when it is used in conventional combustion systems. To understand and avoid such problems, the laminar burning velocities of biogas-air mixtures (either synthetic biogas or not) were studied by many authors, using different experimental and/or numerical methods. The experimental and numerical methods for obtaining the laminar burning velocity of biogas-air mixtures used in the present paper are summarised in Table 4.

Stone et al. [83] reported data from experiments in a closed vessel for several composition of biogas (80% CH_4–20% CO_2; 67% CH_4–33% CO_2; 50% CH_4–50% CO_2; 40% CH_4–60% CO_2), at 1 bar and 298 K under microgravity conditions. They developed a correlation for the laminar burning velocity of CH_4-CO_2-air blends with various equivalence ratios. The correlation was applied for concentrations of CO_2 up to 60%, initial pressures of up to 10 bar and initial temperatures up to 494 K. For the stoichiometric mixture and ambient initial conditions, the laminar burning velocity of biogas composition of 80% CH_4–20% CO_2 was found to be 31.5 cm/s; for 50% CH_4–50% CO_2 the laminar burning velocity was found to be 17.7 cm/s, while for 40% CH_4–60% CO_2 the laminar burning velocity was reported 13.0 cm/s.

Ju et al. [108] determined the laminar burning velocities of synthetic mixtures (equivalence ratio 0.5–1.0) of CH_4 and CO_2 (CO_2: 10, 20 and 30 vol%) by kinetic modelling, adopting two models: the optically thin model and the heat reabsorption model. The last model considers the spectral radiation in the one-dimensional coordinate of heat reabsorption from H_2O and CO_2 that approximately halves the net heat loss and lead to reduce the lower flammability limit and influence the laminar burning velocity. A good agreement between experiments and data from reabsorption model was found.

The laminar burning velocities of CH_4-CO_2-air mixtures with various equivalence ratios (0.65–0.75) and CO_2 content up to 50 vol% were experimentally and numerically determined, at atmospheric pressure, using a stagnation-flow experimental configuration and the detailed GRI 2.11 chemical kinetic mechanism [107]. The results indicate that, for the same equivalence ratio, addition of CO_2 to the fuel significantly reduces the laminar burning velocity. For example, for a flame with equivalence ratio of 0.75, when the carbon dioxide content varies from 0 to 50%, the experimental value of laminar burning velocity decreases from 23.5 cm/s (mixtures without CO_2) to 13.0 cm/s (mixtures with 50%

CO_2). The experimentally measured laminar burning velocities were, in general, in good agreement with the predicted values.

Table 4. Summary of experimental and computed methods for obtaining the laminar burning velocity of biogas-air mixtures presented in the present study.

Type of Method	Method/Model/Mechanism Name		Reference
Experimental	Bunsen burner		Cohé et al. [38]
	Contoured slot burner		Cardona and Amell [56]
	Flat flame burner		Kishore et al. [76] Chan et al. [77] Nonaka and Pereira [78] Zahedi and Yousefi [97] Qin et al. [107]
	Counter-flow twin flames		Park et al. [79]
	Closed vessel		Anggono et al. [20,21,26,84] Pituzzi et al. [23] Hinton and Stone [25] Mitu et al. [29] Halter et al. [80] Bai et al. [82] Stone et al. [83]
Numerical	CHEMKIN software	GRI-Mech 2.11	Cardona and Amell [56]
		GRI-Mech. 3.0	Anggono et al. [26] Cohé et al. [38] Ren et al. [41] Halter et al. [80] Zahedi and Yousefi [97]
		C_1-C_3 mechanism	Cardona and Amell [56]
		Le Cong mechanism	Chan et al. [77]
	Cantera package	GRI-Mech. 3.0	Bai et al. [82]
	1D Premix code	GRI-Mech 2.11	Qin et al. [107]
		GRI-Mech. 3.0	Pituzzi et al. [23] Kishore et al. [76]
		USC-Mech 2.0 mechanism	Park et al. [79]
		San Diego mechanism	Pituzzi et al. [23]
	Chem1D code	GRI-Mech. 3.0	Nonaka and Pereira [78]
		USC-Mech 2.0 mechanism	Nonaka and Pereira [78]
		San Diego mechanism	Nonaka and Pereira [78]
		Konnov mechanism	Nonaka and Pereira [78]
	REGATH package	GRI-Mech. 3.0	Patino et al. [39]
	COSILAB code	GRI-Mech. 3.0	Mitu et al. [29] Boushaki et al. [99]

A study by Kishore et al. [76] focused on the determination of the laminar burning velocity of natural gas by the heat flux method. This study presented the effects of CO_2 (0–60%) on flame properties at various equivalence ratios (0.8–1.3), temperature of 307 K, and atmospheric pressure. Computations were also conducted using the steady one-dimensional laminar premixed flame code PREMIX, together with GRI-Mech 3.0 reaction mechanism. At the stoichiometric equivalence ratio, the experimental laminar burning velocity decreases from 30.7 cm/s for mixtures containing 20 vol% CO_2 to 23.5 cm/s for mixtures with 40% CO_2 and to 14.5 cm/s for mixtures containing 60% CO_2. The numerical results agree well with the experimental data for mixtures with high content of carbon dioxide compared to the mixtures with 20 vol% CO_2.

An experimental study on a lean CH_4-CO_2-air mixture (equivalence ratio equal to 0.6), with different carbon dioxide content (up to 40%), at various initial pressures, was conducted using both, laminar and turbulent Bunsen flame configurations [38]. Data provided by Chemkin package using GRI-Mech 3.0 mechanism complete the experimental results. All data indicated a decrease of the laminar burning velocities with increasing CO_2 dilution rate. The effect of initial pressure on laminar burning velocities was found to be more pronounced than the carbon dioxide content. Moreover, the computations slightly underestimate the reduction of laminar burning velocity caused by the CO_2 content.

The laminar burning velocities of biogas-air mixtures (composition of biogas: 55% CH_4–45% CO_2; 75% CH_4–25% CO_2) were studied experimentally and numerically in the counter-flow configuration at various initial equivalence ratios (between 0.7–1.3), initial pressures ranging from 1 to 4 atm, and ambient initial temperature [79]. The laminar burning velocity is reduced with pressure and CO_2 content increase. The experimental values of laminar burning velocity for stoichiometric mixtures, at standard conditions, are 21.7 cm/s for biogas composition of 55% CH_4–45% CO_2 and 29.5 cm/s for biogas composition of 75% CH_4–25% CO_2. The numerical simulations with PREMIX code and USC Mech II kinetic model reproduced closely the experimental values of the laminar burning velocities over all experimental conditions.

Spherically expanding flames propagating at constant pressure were employed to determine the laminar burning velocity and flammability characteristics of biogas-air mixtures [20,21]. The composition of biogas consisted from 66.4% methane, 30.6% carbon dioxide and 3% nitrogen. The laminar burning velocities were measured at various equivalence ratios (0.5–1.3), at room temperature and atmospheric and sub-atmospheric pressure (0.5 bar). For the ambient initial conditions and equivalence ratio of 1.0, the authors reported a laminar burning velocity of 26.4 cm/s. They found that at sub-atmospheric initial pressure the diffusion time of reactants decreases and consequently, the laminar burning velocity becomes higher than that at atmospheric pressure.

Cardona and Amell [56] measured the laminar burning velocity for biogas-air mixture (66% CH_4–34% CO_2) and biogas-C_3H_8-H_2 mixture (33% CH_4–17% CO_2–40% C_3H_8–10% H_2) with normal and oxygen-enriched air, at 0.828 bar and 295 K, for various equivalence ratios (0.6–1.5). For the stoichiometric biogas-air mixture, they found that the laminar burning velocity is 28.5 cm/s. The GRI-Mech 3.0 and C_1-C_3 reaction mechanisms were used to perform numerical simulations. They observed that the predictions of the GRI-Mech 3.0 mechanism agree quite well the experimental results, except those of oxygen-enriched biogas combustion, where the differences can be due to the high amount of CO_2 in the fuel mixture. The C_1-C_3 reaction mechanism underestimates the laminar burning velocities at all studied mixtures.

Results for higher pressures (up to 18 bar), temperatures (up to 660 K) and equivalence ratios in the range 0.7–1.4 have been obtained using the pressure rise method in a constant volume [25]. A correlation with twelve coefficients fitted the results. The data showed a decrease in the laminar burning velocity with initial pressure and an increase with initial temperature. For the stoichiometric mixture and standard conditions the laminar burning velocity for 60% CH_4_40% CO_2 is 22.5 cm/s wile for 80% CH_4_20% CO_2 is 29.5 cm/s.

