

Article

Investigation of Waste Biogas Flame Stability Under Oxygen or Hydrogen-Enriched Conditions

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Abstract: Increasing production rates of the biomethane lead to increased generation of waste biogases. These gases should be utilized on-site to avoid pollutant emissions to the atmosphere. This study presents a flexible swirl burner (~100 kW) with an adiabatic chamber capable of burning unstable composition waste biogases. The main combustion parameters and chemiluminescence emission spectrums were examined by burning waste biogases containing from 5 to 30 vol% of CH₄ in CO₂ under air, O₂-enriched atmosphere, or with the addition of hydrogen. The tested burner ensured stable combustion of waste biogases with CH₄ content not less than 20 vol%. The addition of up to 5 vol% of H₂ expanded flammability limits, and stable combustion of the mixtures with CH₄ content of 15 vol% was achieved. The burner flexibility to work under O₂-enriched air conditions showed more promising results, and the flammability limit was expanded up to 5 vol% of CH₄ in CO₂. However, the combustion under O₂-enriched conditions led to increased NO_x emissions (up to 1100 ppm). Besides, based on chemiluminescence emission spectrums, a linear correlation between the spectral intensity ratio of OH* and CH* (I_{OH^*}/I_{CH^*}) and CH₄ content in CO₂ was presented, which predicts blow-off limits burning waste biogases under different H₂ or O₂ enrichments.

Keywords: swirl burner; waste biogas; hydrogen; oxygen; combustion; flame stability; blow-off limit

1. Introduction

Due to concerns regarding global climate change, there has been a search for new ways to improve energy efficiency, reduce pollutant emission, and ensure sustainable management of resources. For these reasons, various alternative fuels and by-products (gas or waste fuel) with low calorific value are gaining more attention as an alternative energy source for energy production [1–3]. Basically, such fuels are gained in different fields with different compositions like steel production, refineries, landfills, biogas plants, gasification plants, and other industry sources. For example, a blast furnace gas or syngas consists of different concentrations of CO, H₂, CO₂, and/or N₂, while biogas is mainly composed of CO₂ and CH₄. Considering that the biogas and biomethane production is increasing over the years, rates of the waste biogas from biomethane production are also growing. These gases mainly consist of from few to 25 vol% of methane and should be utilized on-site to prevent pollutant emissions to the atmosphere. However, these gases are not well suitable to combust in existing natural gas-fired systems as the low concentrations of CH₄, and the high amount of CO₂ could result in an unstable flame, blow-off, and a release of unburned gases into the environment. Mainly it caused by the high amount of CO₂, which affects the flame temperature, thereby reducing the stability of the flame.

To avoid these problems and ensure complete and clean combustion of waste biogases, flame stability must be improved, and flammability limits expanded. It could be achieved using new concepts/modifications of burners [4–7]. For example, a new burner design based on modified geometry was proposed and tested for low calorific gas combustion by [8]. It was found that this burner

ensures stable combustion of natural gas and syngas blends, but CO emissions exceed 200 mg/m^3 if thermal shares of syngas are below 70%. Another concept burner based on continuous air staging for low calorific gases was developed by a research group from Gaswärme-Institute. V. Essen (GWI) [9]. The burner (up to 200 kW) was tested on syngas, landfill gas, and mine gas. The obtained results showed that it is possible to achieve a stable combustion process with low NO_x and CO emissions, but there is a need to preheat these gases up to 400 °C before supplying to the combustion zone. A similar study was performed by Mortberg et al. [10]. The authors preheated air up to 900 °C to enhance the low calorific value (LCV) gas combustion stability. During combustion tests, a cross-flow jet of LCV gas mixture (11.9 vol% CH₄ in N₂) was introduced in the combustion airflow at oxygen-deficient conditions. The obtained results showed that this configuration using preheated combustion air is suitable to combust LCV gases, but results in the prolonged ignition delay, higher turbulence levels, and higher vorticity. Another work [11] showed that surface-stabilized combustion (SSC) technology burner is able to burn the LCV biogas with 20–65 vol% CO₂ in CH₄ or hydrogen-enriched CH₄ (15–100 vol% H₂ in CH₄) with low pollutant emissions. However, the burner was more suitable for combustion of high calorific value gases. In the case of biogas with 65 vol% CO₂ in CH₄, the flames were lifted, changing φ from 0.98 to 0.8, while further increase in φ led to blow-off. Another research [12] also focused on the combustion of LCV gases in burners with SiC and Al₂O₃ porous structures. The authors were capable of combusting landfill gas with a methane content of 26 vol%. Though, at higher power (10 kW), the flammability limit decreased by 4 vol% of CH₄ in CO₂ due to increased flow speed. The authors also noticed that preheating the gas mixture, and the flammability limit could be extended by 2 vol%. Al-Attab et al. [13] investigated the combustion of producer gas from biomass gasification in a two-layer porous burner. Results showed that the burner is able to combust the producer gas with a lower calorific value of 5 MJ/m^3 changing equivalence ratios φ in the range of 0.33–0.71, but NO_x emissions were in the range from 230 to 270 ppm. In order to increase the blow-off limit of LCV gases, Song et al. [14] investigated an improved preheating method of the gas mixture, based on an annular heat recirculation. The gas flow was preheated by a high-temperature wall near the gas inlet, which temperature increase, due to improved axial heat conduction and radiation heat transfer of porous media. It ensured stable combustion of the LCV gas of 1.4 MJ/m^3 . However, according to [15], higher volumetric capacity is needed to achieve higher power comparing to existing conventional burning systems. Besides, the use of porous media burners for the combustion of waste biogases with the varying composition is a bit complicated as a material for the porous media should be designed considering gases needed to burn; otherwise, a periodic replacement of the porous media could be needed.

Another option to improve flame stability and flammability is to use an addition of hydrogen/syngas or supply of oxygen-enriched air [16–22]. Chiu et al. [23] investigated the effect of H₂/CO on a premixed methane flame. The research was performed in an impinging burner by changing an H₂/CO concentration from 20/80 to 80/20 vol% and a methane concentration from 10 to 20 vol%. It was determined that the stable flame with H₂/CO of 20/80 vol% is achieved at reach combustion conditions ($\varphi \geq 1.8$) though the flame stability and flammability increased with increasing the hydrogen concentration in simulated syngas. It was also noted that with the increasing amount of H₂ in CO increases the flame temperature. A similar work focused on the hydrogen effect on the LCV gas combustion was performed by [24]. The authors performed numerical and experimental burner tests at a thermal power of 10 kW using syngas ($\sim 4.7 \text{ MJ/m}^3$) and blast-furnace gas ($\sim 3.7 \text{ MJ/m}^3$), including the addition of H₂ (26 and 52 vol%). It was determined that the burner properly burns the syngas, but the blast-furnace gas requires an additional supply of H₂ to avoid blow-off. Though, it was observed that the addition of H₂ negatively affects NO_x emissions. To extend LCV gas combustion flammability and improve flame stability, this burner was also tested under oxy-fuel combustion conditions [25]. Results showed that NO_x emissions decrease; however, to achieve a more stable flame and avoid overheating of the burner wall, a modification for the oxidizer distribution is needed. More detailed investigation of the oxygen enrichment was performed by Ylmaz et al. [26]. The authors investigated

