INVESTIGATION OF CO₂ DILUTED METHANE AND PROPANE SWIRLING PREMIXED FLAMES USING CH^{*} CHEMILUMINESCENCE IMAGING

by

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Utilization of hydrocarbon gaseous fuels, such as biogas, landfill gas and others, is a valuable contribution to sustainable energy production and climate changing control. The presence of CO_2 in these gases decreases heat of combustion, flame temperature, flame speed and can induce flame blow-off and combustion instabilities. In order to better understand the problem, flame geometry and location was investigated using chemiluminescence (CH^{*}) imaging technique. Combustion took place in a purposely built, lean, premixed, unconfined swirl burner, fueled by methane and propane diluted with CO_2 . The fuel type, air-to-fuel equivalence ratio and CO_2 content were chosen as the independent variables. The CH^{*} imaging by means of a commercial CCD camera, fitted with an optical filter, was used for flame investigation. The analysis of images showed that the CH^{*} emission intensity, flame geometry and location were remarkably affected by the fuel type and the air-to-fuel equivalence ratio, while the CO_2 dilution was of minor importance.

Key words: methane-propane combustion, CO₂ addition, commercial CCD camera, swirling premixed unconfined flame, chemiluminescence imaging

Introduction

The Paris climate accord of 2015 aims to support the global action to control the climate change by keeping a global temperature rise in this century below 2 degrees above the preindustrial levels [1]. The proposed action mechanism is to strengthen the mitigation of GHG and to support sustainable development, which includes RES and actions to improving energy efficiency. Utilization of biomass, such as biogas and landfill gases, is an important contribution to such efforts. Biogases are mixtures of CH_4 (45-70% by volume) and ballast components, mainly CO_2 (30-60%), and to far less extent, nitrogen (0-5%). The presence of ballast components in combustible gases decreases their calorific value, flame temperature, laminar flame speed, narrows flammability limits, affects emissions and increases susceptibility of flame to blow-off and instabilities [2-4]

While the adiabatic flame temperature and laminar flame speed are inherent characteristics of a fuel-oxidizer mixture composition, the flame blow-off and flame instabilities are controllable to a certain extent. The principle of flame stabilization is based on enabling chemical reactions to finish in a given space over a range of operating conditions. There are a number of methods to stabilize a flame, of which most often used are bluff body, swirl, dump and piloted

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flame methods. This investigation applies swirl method for flame stabilization. A swirl is the most effective aerodynamic method of flame stabilization.

The present paper deals with a premixed flame using swirl to stabilize the flame. The more detailed explanation of the aerodynamic techniques for flame stabilization can be found elsewhere [5-10]. In short, the bluff body flow is characterized by a recirculation zone behind the body with and shear flow, outer region. The flow produces a turbulent exchange of heat and mass which heats-up the incoming fuel-air mixture, mixes it with the combustion products, and increases the residence time for the chemical reactions. The similar flow pattern stands for dump flows, as well. A swirl is relatively simple yet the most effective aerodynamic method for flame stabilization [5-10]. A swirling flow, schematically shown in fig. 1, induces a complex recirculation zone which entrains outer hot combustion products and mixes them with the incoming fuel and air mixture. It is characterized by strong turbulence field and mixing on both micro and macro levels. Strong turbulence increases a turbulent flame velocity, which further increases flame stability. In addition, due to its excellent flame stabilization characteristics, swirl enables combustion of very lean mixtures which is favorable for very low emission of NO_x [11-13].

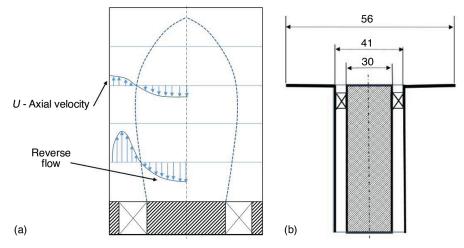


Figure 1. Schematics of swirling flow (a) and swirl burner (b) [mm]

The complex nature of turbulent, swirl flame is still not fully understood. Flame visualization techniques have been used for decades to investigate flames. They are non-intrusive methods that can give instantaneous information on flame geometry, location and flame behavior [14, 15]. Development of CCD cameras and computerized image processing has further fostered interest in the application of visualization techniques for combustion research. Flame chemiluminescence [5, 16] is a valuable and proved imaging technique for flame research. It can provide important information, such as flame geometry, location of zones of maximum heat release rate, air to fuel equivalence ratio, λ , adiabatic flame temperature, flame instabilities and others [5-10, 17, 18].