Nonaka and Pereira [78] experimentally measured the laminar burning velocity of biogas and calculated it for various CO_2 concentrations (0–50%). The experiments were undertaken using the heat flux method, at 298 K and 1 bar, and were supplemented with data from numerical modelling using four detailed reaction mechanisms: San Diego, USC-Mech II, GRI-Mech 3.0 and Konnov. Their results showed that carbon dioxide presence reduces the laminar burning velocity and that the shifting of the maximum burning velocity towards leaner mixtures is determined by a combination of CO_2 dilution and reaction effects. For the most studied composition of biogas (60% CH_4–40% CO_2), the experimental values of laminar burning velocity vary from 16 cm/s for a lean mixture (equivalence ratio 0.8) to 22.0 cm/s for the stoichiometric mixture, and to 13.0 cm/s for a rich mixture (equivalence ratio 1.3). The results obtained using the GRI-Mech 3.0 mechanism agreed well with measurements for mixtures with 10% and 20% CO_2. For mixtures with 30%, 40% and 50% CO_2, the Konnov mechanism presented the best agreement with the experimental data.

Another study [84] was conducted in a spherical closed vessel to find the effect of CO_2 on biogas laminar burning velocities at various initial pressures and equivalence ratios (0.5–1.4) and ambient initial temperature. The composition of CO_2 was 25 and 50%, respectively. The results showed that the laminar burning velocity, at the same equivalence ratio and initial pressure, declined in respect with the increased level of carbon dioxide. For example, for the stoichiometric mixtures at atmospheric initial pressure, the laminar burning velocity is 28.9 cm/s for mixture with 25% CO_2 and 18.9 cm/s for mixture with 50% CO_2.

Carbon dioxide dilution on the laminar burning velocity of methane-air mixture was studied using kinetic modelling to understand the effect of CO_2 concentrations (0 to 50 vol%) on the combustion characteristics of biogas [39]. The simulations were performed using the REGATH package with GRI 3.0 chemical scheme. The computations on mixtures with equivalence ratios from 0.7 to 1.3 were made at initial pressure of 1 bar and an inlet temperature of 298 K. The results showed that increase in CO_2 concentrations reduced the concentrations of the reactants, decreasing the net reaction rate and thus the flame speed.

Pituzzi et al. [23] studied the laminar burning velocity of twelve synthetic biogas mixtures in a closed vessel. Initial pressure between 1 and 5 bar, equivalence ratios between 0.7 and 1.1, and a percentage dilution of a mixture of CO_2 and N_2 between 35 and 55%, have been considered. A comparison with simulation using PREMIX for both GRI-Mech 3.0 and San Diego mechanisms has provided closer agreement for mixtures with the equivalence ratio closer to the stoichiometry, whereas for the equivalence ratio of 0.7 the deviation is larger than 15% for all pressures. The authors noted that the lower values of laminar burning velocity were recorded for the mixtures with lower equivalence ratio, higher dilution percentage and higher initial pressure.

A study was conducted by Anggono et al. [26] on the stoichiometric mixture using both numerical and experimental methods. The experiments were made in a closed combustion chamber under various initial mixture pressures (1 bar and 3 bar), 298 K and CO_2 concentrations up to 70% to simulate the conditions of a combustor working at high pressure conditions. In addition, numerical simulations were carried by CHEMKIN-PRO package with GRI-Mech 3.0 employed as mechanism. The results showed that the laminar burning velocity of methane-air mixtures decreased with an increase in CO_2 concentration and mixture pressure. At 70% CO_2 the experimental laminar burning velocity at 1 bar is 8.0 cm/s, while at 3 bar decrease to 4.3 cm/s. The experimental and computed data agree well.

Bai et al. [82] studied the laminar burning velocities of biogas over various initial temperatures (300–661 K), pressures (0.5–6.9 bar) and equivalence ratios (0.8–1.2) with four different CO_2 concentrations (0%, 20%, 40%, and 60%) using the closed vessel method. Based on the measured laminar burning velocities, two power law correlations for two concentrations of carbon dioxide have been developed. The experimental laminar burning velocities have been compared with the simulation results of Cantera software with GRI-Mech 3.0 mechanism. The comparison showed a good agreement between experimental and computed data. The authors concluded that both pressure and CO_2 concentration present negative effect on the laminar burning velocity. Alternatively, temperature positively affects the laminar burning velocity. They also mention that there is a peak value of laminar burning velocities for each CO_2 concentration, and the equivalence ratios related to the maximum burning velocity shift to lean mixtures with increasing CO_2 concentration. It is also found that the equivalence ratios of the peak burning velocities with low CO_2

concentrations (0–20%) do not change. For high CO_2 concentrations (40–60%), the equivalence ratios at maximum burning velocity shift towards lean mixtures with increasing temperature. The experimental laminar burning velocities of stoichiometric biogas-air mixtures, at standard conditions, obtained by Bai et al. [82] are 30.5 cm/s for 20% CO_2; 22.3 cm/s for 40% CO_2; 13.0 cm/s for 60% CO_2.

Numerical calculations of biogas-air flames are performed using the 1-D COSILAB code using GRI-Mech 3.0 mechanism [99]. Calculations include laminar flame velocity, flame temperature and CO-NO_x emissions with different parameters, such as temperature and pressure of fresh gases, equivalence ratio and gas composition. Several reaction mechanisms were used and compared to the results from the literature. The runs were performed for equivalence ratios between 0.7 and 1.3, CO_2 content up to 50 vol%, 298 K and 1 bar. Results showed that calculations with some of these mechanisms reproduce well experimental results of the literature. But also that, CO_2 addition induces a decrease in laminar burning velocity, an increase in CO emissions and a decrease in NO_x formation.

The effects of adding CO_2 on the laminar premixed combustion characteristics of methane were simulated using the premixed free-propagating flame model based on the CHEMKIN II taken into account the Soret effect [41]. The GRI-Mech 3.0 mechanism was selected to analyse the laminar premixed combustion characteristics of the mixtures with different CO_2 dilutions (0–40 vol%) and equivalence ratios (0.8, 1.0 and 1.2), at an initial temperature of 398 K and an initial pressure of 1 bar. The authors concluded that both the physical and chemical effects of CO_2 reduce the laminar burning velocity. The physical effects are greater than the chemical effects.

The variation of the laminar burning velocities of biogas-air mixtures with various equivalence ratios, at different CO_2 content and standard initial conditions compiled from the above studies are given in Figure 3 for experimental data and in Figure 4 for numerical modelling. As we expected, the dependence of the laminar burning velocity on the biogas composition has the shape of a parabola with the concavity orientated towards the axis of the composition. For the mixtures with higher content of carbon dioxide (e.g., 60 %vol), the maximum of the parabola is not recorded at the stoichiometric composition corresponding to the complete combustion of the biogas to CO_2 and H_2O, but at mixtures with a concentration slightly higher than the stoichiometric concentration. For the mixtures with lower CO_2 content (such as 20 %vol) the maximum of the parabola is recorded at stoichiometric mixture, no matter if the data are obtained from measurements or computation. For the same initial conditions (composition, pressure and temperature), the maximum value of laminar burning velocity corresponds to the maximum value of flame temperature. At this moment the maximum amount of heat is released by the fuel mixture, according to the competition between exothermic and endothermic processes within the flame.

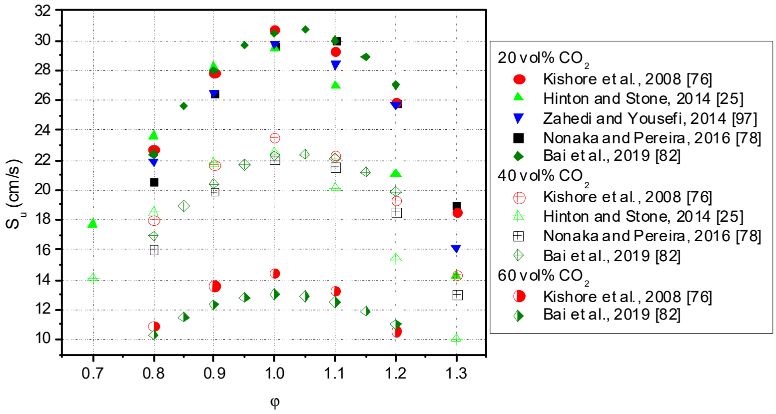


Figure 3. Variation of the experimental laminar burning velocities of biogas-air mixtures with various equivalence ratios, at different CO_2 contents and standard initial conditions.