the biogas flame stability and formation of emissions under different oxygen enrichment conditions in a pilot-scale model burner. It was pointed out that an increase of O₂ to 24% in the oxidizer leads to improved flame stability though CO emissions were equal to 372 ppm. A higher enrichment level (28 vol% of O₂) caused decreased flame stability, but lower CO emissions, till 10 ppm. During the tests, NO_x emissions increased with increased O₂ concentrations in the oxidizer. Ba et al. [20] tried to improve flame stability by coupling oxy-fuel combustion and fuel/air preheating. Firstly, this configuration was experimentally tested using a lab-scale tri-coaxial burner (25 kW), and later, the burner scaled-up to 180 kW was tested on a semi-industrial facility. In both cases, a stable flame of blast furnace gas was achieved with very low levels of CO and NO_x emissions. However, this configuration was not tested on waste biogases, and the effect on flame stability is unknown.

Literature review reveals that there are many works proposing techniques to ensure stable combustion of syngas, furnace blast gases, natural gas blends with LCV gases and etc., but studies related to the combustion characteristics of waste biogas are still lacking. Taking into account that biogas production rates are increasing and to prevent waste biogas emissions to the atmosphere, these gases should be utilized safely on-site. The previous work [27] showed that the addition of up to 20 vol% hydrogen or oxygen-enriched air ensures stable combustion of waste biogases (CH₄ > 30% in CO₂) with low emissions in a flat flame burner. Based on these findings, a flexible swirl burner (~100 kW) with an adiabatic chamber capable of burning unstable composition waste biogases was designed and developed. The developed combustion system provides a solution to recover primary energy and reduce pollutant emissions from biomethane production obtained waste biogases. This study presents the flexible combustion system in detail and performed tests in real conditions. During the experimental tests, flame stability and combustion limits were explored to ensure a wide range operation of the burner with different waste biogas containing from 5 to 30 vol% of CH₄ in CO₂ under air, oxygen-enriched atmosphere, or with hydrogen addition. For a better understanding of waste gas flame behavior under different conditions and to identify a burner performance via flame transition modes and blow-off limits, chemiluminescence emission spectrums (OH* and CH*) from flames of different waste biogas mixtures were obtained. During all tests, an online flue gas analysis was also performed to determine variations of NO_x and CO emissions and compliance with emission standards.

2. Materials and Methods

2.1. Experimental Setup

The experiments were carried out in a 200 kW_{th} semi-industrial scale test facility (Figure 1). In order to simulate waste biogases (LCV) with a low content of methane (CH₄) in carbon dioxide (CO₂) and its combustion with oxygen (O₂) or hydrogen (H₂) addition, a gas supply system was constructed (1–5). CO₂ was acquired in a liquid state (1) by a local supplier (JSC Gaschema), directed to a vaporizer (2) and supplied via the control unit to a gas-mixing system for preparation of LCV gases with pre-assigned compositions. Methane in the form of natural gas (NG) with the composition of 97.5 vol% CH₄, 1.2 vol% C₂H₆, 0.27 vol% C₃H₈ and C₄H₁₀, and 0.08 vol% CO₂ (NG composition is taken from the gas supplier's datasheet) was delivered from a distributed NG pipeline and supplied through a pressure regulating and metering station (3) to the gas-mixing system. For hydrogen or oxygen-enriched combustion, these gases were supplied from gas cylinders (4,5) to a novel swirl LCV gas burner (7). The required gas flow was ensured by a mass flow controller. The combustion air was supplied using an air blower (6).

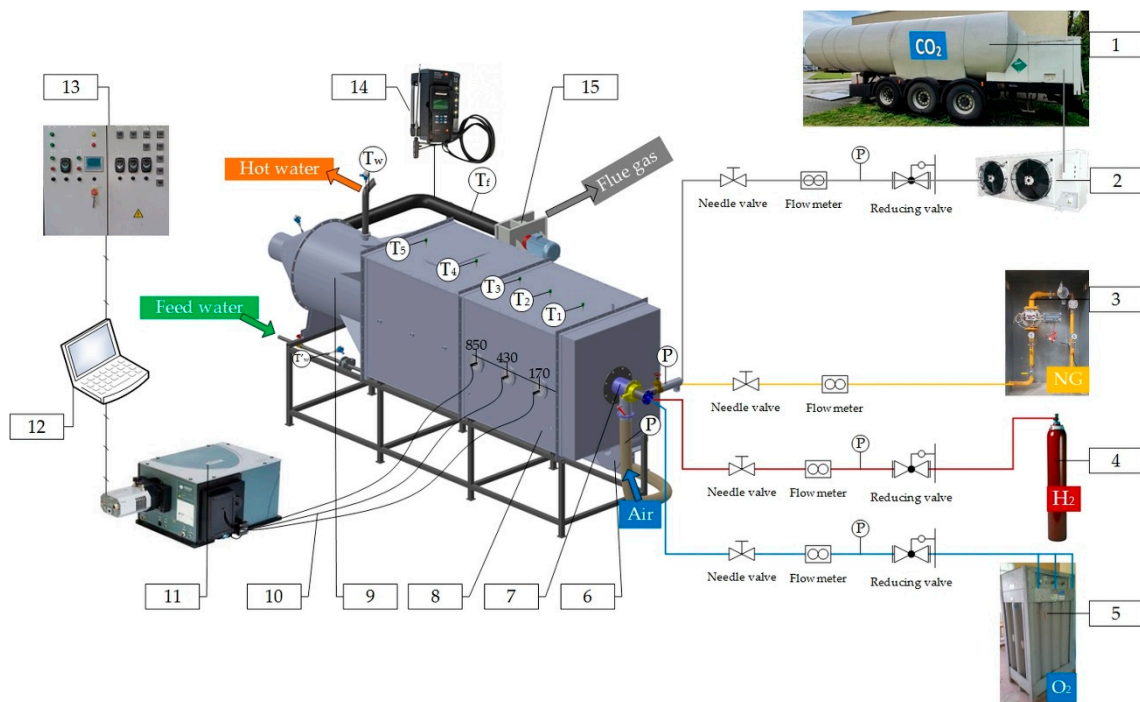


Figure 1. Scheme of the experimental burner testing setup: 1–A liquid CO₂ tank; 2–a CO₂ vaporizer; 3–a natural gas pressure regulating and metering station; 4–a hydrogen cylinder; 5–an O₂ cylinder bundle with a manifold; 6–an air blower; 7–a novel low calorific value (LCV) gas burner; 8–an adiabetic combustion chamber; 9–a boiler; 10–optical fibers with collimating lens; 11–a spectrometer with an ICCD camera; 12–a data collecting system; 13–an operation panel; 14–a flue gas analysis system; 15–a flue gas blower.