Jensen [5] presented state of the art of general techniques and instruments used for luminescence visualization, measurements and analysis. Bheemul *et al.* [6] used a set of CCD cameras to capture 2-D images of flame. A numerical algorithm was developed to present the grabbed images into 3-D ones. The authors showed that the system was capable to present the flame geometric parameters for different combustion conditions. The chemiluminescence emissions of OH^* , CH^* , C_2^* , and CO_2^* in natural gas/air premixed counter flow flames, with

variable equivalence ratio, were measured by a high performance spectroscopic unit [7]. The obtained results showed that chemiluminescence intensities of OH^{*}, CH^{*}, and CO₂^{*} were suitable indicators for heat release rates and equivalence ratios. Guethe et al. [8] overviewed possible application of chemiluminescence for gas turbine combustor development including correlation between the heat release rate and NO_x formation processes. They found that flame chemiluminescence imaging was useful technique for combustor research and development regarding flame stability and emissions. The effect of propane (C_3H_s) , isooctane, ethanol, and methanol on equivalence ratio measurements using chemiluminescence was investigated by Orain and Hardalupas [9]. The authors concluded that the OH^{*}/CH^{*} intensities ratio had monotonic relation with equivalence ratio for all test fuels, except for ethanol and methanol flames, when the effect of flame strain rate had to be considered. The chemiluminescence emissions of OH^* , CH^* , C_2^* , and CO_2^* were analyzed as a function of equivalence ratios in atmospheric CH₄-air premixed flames [10]. The same authors developed a multivariate sensing methodology to match the experimentally measured equivalence ratios in premixed CH₄-air flames. The OH*/CH* intensities ratio was used as a marker for prediction of equivalence ratios. Simona et al. [17] used the chemiluminescence 309 and 282 nm bands to investigate the effect of butanol-diesel fuel blends on spray combustion in a common rail Diesel engine. The ratio of the intensities was used to follow the flame temperature evolution. Jozsa and Sztanko [18] used flame OH^* , CH^* , and C_2^* emission spectroscopy to compare the steam-blast atomization and the airblast atomization of a diesel oil in an atmospheric, lean, prevaporized, premixed burner. The ratios of OH^*/CH^* , OH^*/C_2^* , and CH^*/C_2^* chemiluminescence intensities were used as markers. The authors concluded that the steam blast atomization resulted in lower adiabatic flame temperature compared to the air blast atomization. The temperature reduction lowered the C2 luminescence intensity more than the intensities of OH^{*} and CH^{*}.