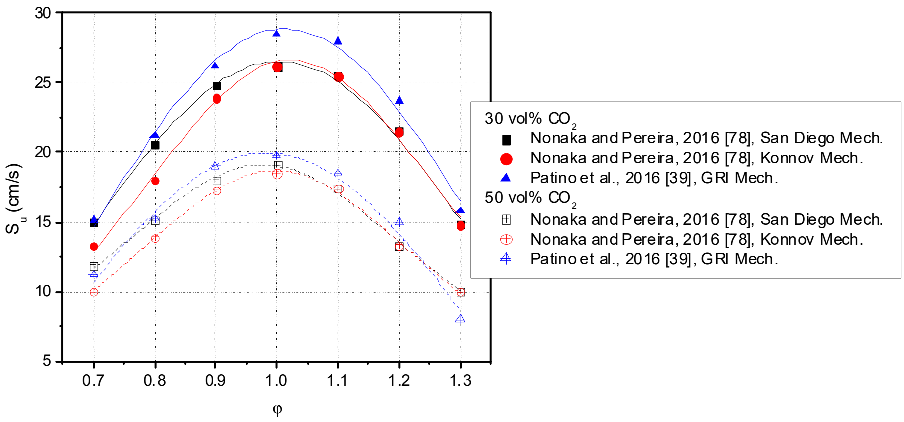


Figure 4. Variation of the calculated laminar burning velocities of biogas-air mixtures with various equivalence ratios, at different CO_2 contents and standard initial conditions.

Beside the above mentioned studies that reported results from experiments and numerical modelling on the laminar burning velocity of biogas (or synthetic biogas) with a high CO_2 dilution, studies were conducted with small amounts of carbon dioxide. One can mention Halter et al. [80] who conducted an experimental and numerical study to obtain the CO_2 dilution effect on the laminar burning velocity of stoichiometric methane-air mixture. The experiments were made in a closed combustion vessel on stoichiometric mixtures diluted with CO_2 up to 20% and ambient initial conditions. The computations were made using the CHEMKIN package with GRI-Mech 3.0 mechanism.

The influence of CO_2 (10 and 20 vol%, respectively) on the laminar burning velocity for CH_4-air mixtures at various equivalence ratios (0.7–1.3), various initial pressures (1–5 bar) and initial temperature of 298 K, was reported by Zahedi and Yousefi [97] from experiments using a flat flame burner and numerical modelling using CHEMKIN package together with the GRI-Mech 3.0 mechanism.

Chan et al. [77] realised experiments using a flat-flame burner at atmospheric pressure and 298K over the methane-air equivalence ratio range of 0.8 to 1.4 and carbon dioxide content of 10 and 15%, respectively. A kinetic modelling was also performed using CHEMKIN-PRO package with the skeletal version of Le Cong kinetic mechanisms.

Flame propagation was studied in methane-air-CO_2 mixtures with various initial pressures and compositions using pressure-time records obtained in a closed vessel with central ignition [29]. The laminar burning velocities obtained from experiments were compared with those obtained from numerical modelling of 1D flame with COSILAB package and GRI-Mech 3.0 kinetic mechanism.

Regardless of the amount of carbon dioxide contained in the biogas, all authors noted that the presence of carbon dioxide in the biogas flames decreases the laminar burning velocity. This can also be observed from Figure 5 where data referring to experimental laminar burning velocities are plotted. In this figure, the laminar burning velocities for mixtures with equivalence ratio of 1.1, at ambient initial conditions vs. carbon dioxide content are given.

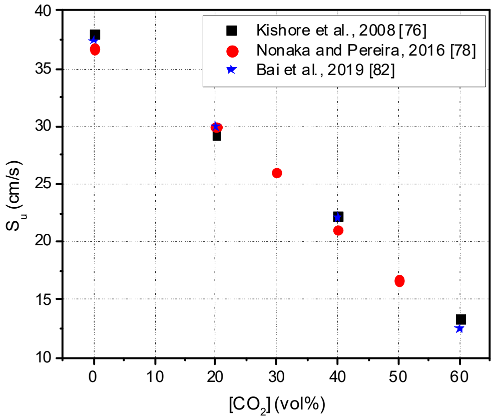


Figure 5. Variation of experimental laminar burning velocities with carbon dioxide content for CH_4-CO_2-air mixtures with equivalence ratio of 1.1, at ambient initial conditions.

Decreasing of the laminar burning velocity with increase in carbon dioxide content is also observed when examining data from numerical modeling. Figure 6 gives experimental and numerical data from Yadav et al. [24] and the calculated data from Boushaki et al. [99].

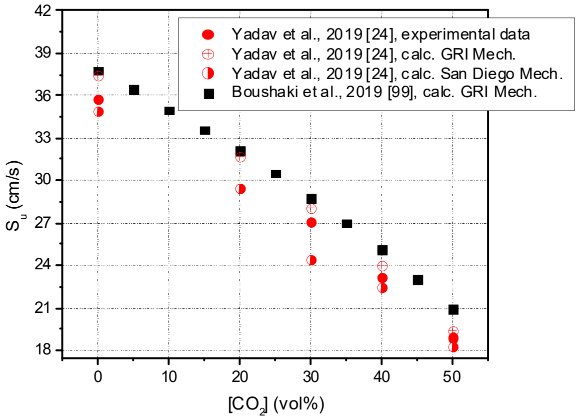


Figure 6. Variation of experimental and calculated laminar burning velocities with carbon dioxide content for mixtures with stoichiometric equivalence ratio, at standard initial conditions.

The decrease of the laminar burning velocity with the increase of CO_2 content occurs due to three effects: the dilution effect, the thermal-diffusion effect and the chemical effect. By adding carbon dioxide to a mixture, the fuel and oxidant content in the mixture are reduced, decreasing the available amount of released heat and the reaction rate. The high CO_2 heat capacity reduces also the adiabatic flame temperature and, hence, the reaction rates, as mentioned by Kishore et al. [76].

The decrease in the adiabatic flame temperature with CO_2 content increase is observed from Figure 7 where data reported by Boushaky et al. [99] with COSILAB code and Ren et al. [41] with CHEMKIN II package for stoichiometric biogas-air mixture at 298 K and 1 bar are given. Xie et al. [81] noted that the flame temperature does not decrease linearly with the increase of the CO_2 content. For a mixture with a higher CO_2 content, the carbon dioxide has a stronger thermal effect. Meanwhile, the mixtures at lean or rich ratios show a stronger thermal effect than that at stoichiometric ratio.

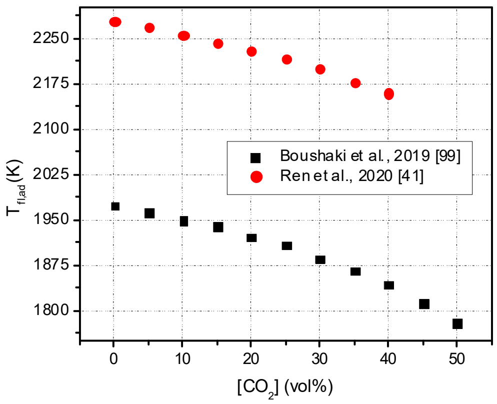


Figure 7. Adiabatic flame temperature variation with CO_2 content for the stoichiometric biogas-air mixture, at 298 K and 1 bar.

In addition to a decrease of flame temperature, a decrease in NO_x emissions by increasing CO_2 content in the biogas mixtures was observed [99]. This observation can be beneficial to reduce the NO_x emissions provided by the internal combustion engines by redirecting a part of the exhaust gases into the intake manifold, lowering the combustion temperature and hence decrease of NO_x emissions.

As mentioned by Ren et al. [41], CO_2 is involved in the reaction $CO_2 + H \Leftrightarrow CO + OH$. This reaction is competing for H atoms in reactions $H + O_2 \Leftrightarrow OH + OH$ and $H + O_2 \Leftrightarrow O + OH$, which increase the OH/H ratio. The increase of OH/H ratio affects the propagation of the reaction chain and reduces the overall combustion reaction rate, and thus, the laminar burning velocity. The decrease of this parameter is more pronounced at rich mixtures (equivalence ratios > 1.0). The presence of CO_2 in biogas mixtures reduces the laminar burning velocity due to its physical and chemical effects. The authors mentioned that the physical effects were higher than the chemical effects. Additionally, under the flame condition, CO_2 can dissociate leading to an increase in its inerting effect. Thus, examination of the total mass fraction of H and OH radicals, which are the main chain carriers and influence most significantly the fuel consumption reaction, gives us information regarding the influence of CO_2 dissociation on flame propagation, as Movileanu et al. pointed out [109]. Halter et al. [80] concluded that the dissociation of CO_2 is not negligible, but the effect of its dissociation is less important compared with additive amount increase. This is due to the fact that the flame temperature is lower and does not support an extensive dissociation.