The combustion of the prepared LCV gas mixture was performed in an adiabetic chamber (8) with internal dimensions of $600 \times 600 \times 2500$ mm. The temperatures in the chamber were measured with four K type thermocouples (T_1 – T_4). The thermocouples were positioned at the 530, 900, 1690, and 2410 mm from the inner front wall, respectively. For spectral analysis of selected excited radicals from the flame, a spectrometer Andor Shamrock SR-303i coupled with an ICCD (Intensified Charge Coupled Device) camera Andor iStar DH734 (11) was used. Three optical fibers with collimating lenses (10) were installed to transmit the light to the spectrometer. The optical fibers were oriented horizontally to provide a radical distribution along with the flame and fitted at the distances of 170, 410, and 830 mm from the front wall. The combustion products are cooled down to appropriate temperature, which is measured with a K type thermocouple, by passing the boiler (9) mounted at the exit of the combustion chamber. A flow of cooling water is controlled, and water temperature is monitored before and after the boiler. The cooled flue gases are analyzed using a portable flue gas analyzer Testo 350XL (14) via a measurement point installed between the stack (15) and the boiler (9). The entire combustion process is automated and controlled from a control desk (13) installed outside the site. All operating signals from measuring equipment or sensors were collected and analyzed further.

2.2. Waste Biogas Burner

A novel swirl burner was developed for the waste biogas (LCV) combustion (Figure 2). The burner is designed to operate in a flexible mode for a variety of waste biogas compositions, including the possibility of hydrogen or oxygen addition. The burner has multiple inlets for different gases and operational regimes. The ignition of the main fuel (LCV gas) is executed with a pilot natural gas flame. After the ignition of the main fuel, a supply of pilot gas is stopped. The LCV gas is supplied via an annular channel installed in the center of the burner. Air was used as the main oxidizer, which enters

the burner through a peripheral annulus. When a calorific value of waste biogas becomes very low, a stable operation is ensured by the oxygen or hydrogen addition. The supply of these gases is designed in the center of the burner.

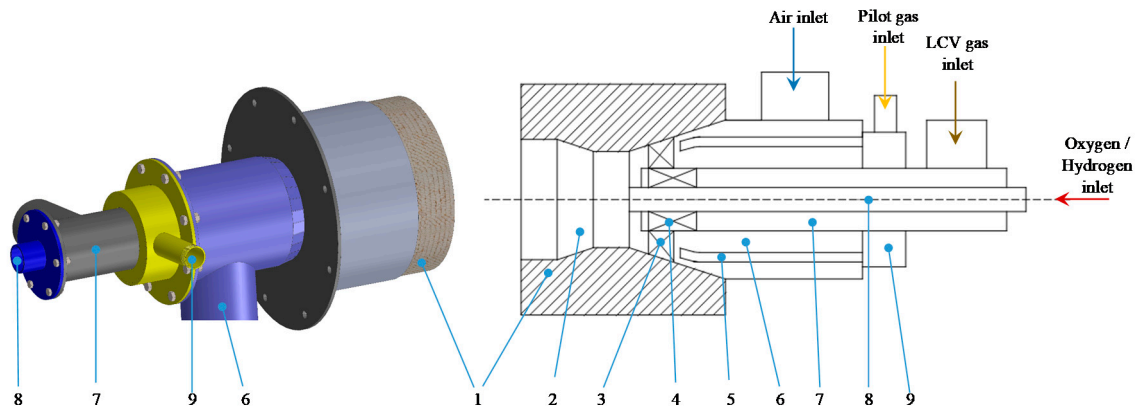


Figure 2. Schematic view of the experimental LCV gas burner: 1–A burner embrasure; 2–a burner quarl; 3–an axial airflow swirler; 4–an axial LCV gas swirler; 5–a pilot gas lance; 6–an air annulus; 7–an LCV gas annulus; 8–an oxygen or hydrogen orifice; 9–a pilot gas annulus.

The flame stabilization is achieved by swirling both, the combustion air and LCV gas flow with an axial vane swirl angle of 30° . Both swirlers has a swirl number of $S_w = 0.5$, which was defined by Equation (1) [28]:

$$S_w = \frac{2}{3} \tan(\alpha_{sw}) \left(\frac{1 - R^3}{1 - R^2} \right) \quad (1)$$

where α_{sw} is the axial vane swirl angle, R is the ratio of the center-body diameter and outer inlet tube diameter.

2.3. Experimental Procedure

The stability limits of waste biogas (LCV) combustion were determined by running the combustor at a fixed heat load of 80 kW_{th} . The heat load was calculated using the lower heating values (LHV) and measuring flows of combustible gases, namely, CH_4 , and H_2 when it was used. Thus, the main variables were CO_2 content in LCV gas, an amount of the oxidizer, and the fuel to air ratio (ϕ).

Three sets of experiments were performed to determine the limits of stable waste biogas (LCV) combustion. The first trial was associated with a determination of the lowest possible, stable combustion regime by decreasing LHV of LCV gas, which was changed by increasing the volumetric fraction of CO_2 . The air was used as the oxidizer. At each LCV gas composition, the burner performance was tested for different fuel to air ratios, which varied in the range from 0.7 to 1.0. The fuel to air ratio was determined by measuring the O_2 concentration in flue gases.

The second round of experiments was conducted with the H_2 addition to extend LCV gas combustion limits. In a previous work [29], the possibility to produce H_2 on-site from LCV gas was explored. The idea was based on the power of syngas (P2SG) technology. Therein to produce H_2 rich syngas, the thermal plasma was used to run a dry reforming of gases containing a low concentration of CH_4 in CO_2 . Finally, it was concluded that from an economic point of view, this technology is cost competitive when a produced H_2 content for 1 m^3 LCV gas combustion will constitute up to 5 vol%. To simulate this situation in the present work, the hydrogen addition of 2.5 and 5 vol% was investigated. As in the previous case, the air was used as the oxidizer with ϕ ranging from 0.7 to 1.0.

During the third set of experiments, the stability limits were determined using O_2 -enriched air instead of the H_2 addition. The amount of oxygen was gradually increased until the flame becomes stable to avoid possible problems described in some works [21,26] like increased CO emissions,

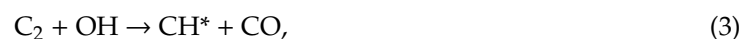
high flame temperature, or even unstable combustion. When the flame became stable, the burner performance for different LCV gases against the various fuel to air ratios from 0.7 to 1.0 was tested. The expression of the volumetric oxygen content in this work corresponds to the O₂ concentration in air, which was defined as follows:

$$\text{O}_2 \text{ in air} = \frac{V_{\text{air}}0.21 + V_{\text{O}_2}}{V_{\text{air}} + V_{\text{O}_2}} \quad (2)$$

where V_{air} and V_{O_2} are the volume flow rates of air and O₂, respectively. The upper value of O₂ in the air was pre-selected to not exceed 30 vol%.