Beside the carbon dioxide content influence on laminar burning velocity, the initial pressure affects also this combustion parameter. At constant equivalence ratio, carbon dioxide content and initial temperature, the laminar burning velocity decreases with the initial pressure increase as we can observe from Figure 8, where experimental and computed data delivered by Park et al. [79] on the biogas mixture (55 % CH_4–45 % CO_2) at 298 K and on the lean, stoichiometric, and rich mixtures are plotted. This behaviour can be attributed to the fact that the effect of CO_2 addition is insignificant on the flame wrinkling parameters and flame surface density, while the pressure increase strongly influences both parameters. Another representative set of data compiled from Hinton and Stone [25] is represented in Figure 9, for stoichiometric equivalence ratio, biogas composition of 60% CH_4–40 % CO_2 and initial temperature of 400 K. The decrease of the laminar burning velocity with the initial pressure increase is not linear, this decrease is rather exponential. According to results of Hu et al. [110] on H_2-air flames the decrease of laminar burning velocity with the increase of initial pressure can be understood by the suppression of overall chemical reaction due to the decrease of H and OH mole fractions in flames. Hu et al. [110] found strong correlations between the laminar burning velocity and the maximum radical concentrations of H and OH radicals in the reaction zone of premixed flames of H_2-air flames and concluded that high laminar burning velocities correspond to high radical concentrations in the reaction zone.

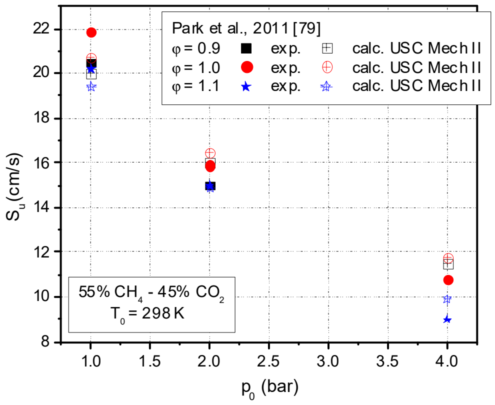


Figure 8. Initial pressure influence on laminar burning velocity, at various equivalence ratios and 298 K.

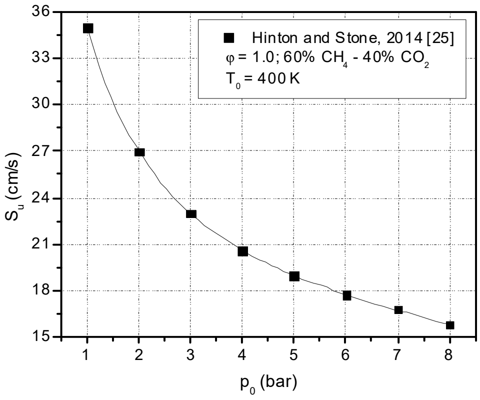


Figure 9. Initial pressure influence on experimental laminar burning velocity for stoichiometric equivalence ratio and 400 K.

If the increase of the initial pressure decreases the laminar burning velocity, the increase of the initial temperature has an opposite effect. This behaviour is represented in Figure 10, where data from Hinton and Stone [25] for stoichiometric equivalence ratio, biogas composition of 60% CH_4–40 % CO_2 and initial pressure of 5 bar are given. Data from Bai et al. [82] at various equivalence ratios, ambient initial pressure and two initial temperatures are given in Figure 11. Bai et al. [82] noted that the initial temperature has a positive effect on the laminar burning velocity for all conditions, with all carbon dioxide concentration.

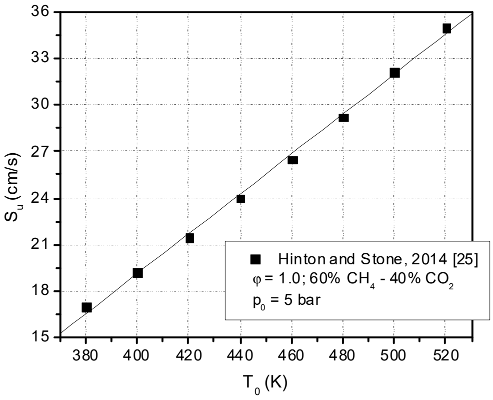


Figure 10. Initial temperature influence on experimental laminar burning velocity for stoichiometric equivalence ratio and 5 bar initial pressure.

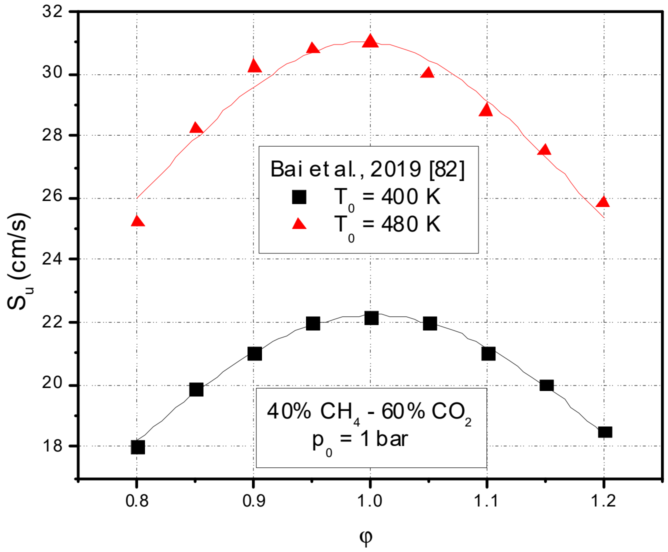


Figure 11. Initial temperature influence on experimental laminar burning velocity, at various equivalence ratios and 1 bar initial pressure.

6.2. Effect of Hydrogen Addition on the Laminar Burning Velocity of Biogas-Air Flames

Because the biogas has a low heating value, some studies have turned their attention to improve its relatively poor combustion characteristics. To improve the combustion properties of the biogas, it can be mixed with another combustible gas whose combustion properties, mainly the flame velocity, are greater. Given that, the hydrogen, which is a high-quality fuel with zero greenhouse potential, was chosen by some researchers to be added to biogas. The results showed a higher heat release rate, an improved stability of the flame and an increased flame temperature [42,111,112]. A significant improvement in combustion properties was observed by adding a trace amount of hydrogen into the biogas mixtures causing the acceleration of the reaction rate which counteracts the dilution effect of the large amount of CO_2 from biogas [111].

The experimental and numerical methods for obtaining the laminar burning velocity of biogas- H_2-air mixtures presented in this work are summarised in Table 5.

Table 5. Summary of experimental and numerical methods for obtaining the laminar burning velocity of biogas-H_2-air mixtures presented in the present study.

Type of Method	Method/Model/Mechanism Name		Reference
Experimental	Bunsen burner		Acero-Caballero et al. [45] Nurmukan et al. [48] Zhen et al. [111]
	Flat flame burner		Yadav et al. [24]
	Closed vessel		Suhaimi et al. [44] Wei et al. [46]
Numerical	CHEMKIN software	GRI-Mech 2.11	Acero et al. [43] Acero-Caballero et al. [45]
		GRI-Mech. 3.0	Yadav et al. [24] Acero-Caballero et al. [45] Wei et al. [46]
		San Diego mechanism	Yadav et al. [24] Wei et al. [46]
	ANSYS Fluent software	reduced GRI-Mech 3.0	Nurmukan et al. [48]
	Cantera package	USC-Mech 2.0 mechanism	Quintino et al. [49]

Zhen et al. [106] measured the laminar burning velocities of biogas-H_2-air mixtures (biogas compositions 60% CH_4–40% CO_2; equivalence ratios from 0.8 to 1.2) using a Bunsen burner. The H_2 content varied from 10 to 50%. The authors determined that there was a corresponding increase in the biogas burning rate with H_2 percentage increase. For the stoichiometric biogas-H_2-air mixture the laminar burning velocity increases from 25 cm/s for 20% H_2, to 37 cm/s for 50% H_2. For rich (equivalence ratio 1.2) biogas-H_2-air mixture, the laminar burning velocity increases from 19 cm/s for 10% H_2, to 47 cm/s for 50% H_2. For lean (equivalence ratio 0.8) biogas-H_2-air mixture, the flame is stable only at 50% H_2, and thus the corresponding laminar burning velocity is 25 cm/s.

Another study [43] was conducted to find the effect of hydrogen addition to biogas (60% CH_4–40% CO_2) on the laminar burning velocity using the CHEMKIN software and GRI-Mech 2.11 mechanism. The runs were performed under normal conditions using different aeration factors. The H_2 content from mixtures was 5% and 15%, respectively. The results showed that the hydrogen addition to the biogas generates an acceleration of the combustion reaction and an increasing of the laminar burning velocity. For stoichiometric mixture (aeration factor of 1), the laminar burning velocity increase from 27.4 cm/s for pure biogas, to 28.5 cm/s for biogas with 5% H_2 and to 32.2 cm/s for biogas with 15% H_2. For the lean mixtures (aeration factor of 1.4), the effect of hydrogen addition on the laminar burning velocity leads to a small increase, from 15.8 cm/s for pure biogas, to 18.4 cm/s for biogas with 15% H_2. The authors concluded that a significant improvement on the laminar burning velocity of biogas requires a hydrogen addition near 15% or more in volume.

Suhaimi et al. [44] reported the laminar burning velocities of hydrogen enriched biogas-air mixtures (biogas composition 50% CH_4–50% CO_2) with an equivalence ratio from 0.4 to 0.9 using a constant volume combustion chamber, at initial pressure of 1 bar and temperature range of 25 °C to 29 °C. They found that the H_2 enrichment increases the biogas burning rate to a significant extent, especially at 30–40% enrichment, while for smaller enrichment (i.e., 10–20%) an insignificant increase in the burning rate is observed. Suhaimi et al. [44] pointed out that the maximum value of the laminar burning velocity occurs at equivalence ratio of 0.8 for all mixtures regardless of the hydrogen enrichment percentage. At this equivalence ratio the reported laminar burning velocity increase from 22.0 cm/s at 10% H_2 addition, to 48.0 cm/s at 40% H_2 addition.

The laminar burning velocities of biogas-H_2 mixtures (5% or 15% H_2 by volume) were measured by using the method of the inner cone of combustion zone, under normal conditions and various aeration factors (1.0–1.4) [45]. Additionally, computational results on a biogas mixture with 60% CH_4 and 40% CO_2 obtained using CHEMKIN software and two kinetic mechanisms (GRI-Mech 2.11 and GRI-Mech 3.0) were reported. They found that the maximum value of the laminar burning velocity for biogas-H_2 mixtures is reached in the stoichiometric flames, with experimental values ranging from 27.7 cm/s for pure biogas and 29.9 cm/s for biogas with 5% H_2. The authors noted that for biogas mixture with 15% H_2, it was not possible to obtain the deflagration rate in stoichiometric flame due to the flame instability. For aeration factors of 1.1, the values obtained were 26.1 cm/s, 26.7 cm/s and 30.3 cm/s for pure biogas, a mixture with 5% H_2 and a mixture with 15% H_2, respectively. For mixtures with 15% H_2, the effect of hydrogen in the increase of the biogas flame speed is appreciated. The computed results have values that are close to the experimental ones.

An experimental investigation of the adiabatic laminar burning velocity of hydrogen-enriched biogas mixtures was conducted using a flat flame burner, at 1 bar and 298 K [24]. The biogas compositions used in this paper are 50% CH_4–50% CO_2, 60% CH_4–40% CO_2, 70% CH_4–30% CO_2 and 80% CH_4–20% CO_2, respectively. The amount of hydrogen was 20 or 40%. Additionally, computations using CHEMKIN program with GRI-Mech. 3.0 and San Diego reaction mechanisms were used to obtain the laminar burning velocity of H_2-enriched biogas mixtures with different equivalence ratios (range of 0.7 to 1.4). The same substantial increase in the laminar burning velocity was observed with an increase in H_2 content in the fuel, at all equivalence ratios and all studied biogas compositions. For example, for the mixtures with 60% CH_4–40% CO_2 and stoichiometric ratio, the experimental laminar burning velocity increases from 19.5 cm/s for 0% H_2, to 26.5 cm/s for 20% H_2 and to 40.0 cm/s for 40% H_2. The predictions of GRI Mech. 3.0 were in better agreement with the experimental values compared to other mechanism used in this work.

Wei et al. [46] conducted a study on the laminar burning velocities of biogas-hydrogen-air mixture at different fuel compositions (60% CH_4–40% CO_2, 50% CH_4–50% CO_2, 40% CH_4–60% CO_2) and equivalence ratios (0.9–1.3) using the closed vessel method. During the experiments the initial pressure and temperature were set at 1 bar and 298 K, respectively. The amount of hydrogen was varied between 0 and 50%. In addition, simulations of the laminar burning velocity were conducted using the CHEMKIN code and GRI Mech. 3.0 and San Diego reaction mechanisms. On the whole, the GRI 3.0 mechanism had a better accuracy on predicting the laminar burning velocity of biogas-hydrogen mixture. It is obvious that the laminar burning velocity is enhanced with the increase in hydrogen fraction and the methane proportion. For the most studied mixture in literature with stoichiometric equivalence ratio (60% CH_4–40% CO_2), the experimental results of the laminar burning velocity increase from 22.5 cm/s for 0% H_2, to 38.8 cm/s for 50% H_2.

The biogas mixtures (60% CH_4–40% CO_2) enriched with H_2 (0 to 40% by volume) were experimentally examined using the Bunsen burner method, over a wide range of equivalence ratios (0.8–1.3), at atmospheric pressure and 298 ± 1 K [48]. The experimental values were compared with computational results obtained over various equivalence ratios using the ANSYS Fluent with reduced GRI-Mech 3.0 chemical mechanism. The obtained computed values were similar to experimental ones. The laminar burning velocities of biogas-air flames increase with the H_2 enrichment of the mixture. For lean mixtures (equivalence ratio of 0.8), the experimental laminar burning velocities increase from 16.3 cm/s for 0% H_2, to 30.0 cm/s for 40% H_2. For stoichiometric mixtures, the experimental laminar burning velocities increase from 22.5 cm/s for 0% H_2 to 43.0 cm/s for 40% H_2. For rich mixtures (equivalence ratio of 1.2), the experimental laminar burning velocities increase from 21.0 cm/s for 10% H_2, to 39.5 cm/s for 40% H_2. The authors concluded that improvements in burning velocities are more pronounced in the stoichiometric and rich fuels compared with lean mixtures.

A recent paper [49] numerically investigated the laminar burning velocity of three biogas blends (100% CH_4, 90% CH_4–10% CO_2, 80% CH_4–20% CO_2) with hydrogen addition up to 50% in volume by Cantera package with USC-Mech 2.0 mechanism. The runs were performed at equivalence ratios between 0.8 and 1.0, initial pressure of 1 bar and initial temperature of 298 K. Naturally, in all three cases, an increase in hydrogen content produced an increase in laminar burning velocity. For the stoichiometric mixture containing 80% CH_4, the laminar burning velocity increase from 31.0 cm/s for 0% H_2, to 52.5 cm/s for 50% H_2.

Figure 12 compares results of the laminar burning velocities for biogas mixtures enriched with hydrogen from experiments at standard conditions and various equivalence ratios. The biogas referred to in Figure 12 consists of 60% CH_4 and 40% CO_2 and the hydrogen concentration is 20 and 40 vol%, respectively. The results of Yadav et al. [24], Wei et al. [46] and Nurmukan et al. [48] are in closer agreement for mixtures without hydrogen and with 20% H_2. For H_2-enriched mixtures with higher hydrogen content (e.g., 40% H_2) the values reported by Yadav et al. [24] and Nurmukan et al. [48] are in better agreement than the data from Wei et al. [46].

Another representative set of data is given in Figure 13 were data referring to a biogas with the composition of 50% CH_4 and 50% CO_2 enriched with hydrogen (20 and 40 vol%) from experiments and modelling, at different equivalence ratio and ambient initial conditions are plotted.

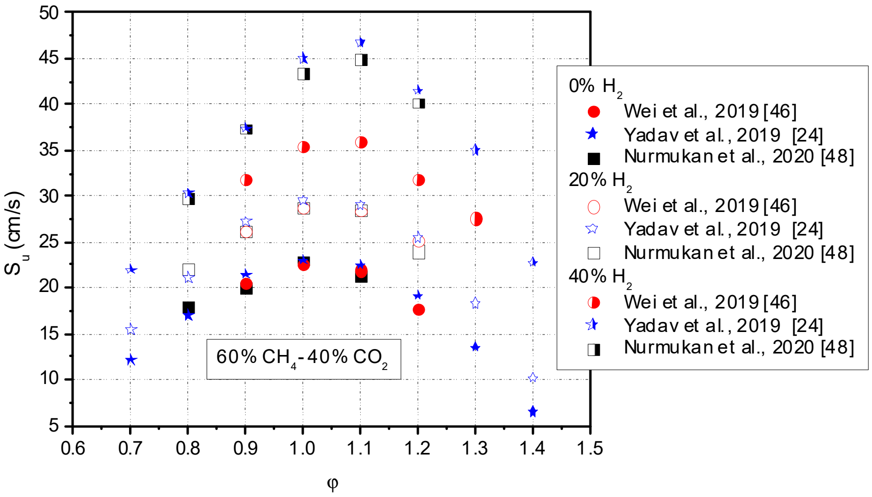


Figure 12. Variation of the experimental laminar burning velocities of H_2-enriched biogas mixtures at various equivalence ratios and standard initial conditions; Biogas composition: 60% CH_4–40% CO_2.