During all experimental runs, emissions of CO and NO_x in flue gases were measured continuously. CO is considered as the main indicator for incomplete combustion, thus despite the existence of flame stability, it demonstrates the inappropriate burner operation regime. NO_x is one of the main regulated gaseous pollutants, which should be controlled. It is well known, that the main source of NO_x formation is attributed to the thermal pathway. Thus, increasing oxygen content in the oxidizer during LCV gas combustion could lead to noticeable higher NO_x emissions [3]. In order to know the level of this pollutant during combustion and compliance with emission standards (EU directive 2015/2193 [30]), it was measured and analyzed regarding the LCV combustion mode.

In addition, the chemiluminescence based optical diagnostic tool was used to identify the possible flame transition mode. The main chemiluminescent species of interest were OH* and CH* with a wavelength of 308.9 ± 2.0 nm and 431.4 ± 2.0 nm, respectively. According to [31–34], CH* formation is related to high temperature, and the main formation pathways are:



The formation and excitation of the OH* radical are also attributed to the thermal excitation, and the main formation reactions are [31–34]:



The optical data were collected from three-measurement points using a spectrometry system (Figure 1). The light coming from the flame is dispersed, and entire spectra is collected on the split area of the ICCD camera sensor. The camera exposure duration was set to 3 s, a side input slit to 100 μm, and grating to 300 L/mm. Each spectrum consisted of 10 acquisitions.

3. Results and Discussion

3.1. Stability Maps of Waste Biogas (LCV) Combustion

This section presents combustion stability maps for developed LCV gas burner under different tested combustion modes. Firstly, LCV gases with different concentrations of CH₄ and CO₂ were burned in the air over a wide range of fuel to air ratios to determine the limit of stable combustion under ordinary conditions. As can be seen from Figure 3, the stable combustion of the waste biogas (LCV) with CH₄ ≥ 25 vol% in CO₂ can be reached in the air under all tested range of φ. Compared to previous results [27], the combustion of waste biogas in a flat flame burner under the air atmosphere was only able with a mixture of 30 vol% CH₄ in CO₂. In that case, the developed burner shows wider flammability ranges under air conditions. A reduced CH₄ content to 20 vol% in the waste biogas (LCV) leads to flame stability problems, and blow-off occurs at a richer combustion mode,

$\phi \geq 0.87$. According to obtained results (Figure 3), the stable combustion of LCV gases with lower CH_4 content (>20 vol%) is only possible with H_2 or O_2 addition. The addition of H_2 to the biogas flame was studied in detail by others [16,35,36]. Zhen et al. [35] noted that the H_2 addition of 5 vol% improves biogas flame stability, and CO emissions decrease, but further addition increase to 10 vol% of H_2 is insignificant on flame stability. Similar findings were also reported by Leung and Wierzbka [17]. The authors investigated the effect of 10 vol%, 20 vol%, and 30 vol% H_2 addition on the biogas flame stability and determined that the most significant results are achieved by supplying a small amount of H_2 .

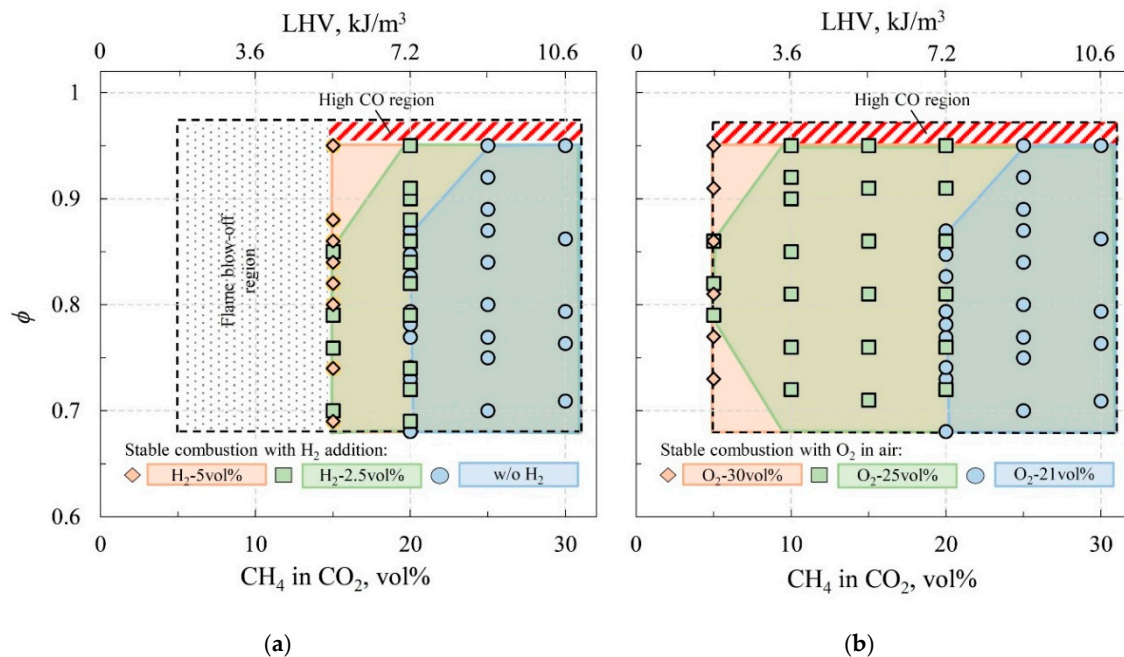


Figure 3. Stability maps of waste biogas (LCV) combustion with different content of CH_4 in CO_2 for a range of (a) H_2 addition and (b) O_2 air enrichment.

Even though the concentration of CH_4 in biogas is higher and varies from 40% to 60% compared to the studied range of CH_4 concentrations (5–30 vol%), the provided findings are in good agreement with the obtained results. The stable combustion of LCV gas containing 15 vol% of CH_4 is obtained within the range of ϕ from 0.7 to 0.85 by adding H_2 of 2.5 vol%, while an increased addition of H_2 to 5 vol% has an insignificant effect, and the flammability limit is extended only to a richer region, up to ϕ of 0.95. However, further decrease of CH_4 content in LCV gases results in flame blow-off even in both cases of H_2 addition (Figure 3a). In order to extend the flammability limit and ensure stable combustion of LCV gases with very low CH_4 content (5–15 vol%), O_2 -enriched air was introduced. Considering that a high oxygen enrichment level could lead to the unstable flame and increased CO emissions [26], two cases with 25 vol% and 30 vol% of O_2 were tested (Figure 3b). As was expected, using O_2 -enriched air with O_2 content of 25 vol%, the stable combustion of waste biogases with CH_4 content from 30 to 5 vol% was ensured. However, in this case, the stable combustion of LCV gas containing 5 vol% of CH_4 is achieved only in the narrow range of ϕ , from 0.79 to 0.86. Meanwhile, at a higher enrichment level (30 vol% of O_2), the stable combustion of LCV gases is acquired in all tested points (Figure 3b). Similar results were also acquired numerically studying methane combustion under CO_2/O_2 atmosphere, and researchers pointed out that the most effective and stable combustion is achieved at O_2 concentrations of 28–32 vol% [37]. Though another work [21] showed a bit higher level of O_2 (31–35 vol%) is needed to achieve a stable flame. Considering the obtained results, it could be assumed that the designed burner with the adiabatic chamber ensure stable combustion of waste biogases (5–30 vol% of CH_4 in CO_2) at lower concentrations of O_2 (25–30 vol%), due to a special design