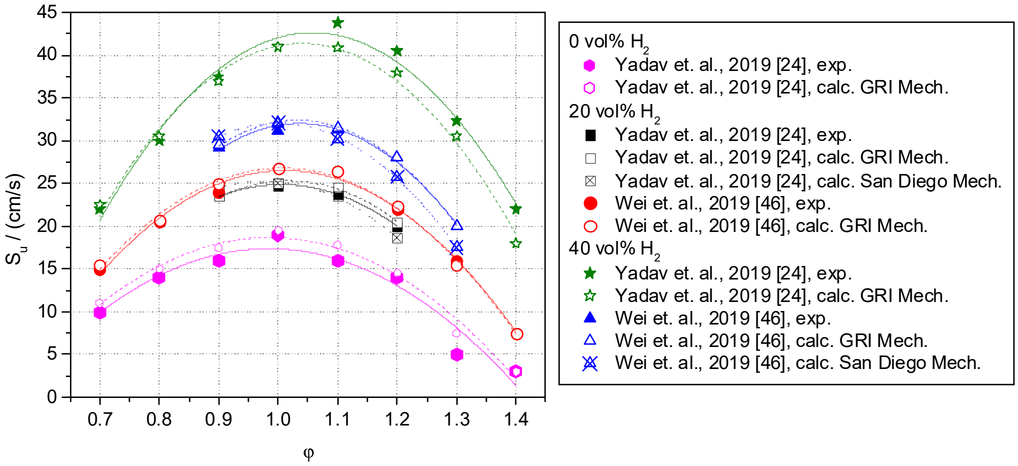


Figure 13. Variation of the laminar burning velocities of H_2-enriched biogas mixtures at various equivalence ratios and standard initial conditions; Biogas composition: 50% CH_4–50% CO_2.

From Figures 12 and 13 is observed that the laminar burning velocities increase with an increase in hydrogen content. The effect of hydrogen on the biogas blends is more visible from data presented in Figure 14 where a comparison of some of the experimental and numerical results obtained in the above mentioned studies regarding the one of the most common studied compositions of biogas (60% CH_4–40% CO_2) is given. The results refer to the stoichiometric mixture, at standard conditions and various hydrogen contents. The increase in the laminar burning velocity is monotonous and is similar to the case of methane-air flames enriched with fractions of hydrogen. However, the laminar burning velocity of hydrogen-enriched biogas mixtures is higher than laminar burning velocity of methane. This is due to the fact that the laminar burning velocity of hydrogen is higher than that of methane [113,114]. The increase of the laminar burning velocity with increase of H_2 content was also observed by Xie et al. [115] who reported data on $CO/H_2/air/H_2O$ mixtures with hydrogen content between 5% and 50%. However, the authors observed that the laminar burning velocities of $CO/H_2/air/H_2O$ mixtures decrease with the increase of water content, at all studied equivalence ratios.

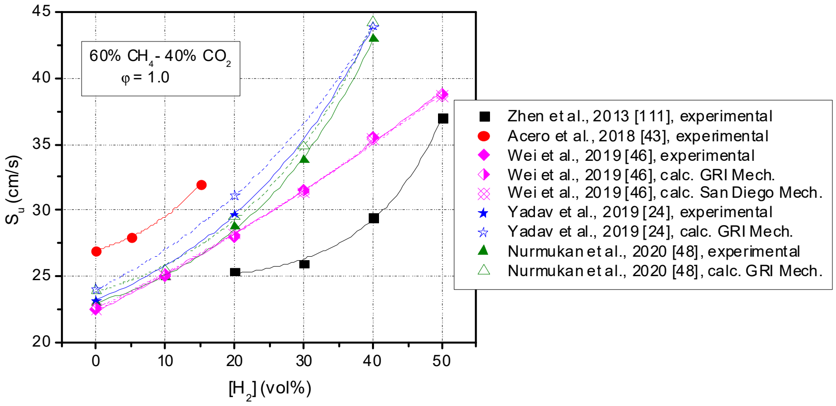


Figure 14. Comparison of the experimental and numerical results referring to the stoichiometric mixture, at standard initial conditions and various hydrogen content.

Even if the carbon dioxide concentration is high, the presence of hydrogen causes an increase in the laminar burning velocity by mitigating the thermal diffusivity and concentration effects of CO_2 on the laminar burning velocity. This can be seen in Figure 15 where data regarding the biogas mixtures enriched with 40 vol% H_2, at equivalence ratio of 1.2, different content of carbon dioxide and standard conditions are plotted. Figure 15 shows that literature data are scattered even if both concentrations of hydrogen and carbon dioxide are maintained constant (each of them 40 vol%). This is probably due to different experimental techniques: flat flame burner in the case of Yadav et al. [24]; closed vessel in the case of Wei et al. [46]; Bunsen burner in the case of Nurmukan et al. [48]. Different kinetic mechanisms also led to different results: GRI Mech. 3.0 used by Yadav et al. [24] and Wei et al. [46]; San Diego used by Wei et al. [46]; reduced GRI used by Nurmukan et al. [48].

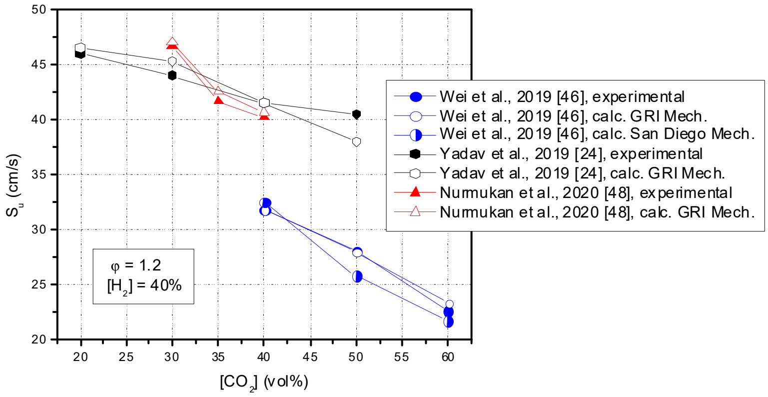


Figure 15. Influence of CO_2 concentration on the laminar burning velocities of biogas mixtures with equivalence ratio of 1.2 enriched with 40 vol% H_2, at standard initial conditions.

Despite the fact that different values of laminar burning velocities were obtained from measurements or modelling, for practical applications, large quantities of H_2 added to biogas may avoid the need to completely remove of the carbon dioxide from the biogas blends. Removal of CO_2 is necessary in order to increase the laminar burning velocity and the calorific value of the biogas. CO_2 removal methodologies use separation by membranes, adsorption or absorption. This process involves laborious, costly, and time-consuming work, therefore CO_2 removal can be replaced by the addition of hydrogen especially in some internal combustion engines which use biogas as fuel. Studies on combustion of hydrogen enriched methane and biogas were conducted in internal combustion engines [51,116,117]. The studies pointed out that, at constant initial temperature, hydrogen promotes ignition, which then occurs earlier, as a consequence of higher chemical reactivity. At constant initial pressure, hydrogen presence shifts the operating range towards lower initial gas temperature and lower equivalence ratio and reduces NO_x emissions. The addition of hydrogen to biogas also improves combustion stability, reducing cycle-by-cycle variations. Additionally, hydrogen improves the thermal efficiency and increases the heat release rate.

The laminar burning velocity depends on the reaction rate, the thermal diffusivity and flame temperature [67]. Thus, adding hydrogen to the biogas mixture impacts the laminar burning velocity by changing the mixture properties such as the thermal diffusivity and the specific heat capacity of the mixture. Hydrogen enrichment increases the thermal diffusivity and the reaction rate, as well as the flame temperature, therefore leads to laminar burning velocity increase. Rocha et al. [118] studied the relative impact of temperature, species concentration and chemical/kinetics effects on chemiluminescence due to hydrogen enrichment in biogas-air flames using both, numerical and experiments. The authors evaluated this impact and concluded that the chemistry effect is the dominant factor in chemiluminescence. Alternatively, the kinetic or chemical effects occur because of the participation of those species containing hydrogen to the chemical reactions or pathways, while the concentration effects result from a reduced amount of a certain species, caused by an increase of another [49]. On the other hand, Fu et al. [119] observed that the flame structure of syngas mixtures with various equivalence ratio (0.5–1.8) is affected by the increase of H_2 content. The authors mentioned that the flame structure is affected by both curvature effect and preferential diffusion of the fuel.