of the burner allowing the faster mixing of fuel oxidizer, faster reaction kinetic and minimal heat loss via the adiabatic combustion chamber walls. However, the presented design of the LCV gas burner shows an opposite relation in non-premixed combustion. The flame extinguish is more likely at higher fuel to air ratios than at lower ones, and this was found for all studied cases (Figure 3). Typically, the flame is more stable, or a flash-back phenomenon is more likely at lower fuel to air ratios, even in premixed flames [25]. The reasons for an instability arose might be related to ongoing several complex processes: The aerodynamic of the flame shape, which allows a proper internal hot gas recirculation zone, and an amount of heat provided from both recirculating gases and re-radiation from furnace walls [37].

In order to study the impact of those two parameters, the flame core temperature and averaged axial flow speed were analyzed. Figure 4 represents data obtained by T_1 thermocouple and shows temperature ranges during the combustion of waste biogases (LCV) with different content of CH_4 in CO_2 for all three tested cases: Under air only combustion, under H_2 addition, and under O_2 -enriched air. The upper and bottom limits stand for the maximum (ϕ_{max}) and minimum (ϕ_{min}) fuel to air ratio ϕ at which stable combustion still occurs. As can be seen from the temperature map (Figure 4a,b), the temperature in the core of the flame also rises with an increase of ϕ . While in opposite the temperature decreases with a decrease of CH_4 content in CO_2 . The latter trends are well known and logical. However, the experiments showed that the temperature increase not always plays a key role and is sufficient to maintain a stable flame. Meanwhile, considering the axial velocity ranges (Figure 5) can be assumed that a change in velocities is contrary to the temperature and a decrease at higher ϕ ratios occurs. Based on this can be stated that a reduction in flow velocity affects an inner recirculation zone, and thus, prevents passing a sufficient amount of high temperature gas, which in turn negatively affects the combustion stability. This is well proven comparing the cases with H_2 addition and O_2 enrichment at 15 vol% of CH_4 in CO_2 as the flame temperature was higher almost by 100°C under the oxygen-enriched conditions than in the case of H_2 addition (Figure 4a,b). Though, the flow velocities were in the same order of magnitude. Taking into account these findings, it can be concluded that a certain amount of heat is critical and should be ensured and maintained to ignite the incoming cold air-fuel mixture.

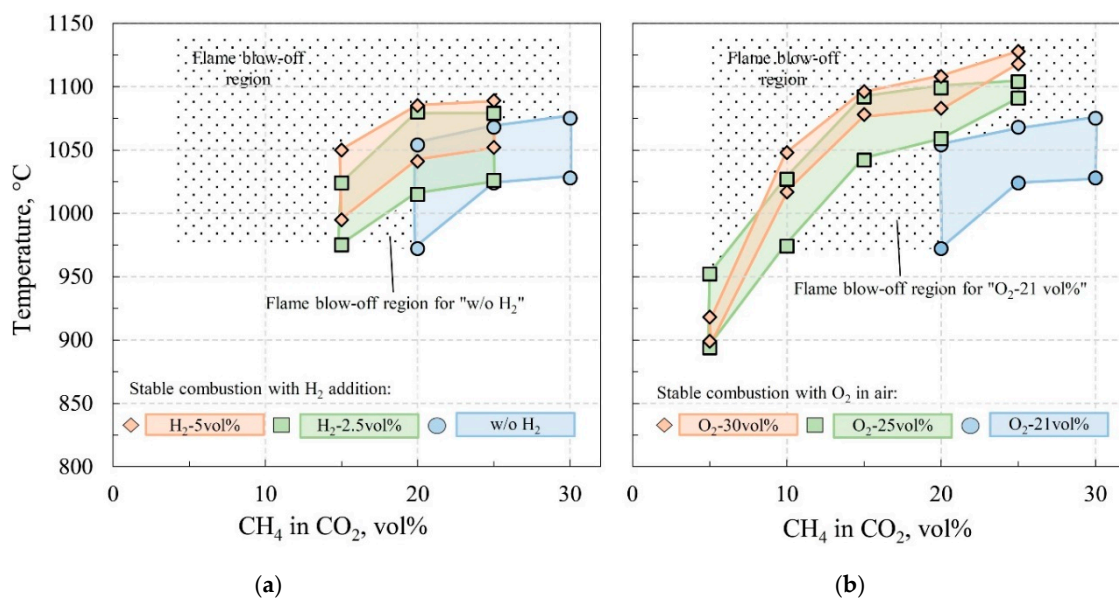


Figure 4. Temperature ranges during waste biogas (LCV) combustion with different content of CH_4 in CO_2 for a range of (a) H_2 addition and (b) O_2 air enrichment. The upper and bottom limit stands for a ϕ_{max} and ϕ_{min} , respectively.

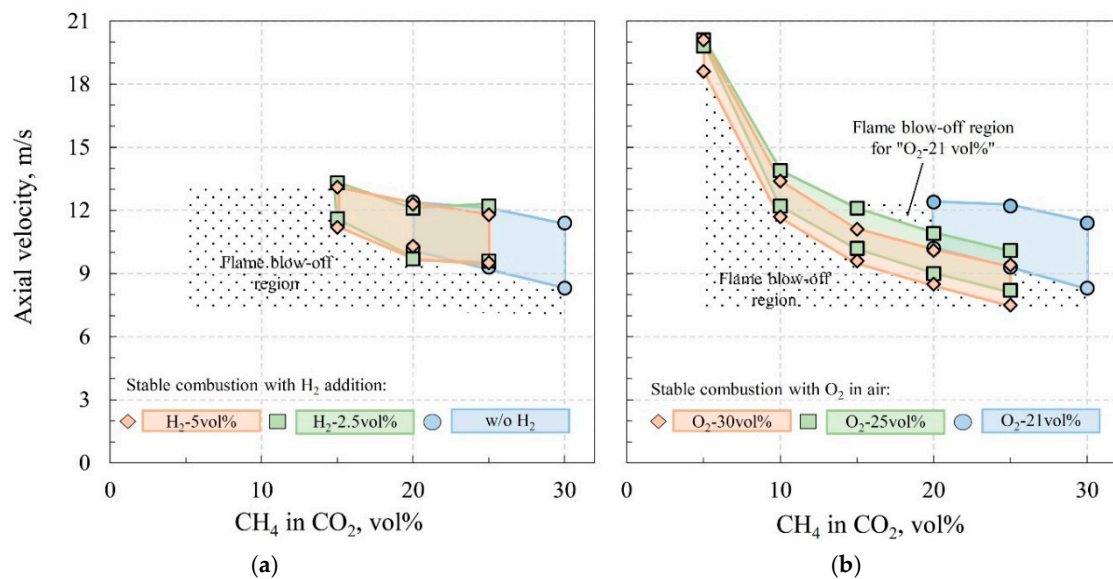


Figure 5. Axial velocity ranges during waste biogas (LCV) combustion with different content of CH₄ in CO₂ for a range of (a) H₂ addition and (b) O₂ air enrichment. The upper and bottom limit stands for a ϕ_{max} and ϕ_{min} , respectively.

3.2. Changes in Flame Chemiluminescence

For a better understanding of waste biogas (LCV) flame behavior under H₂ or O₂ addition and identify a possible performance for flame transition modes, chemiluminescence emission spectrums from flames of different LCV gas mixtures were obtained. The obtained OH* emission intensities distributed per the chamber length at $\phi = 0.85$ are presented in Figure 6. The highest OH* intensities were observed at the front of the flame (17 cm from the burner) where the main reactions take place. Comparing cases with and without H₂ enrichment, it was observed that the H₂ addition of 2.5 vol% has a negligible effect on the LCV flame enhancement, especially for the mixture with 25 vol% CH₄ in CO₂ as the combustion chamber temperatures are near identical to ones without the H₂ addition (Figure 4a). Similar results were also determined analyzing the obtained OH* emission intensities of hydrogen-enriched LCV flames (Figure 6a). Though, the increase of hydrogen addition to 5 vol% led to higher OH* emission intensities by ~1.6 times and a higher chamber temperature at point T₁ by ~30 °C burning LCV gas mixtures with CH₄ content of 25 vol% and 20 vol%. According to previous work [27], the increased OH* intensities and chamber temperatures could be attributed to an improved flame stability. However, the waste biogas (LCV) flammability limit using the H₂ enrichment was achieved burning mixtures with CH₄ content of 15 vol%, and a further decrease in CH₄ content led to the flame blow-off. In the case of the mixture with 15 vol% CH₄, lowermost OH* emission intensities, and chamber temperatures were determined using both H₂ additions (2.5 and 5 vol%). This could be related to a high level of the diluent (CO₂), which in turn lowers the flame temperature, and thus, OH* emission. The latter findings are in agreement with those found in the work of Guiberi et al. [38], where pointed out that OH* intensity decrease of CO₂-diluted flame is more intense with the increasing diluent concentration.

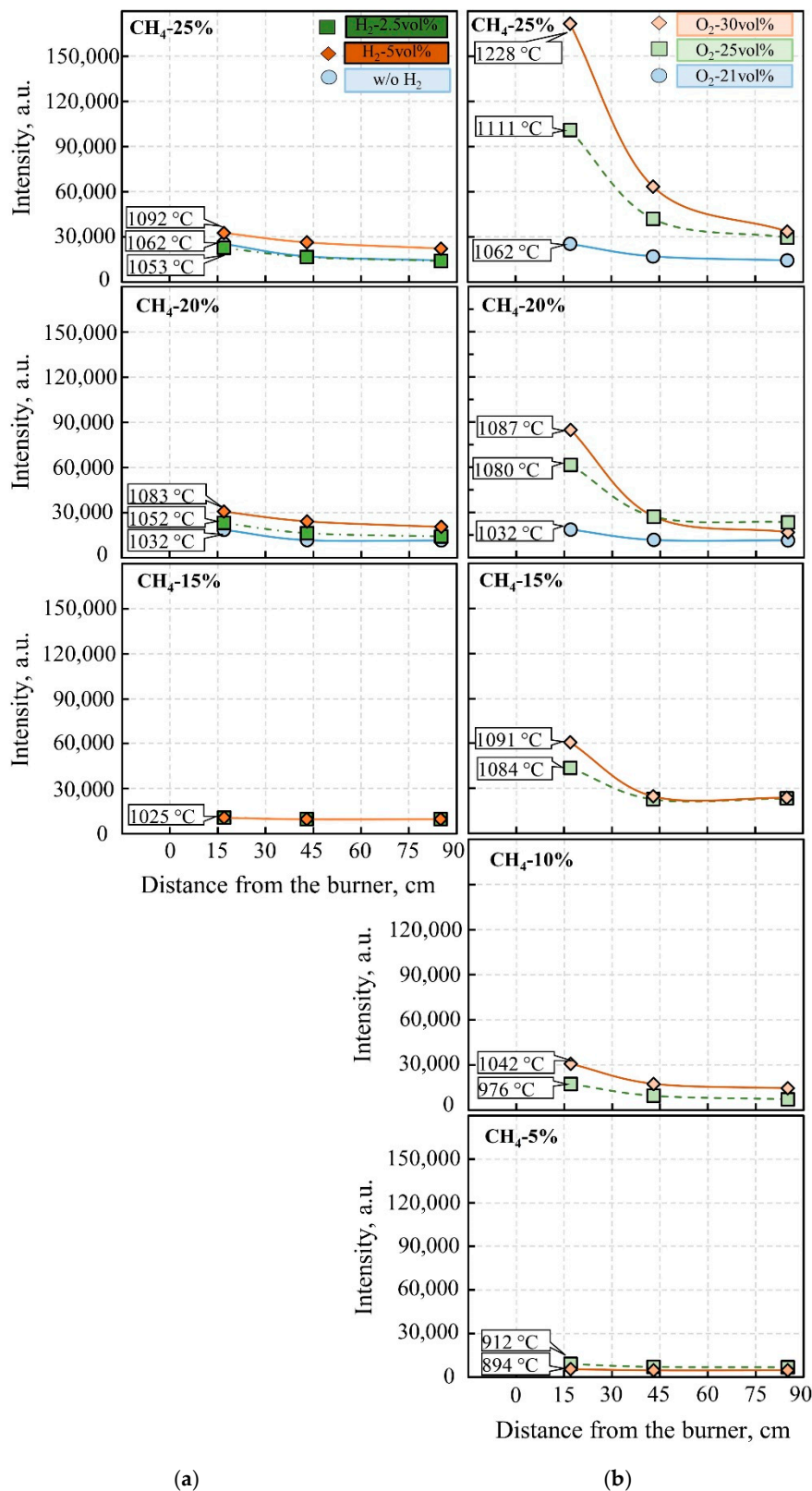


Figure 6. OH* spectral intensity distribution per distance at $\phi = 0.85$ under (a) hydrogen-enrichment and (b) oxygen-enriched air.

A bit different results were obtained using oxygen-enriched air (Figure 6b). As an example, an increased O₂ content by 4 vol% in the air affected the mixture with 25 vol% of CH₄ combustion

process significantly, and the OH* emission intensity at the flame front was almost four times higher compared to the intensity obtained under ordinary combustion in air. Besides, higher chamber temperatures were also observed (Figure 6b). Further increase in O₂ content from 25 to 30 vol% also led to increased OH* emission intensities and the chamber temperatures. A similar trend of OH* emission intensity increase, due to a higher O₂ content in the oxidizer was also determined in previous works [27,39]. According to He et al. [18], the OH* formation reaction (7) dominates under oxygen-enriched conditions as the intensity of reaction (7) is enhanced when the concentration of O₂ in the oxidizer increases and vice versa. However, with increasing concentration of diluent or decreasing concentration of methane, the OH* emissions weaken gradually, resulting in decreased spectrum intensity. This tendency was determined by burning LCV gases with lower content of CH₄ (20–5 vol%). Even though the supply of oxygen-enriched air expanded LCV gas flammability and was able to burn the mixture with CH₄ content of 10 and 5 vol%.