6.3. Effect of Oxygen Addition on the Laminar Burning Velocity of Biogas-Air Flames

Another way to improve the combustion properties of biogas is the oxygen addition to the mixture. This increases the reactivity of the mixture and therefore the laminar burning velocity. Nowadays, the oxy-fuel combustion is also considered as a promising approach for CO_2 emission, thus reducing greenhouse gas emissions. This procedure consists in replacement of air in traditional combustion with pure O_2 mixed with recycled flue gas (mainly containing CO_2 and H_2O) for material safety and temperature control. The use of pure oxygen solves the problem of the NO_x emissions during the combustion process. NO_x emission can be reduced by removal of nitrogen from flue gases or removal of nitrogen from air before combustion. In oxy-fuel combustion, the nitrogen from air is missing, pure oxygen being used as oxidizer and therefore there are less NO_x emissions from biogas combustion [120]. Additionally, the carbon dioxide concentration in the exhaust gas is high, and thus, carbon capture and storage technology are needed. As Xie et al. [81] wrote, the carbon capture and storage involves a series of processes for safe storage of CO_2 emissions from fossil fuel combustion into geological storage, rather than to be released into the atmosphere. Therefore, the oxy-fuel combustion is one of the most promising technologies for carbon capture and storage. Along with carbon capture and storage technology, biogas oxy-combustion can achieve zero emissions, including CO_2 [81]. Thus, some researchers conducted studies on oxy-biogas flames for a better understanding of its behaviour, in order to apply it to industrial facilities. The experimental and numerical methods for obtaining the laminar burning velocity of biogas-O_2-air mixtures described in the present study are summarised in the Table 6.

For example, Cardona and Amell [56] studied numerically and experimentally the laminar burning velocity of biogas oxygen-enriched air mixtures. The experiments were made using a contoured slot burner at different air/fuel ratios, initial pressure of 0.828 bar and initial temperature of 295 K. The biogas composition was 66% CH_4–34% CO_2 and oxygen concentration was between 21 and 32 vol%. The kinetic modelling was made using CHEMKIN-PRO package together with GRI-Mech 3.0 mechanism. The authors concluded that the addition of oxygen to the air results in more stable flames and a significant increase in the burning velocity. The experimental laminar burning velocity for stoichiometric mixtures with 21 vol% O_2 is 28.5 cm/s; for mixtures with 24 vol% O_2 is 42.5 cm/s, while for mixtures with 32 vol% O_2 is 58.5 cm/s.

Table 6. Summary of experimental and numerical methods for obtaining the laminar burning velocity of biogas-O_2-air mixtures described in the present study.

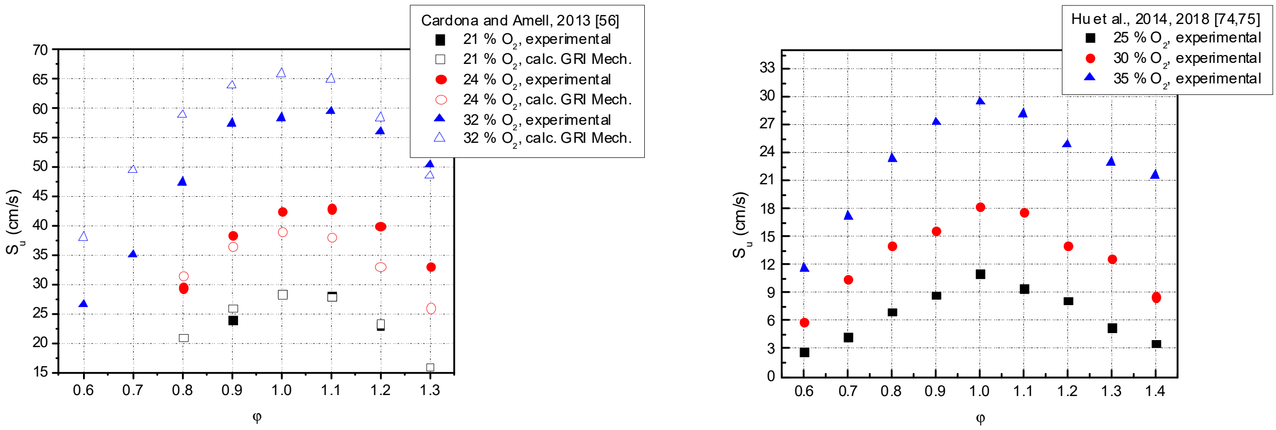
Type of Method	Method/Model/Mechanism Name		Reference
Experimental	Bunsen burner		Hu et al. [74]
	Contoured slot burner		Cardona and Amell [56]
	Flat flame burner		Wang et al. [59]
	Closed vessel		Xie et al. [81]
Numerical	CHEMKIN software	GRI-Mech. 3.0	Cardona and Amell [56] Wang et al. [59] Hu et al. [74] Xie et al. [81]
		HP-Mech mechanism	Wang et al. [59]

Xie et al. [81] reported the laminar burning velocity of $CH_4/CO_2/O_2$ mixtures from experiments and modelling at various CO_2 mole fractions (0.4–0.7), equivalence ratios (0.4–1.6), initial pressures (1–3 bar) and 300 K. The closed vessel method and CHEMKIN package (mechanism GRI-Mech 3.0) were used. The experimental results for the stoichiometric mixture and standard conditions range between 15.0 cm/s for CO_2 fraction of 0.7, to 122.0 cm/s for CO_2 fraction of 0.4. Generally, the calculated data show a reasonable agreement with the experimental ones, especially at atmospheric pressure. However, there exists a large discrepancy when the pressure is increased. It was found that the GRI 3.0 mechanism under predicts the laminar burning velocities of CH_4 oxy-fuel combustion. The discrepancy becomes larger with the decrease of the CO_2 fraction.

Another study was conducted by Hu et al. [74] on $CH_4/O_2/CO_2$ mixtures in atmospheric conditions (300 K and 1 bar) using a Bunsen burner. This study was performed to obtain the laminar burning velocities for a range of equivalence ratios (from 0.6 to 1.4) and oxygen concentrations (from 25% to 35%). The experimental data were compared to numerical predictions obtained with GRI-Mech 3.0 mechanism using a CHEMKIN package. With an increase in O_2 concentration, the flame speeds increased. The results showed that the maximum of the experimental laminar burning velocities appeared at the stoichiometric composition. In this case the laminar burning velocity values are as follows: 11.1 cm/s, 15.7 cm/s, 20.0 cm/s, 26.1 cm/s, 29.6 cm/s, corresponding to O_2 concentration 25%, 29%, 31%, 33%, 35%. Discrepancies between experimental and numerical data were reported, but the trend was well predicted. Later Hu and Yu [75] extended their study to elevated initial temperatures (300 K, 400 K and 543 K). The authors found that the adiabatic flame temperature increase with initial temperature increase and hence the laminar burning velocity increases. For example, the laminar burning velocities of stoichiometric mixture enriched with 30% O_2 increase from 18.2 cm/s at 300 K, to 49.5 cm/s at 543 K.

Laminar burning velocities of $CH_4/O_2/CO_2$ flames were measured at elevated pressures up to 0.5 MPa and equivalence ratios ranging from 0.6 to 1.6 using the heat flux method [59]. The oxygen molar fraction was varied from 0.31 to 0.42. The laminar burning velocities of stoichiometric mixture obtained from measurements are 19.8 cm/s, 37.0 cm/s, 48.8 cm/s, corresponding to 31 vol%, 38 vol% and 42 vol% oxygen. Kinetic modelling was also performed using the GRI-Mech 3.0 and the HP-Mech mechanisms. Data present larger discrepancies between the measured and predicted laminar burning velocities, but overall, both mechanisms predict reasonably well the laminar burning velocities.

Data from the above studies regarding the effect of oxygen enrichment on the laminar burning velocities are given in Figures 16 and 17, at various equivalence ratios and initial temperature around 300 K. The results represented in these figures are both from experiments and modelling. As Cardona and Amell [56] noted, moderate addition of oxygen has great impact on the laminar burning velocity of pure biogas.



(**a**) (**b**)

Figure 16. Laminar burning velocities of biogas mixtures enriched with oxygen. Data at: (**a**) 0.828 bar and 295 K; (**b**) 1 bar and 300 K.