At the lowest CH₄ concentration (5 vol%) under oxygen-enriched air (25 and 30 vol% of O₂), OH* emission intensities were lower than that in the case of the mixture with 20 vol% CH₄ burned under air combustion mode. Though the chamber temperature at point T₁ was about 900 °C, the combustion process was stable using both oxygen-enrichments, possibly due to enhanced fuel oxidation. According to obtained results under oxygen-enriched conditions, the OH* intensity decrease coincides with the decrease of chamber temperatures, and the OH* emission intensities are related to the flame temperature. In overall, this tendency is in good agreement with spectral intensity ratios of I_{OH*}/I_{CH*} changing CH₄ content in CO₂ (Figure 7) even though the ratio of I_{OH*}/I_{CH*} is mostly used to determine the global equivalence ratio [40,41]. Increasing the CO₂ dilution level leads to decreased flame temperature as the LHV decreases, and the ratio of I_{OH*}/I_{CH*} also decreases. The supply of O₂-enriched air results in rapidly increased ratios of I_{OH*}/I_{CH*}, but it also depends on the O₂ level in the air and CO₂ levels in LCV gases (Figure 7). Considering the stability of waste biogas (LCV) combustion, the ratios of I_{OH*}/I_{CH*} determined burning LCV gases under air combustion conditions could be assumed as indicators for a threshold of the stable combustion as a further decrease in the ratios of I_{OH*}/I_{CH*} lead to blow-off. Under oxygen-enriched conditions, the lowest ratio of I_{OH*}/I_{CH*} is achieved by burning LCV gas with the lowest CH₄ content (5 vol%), and a further decrease in CH₄ content or O₂ enrichment level also leads to the flame extinguish and blow-off. Taking into account these observations, a dashed line in black was introduced in Figure 7 to represent the stability threshold of LCV gas combustion. The ratios of I_{OH*}/I_{CH*} on or above the dashed line show the stable combustion, and the combustion stability increases with increase of CH₄ content in CO₂ and O₂ level in the air. Meanwhile, the flame blow-off occurs below the stability line. Besides, the proposed indication of the stable combustion is in close agreement with the determined stability maps (Figure 4b). However, this tendency is not fully valid for the cases with H₂ addition. This could be related to low content of H₂ addition, as it might not be sufficient to represent the flame enhancement via flame chemiluminescence. The previous research showed similar results with the same level of H₂ addition, and the significant combustion improvement and higher intensities of OH* and CH* were achieved only at higher levels of H₂ addition [27].

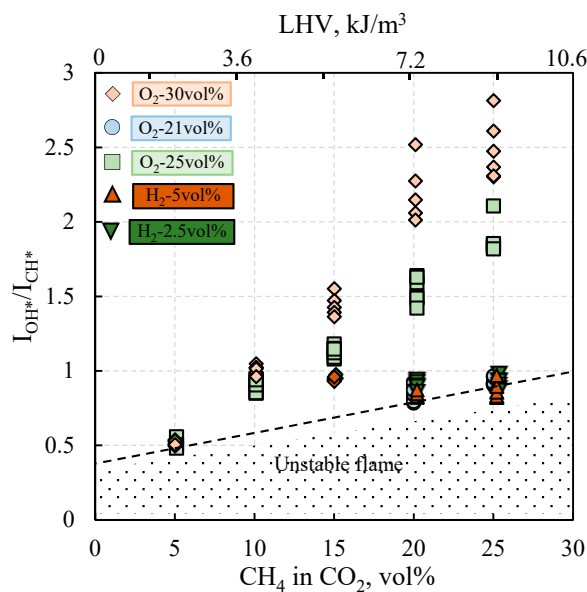


Figure 7. The tendency of the spectral intensity ratio of I_{OH^*}/I_{CH^*} with the increase of CH_4 content in CO_2 .

3.3. Addictive Influence on NOx and CO Emission

During the waste biogas (LCV) combustion tests, an online flue gas analysis was performed to determine the behavior of NOx and CO emissions under all tested cases. The obtained concentrations of NOx and CO are presented graphically in Figure 8. The burner configuration ensured low NOx (up to 25 ppm) and zero CO emissions burning LCV gases with CH_4 content of 25 and 20 vol% with air in the range of ϕ from 0.7 to 0.9. However, as noted before, the combustion at higher ϕ values (>0.9) becomes unstable, and CO is formed, in which concentration in the exhaust gases rises up to 45 ppm. This trend was determined in all tested conditions. At ϕ values below 0.9, the stable combustion is achieved, and variations in CO and NOx emissions mainly depend on the CH_4 content in LCV gases and the level of O_2 or H_2 addition. In the case of H_2 addition (Figure 8a), the NOx concentrations increased with increasing addition of H_2 compared to emissions obtained at air combustion mode. For example, in the case of the mixture with 20 vol% CH_4 , the NOx concentrations increased by 2–5 ppm and by 5–7 ppm using the H_2 addition of 2.5 vol% and 5 vol%, respectively. According to Figure 4a, it could be considered that NOx concentrations increase due to increased flame temperature, which in turn is affected by enhanced flame speed, and thermal NOx formation intensifies with higher levels of H_2 addition. Meanwhile, the CO concentrations stand at zero, due to enhanced combustion. It is important to point out, that the lowest NOx (up to 14 ppm) and zero CO concentrations using the H_2 addition are obtained burning the LCV gases with CH_4 content of 15 vol%. In comparison, this is not able to combust under normal conditions (Figure 8a). Besides, the obtained emissions of NOx during the waste biogas combustion with hydrogen addition does not exceed the emission limit value (97 ppm) according to EU directive 2015/2193 [30]. Considering the oxygen-enriched conditions, the stable combustion of the 15 vol% CH_4 in CO_2 is also achieved, but NOx concentrations increase up to 78 ppm (Figure 8b). Moreover, with the increase of O_2 enrichment in air, NOx increases gradually, even an N_2 concentration in air decreases. This tendency was also determined with LCV gases containing higher LHV values, and the highest NOx concentrations were determined (up to 1100 ppm) burning the LCV gas with CH_4 -25%. However, minor emissions of NOx (up to 20 and 40 ppm) were observed burning LCV gases with CH_4 content of 5 vol% and 10 vol%, respectively. Besides, the results show that during the combustion of waste biogas (up to 20 vol% CH_4) under oxygen-enriched conditions (25 and 30 vol% of O_2), the NOx and CO emissions do not exceed the established emission limits [30]. But the obtained results during the combustion of mixtures with a higher amount of CH_4 in CO_2 confirm that the NOx

formation strongly belongs to the thermal NO formation route under oxygen-enriched conditions. As it mainly depends on the flame temperature, the O₂ enrichment level should be controlled based on the LHV value to prevent unwanted NO_x formation.