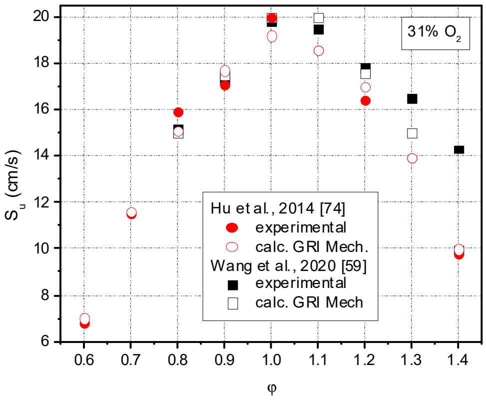


Figure 17. Laminar burning velocities of biogas mixtures enriched with oxygen at standard conditions.

The influence of oxygen addition is better seen from Figure 18 where data from experiments on lean and stoichiometric mixtures, at ambient initial conditions are plotted. Hu et al. [74] observed that as the oxygen concentration increases, the laminar burning velocity increase in a nonlinear relation. Experimental and modelling results indicated, through data fitting, a nearly quadratic function relationship between flame speeds and O_2 concentration [74]. The authors mention that this trend is due to two reasons. First, carbon dioxide has a higher specific heat than oxygen. Thus, an increase in O_2 concentration means a decrease in CO_2 concentration, which raises the flame temperature to generate more radicals in chain reactions. Second, the reduction of carbon dioxide brings more chance for particle collisions, such as the [O], [OH], [H] to cause more new chain reactions, which speed up the combustion reaction. Both reasons lead to the laminar burning velocity increase.

The increase in the amount of oxygen in the mixture results in increase of adiabatic flame temperature as it results from Figure 19, where data on stoichiometric mixture from Cardona and Amell [56], Wang et al. [59] and Hu and Yu [75] are given. The increase in the adiabatic flame temperature leads to increase in the laminar burning velocity due to the increase in mixtures' reactivity. As it results from Figures 19 and 20, the adiabatic flame temperature increases with the initial temperature increase and hence the laminar burning velocity increases. This behaviour can be also seen from Figure 21 where data collected from Hu and Yu [75] at various initial temperatures, equivalence ratios and ambient pressure are plotted. As Cardona and Amell mentioned earlier [56], these increases are partly due to the fact that the additional oxygen leads to a reduction of inert concentration in the unburned mixture and its consequent ability to absorb heat during the reaction, resulting in a higher flame temperature. In practical terms, the use of additional oxygen in combustion results in increased productivity in the industrial heating processes and improved flame stability.

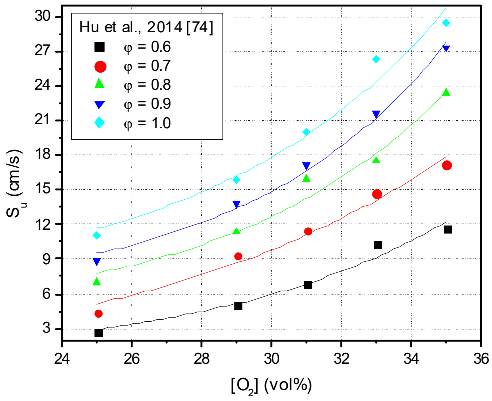


Figure 18. Influence of O_2 concentration on the experimental laminar burning velocities of biogas mixtures.

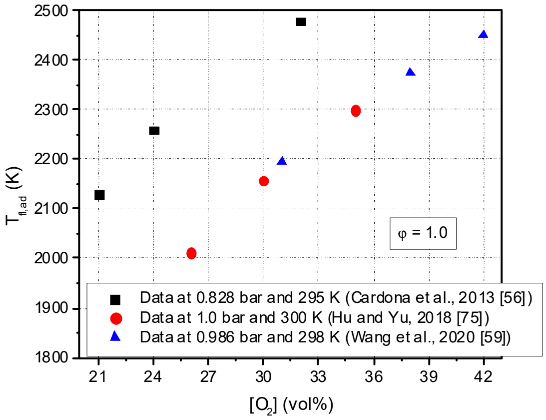


Figure 19. The adiabatic flame temperature variation with O_2 content for stoichiometric biogas-air mixtures.

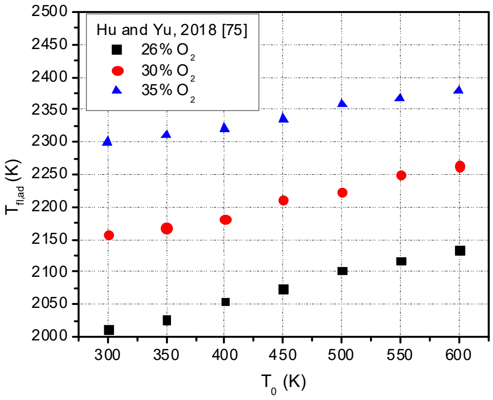


Figure 20. The adiabatic flame temperature variation with initial temperature for oxygen-enriched biogas mixtures.

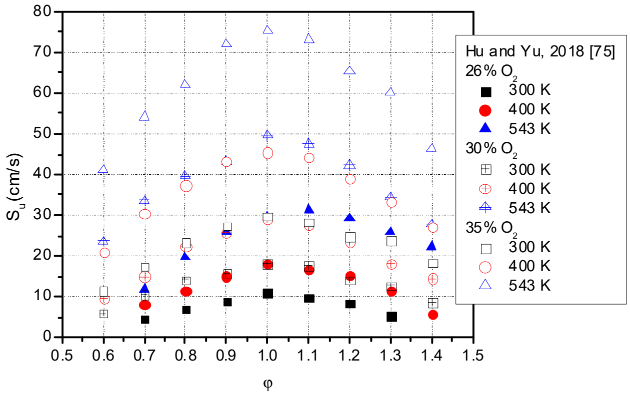


Figure 21. Influence of initial temperature on experimental laminar burning velocity of oxygen-enriched biogas mixtures.

It was found that there are three ways for the initial temperature affecting the laminar burning velocity [67]. The first factor is through the reaction rate and the adiabatic flame temperature. The increase in the adiabatic flame temperatures accelerates the reaction rates. The second factor is through the transport properties of the mixtures. The increase in the adiabatic flame temperature enhances the transport effect of CO_2 due to the parameters of the transport property, such as the heat conductivity, which depend on the temperature. The third factor is due to the change in density. An increase in the initial temperature increases the laminar burning velocity through the decrease in density.

Although an increase in the amount of oxygen increases laminar burning velocity, an increase in the initial pressure decreases the laminar burning velocity. This behaviour is observed from Figure 22 where experimental data on stoichiometric biogas-air-oxygen mixture, at various oxygen contents and 298 K are presented [59].

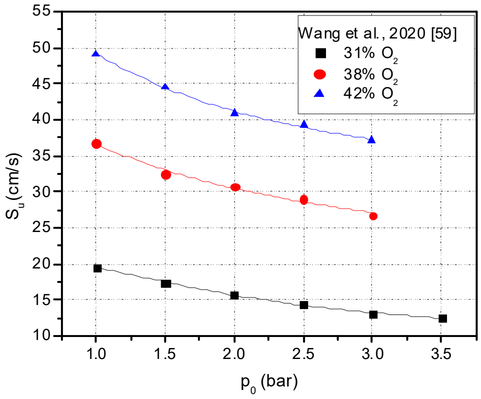


Figure 22. Initial pressure influence on experimental laminar burning velocity for stoichiometric biogas-air-oxygen mixtures. Data refers to various oxygen contents and 298 K initial temperature.

A small increase in the oxygen content significantly enhances the laminar burning velocities for atmospheric and elevated pressures, as well as for ambient and elevated temperatures. Therefore, the combustion properties and flame stability are significantly improved when air is enriched with pure oxygen.

7. Conclusions

The available literature regarding the laminar burning velocities of biogas-air mixtures has been reviewed. The results are summarized as follows:

- The increase in carbon dioxide content decreases laminar burning velocities no matter the initial biogas composition, pressure and temperature. This phenomenon significantly affects the stability of the flame, which can make it difficult to use biogas as a fuel in spark ignited engines and conventional burners.

- Addition of hydrogen to a biogas-air mixture increases the laminar burning velocity due to the increase in thermal diffusivity and the reaction rate, as well as the flame temperature of the mixture. For practical applications, large quantities of H_2 added to biogas could avoid the need to completely remove of the carbon dioxide from the biogas blends.
- The addition of oxygen to biogas mixtures leads to increase of the laminar burning velocity due to the increase in the adiabatic flame temperature. Oxygen-enriched combustion of biogas can be efficient for power generation not only because of the lower NO_x emissions but also because of the suitability for CO_2 capture and storage.
- Small quantities of hydrogen and oxygen added to biogas leads to greater flame stability and improve the combustion properties of these flames.
- The increase of the initial temperature and the decrease of the initial pressure of fuel-oxidant mixture cause the increase of both, experimental and computed laminar burning velocities.

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