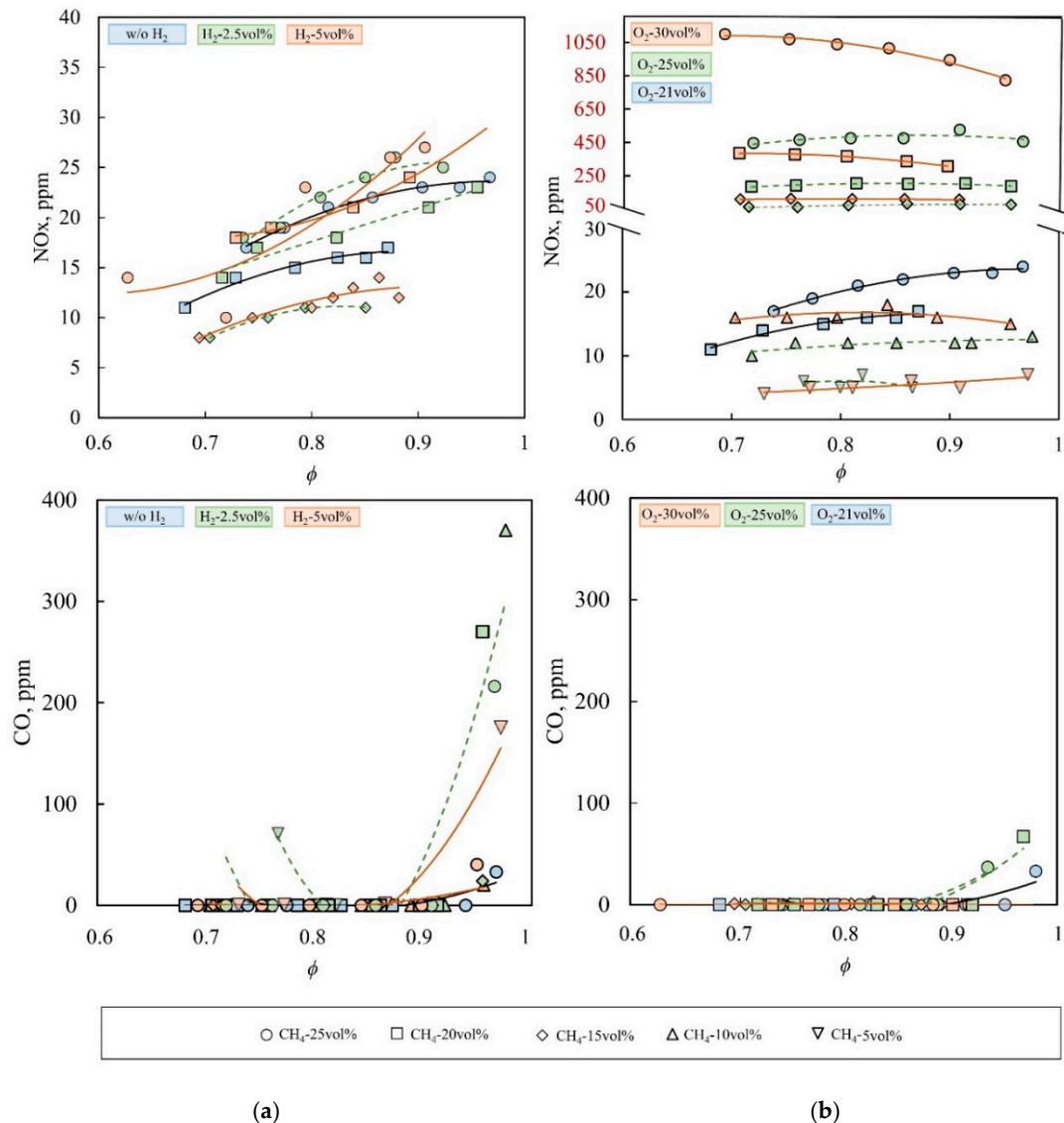


Figure 8. NO_x and CO emissions of all tested waste biogas (LCV) mixture combustion under hydrogen-enrichment (a) and oxygen-enriched air (b) versus fuel-to-air ratio.

4. Conclusions

This study describes a flexible swirl burner (~100 kW) with an adiabatic chamber, which was designed and developed to burn unstable composition waste biogases (5–30 vol% of CH₄ in CO₂) and performed tests at real conditions. During the experimental tests, the flame stability and combustion limits were explored under air, oxygen-enriched atmosphere, or with hydrogen addition. The stable operation of non-premixed swirl combustion of waste biogas in the air was achieved for CH₄ content in CO₂ with not less than 20 vol%. Further decrease of CH₄ content in CO₂ requires H₂ or O₂ enrichment to extend flammability and improve flame stability. It was determined that by adding up to 5 vol% of H₂, the stable combustion of waste biogas with CH₄ content of 15 vol% is obtained. A greater improvement in flame stability of waste biogas was established using O₂-enrichment. The supply of O₂ by 25 and 30 vol% resulted in the stable combustion of waste biogases gases containing 10 vol% and

5 vol% of CH₄, respectively. Besides, the tests of the developed swirl burner at real conditions revealed that the flame blow-off is more likely at higher fuel to air ratios ($\phi > 0.95$), and this was established for all studied cases.

This work shows the different stabilization modes and the changes in intensities of OH*chemiluminescence spectra, which enables to determine the possible transition between regimes and when flammability limit might occur. The linear correlation between the spectral intensity ratio of OH* and CH* (I_{OH^*}/I_{CH^*}) and CH₄ content in LCV gas is determined, which clearly demonstrates the change in intensity for the tested modes and can be used for the prediction of blow-off limits burning different LCV gases under different H₂ or O₂ enrichments.

During all tests, the online flue gas analysis was performed to determine the behavior of NO_x and CO emissions and explore its relation to flame stability. Minor emissions of NO_x (up to 20 and 40 ppm) are observed only burning waste biogas (LCV) in air and with H₂ addition. During combustion under oxygen-enriched conditions, such concentrations can be obtained only by combusting waste biogases containing the CH₄ content of 5 vol% and 10 vol%, respectively. The CO emissions in most cases, were determined to be at zero levels and formed only by approaching the blow-off limit. In that case, the proposed combustion system meets the EU established emission standards for gas-burning systems.

However, if waste biogas calorific value increase, O₂ enrichment should be minimized to prevent unwanted high NO_x formation. Besides, thermal damage to the combustion system and high NO_x emissions are possible if the oxygen-enriched air is supplied with higher O₂ concentrations than 30 vol%.

Overall, the developed swirl burner with the adiabatic combustion chamber is a flexible combustion system that could be installed in the biogas plant to produce heat and electricity and also provides a solution to recover primary energy and reduce pollutant emissions from biomethane production obtained waste biogases.

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