While many investigations deal with CH4, C3H8, isooctane, methanol, ethanol, butanol, diesel, kerosene and other fuels [5-10, 17, 18], few were concerned with biogas, or CH₄ and CO₂ blends [19-25]. Guiberti et al. [19] used combined numerical and experimental investigation of OH^* , CH^* , and CO_2^* chemiluminescence intensities of a laminar, premixed, conical CH_4 flames, as a function of dilution with CO₂ and N₂ using a spectrometer and an ICCD camera fitted with optical filters. Marsh et al. [20] used a CCD camera coupled with an optical filter for OH^{*} chemiluminescence in CH4-air premixed confined swirl flames, diluted with CO2 and N2, to investigate the effect of dilutions on flame location, shape and heat release rates. The authors concluded that the effect of N_2 was negligible, while the effect of CO_2 dilution on flame location was measurable. Cosić [21] used a CCD camera with an optical filter to investigate a correlation between CH^{*} and air coefficient in a laminar premixed C₃H₈ flame of an atmospheric burner with controlled flow rates of air and fuel. The linear correlation between CH^{*} and air coefficient was found. Ballachey and Johnson [22] investigated blow-off conditions of a low swirl burner using particle image velocimetry (PIV) for different compositions of fuel (CH₄, H₂, CO₂, and CO), different burner geometries and swirl number. Littlejohn and Cheng [23] conducted experimental investigation of different fuel compositions (CH₄, C₂H₄, C₃H₈, H₂, CO₂, and N₂) on lean blow-off velocity in a gas turbine low swirl injector, using PIV. The authors concluded that the swirl injector operated well without need for significant changes, but further studies were needed when burning mixtures with H₂. Khallil and Gupta [24] investigated turbulent, confined, swirling combustion, using premixed and non-premixed CH₄-air mixture with the air-to-fuel equivalence ratio from 1.25 to 2.0, at constant heat load. The flame OH* chemiluminescence was captured using an ICCD camera fitted with a 307 nm optical filter. The NO_x and CO emissions were

measured and analyzed. The images of OH^* showed the increased distributed combustion when an extended flue gas exit tube inside the combustor was present, leading to a higher residence time, improved CO emission at low levels of NO_x emission.

Regarding premixed, lean, turbulent, swirl, confined combustion of biogas, using CH* chemiluminescence imaging technique, there are a few published papers in the available literature. Shi et al. [25] used tubular confined flame, CH4 fueled burner with tangential injection of pure CH₄, or CH₄-CO₂ mixtures, and through separate ports, pure oxygen, or oxygen-CO₂ mixture. The conventional digital cameras for flame imaging were used, while the OH*, CH* chemiluminescence images were acquired by a high speed intensified video camera and optical filters. The authors found that the direct images were unclear to understand while CH* chemiluminescence images enabled understanding of the flame position, structure and behavior. No Abel deconvolution was applied, despite the flame was axisymmetric. Instead, the images were taken from the flame-sidewise and flame-top positions. The CHEMKIN-Pro software was used to calculate the chemical reaction times and laminar burning velocities. The authors analyzed flame images, flame stability and CH^{*} radical distribution as a function of oxygen-CO, dilution and equivalence ratio variables. Lafay et al. [26] investigated effects of CO₂ on combustion of CH_4 and simulated biogas (CH_4 , CO_2 , and N_2) at different air coefficients, in a swirl, premixed, confined flame burner, using a scientific grade ICCD camera for visualization of CH^{*} radical. The velocity profiles were provided by a laser Doppler anemometry. The authors investigated flame stability and flame structure. Biogas showed narrower flame stability limits than pure methane. The presence of CO_2 , for the same values of air coefficient, strongly affected reaction zone location. The authors found that the laminar flame speed was the main parameter for the prediction of flame structure.

The present investigation aims at characterization and analysis of premixed, lean, turbulent, swirling, unconfined flames of CH_4 and C_3H_8 diluted with CO_2 , using flame CH^* emission imaging technique by means of a commercial CCD camera fitted with an optical filter. The investigation has shown that under laboratory conditions, the applied CH^* imagine technique can reveal flame geometry and locate the flame front with the precision of the order of millimeters. There have been numerous investigations of lean, premixed, confined swirling flames of biogas, using chemiluminescence spectroscopy, ICCD and CCD cameras. Among them only a few investigations used a commercial CCD camera for the flame CH^* emission imaging, and to our best knowledge, there is a lack of such investigations under flame unconfined conditions in the open literature.

Experimental facility

The experimental facility consisted of a laboratory scale swirl burner, a fuel and air supply system. Schematics of a swirling flow and the burner are shown in fig. 1. The burner was purposely designed for research of lean, premixed, low NO_x and CO emission, gaseous fuel combustion [27]. The burner can burn low calorific value gases, such as, biogas, landfill and natural gas with increased content of CO_2 and N_2 , with 10:1 thermal power dynamic range, avoiding flame flashback and combustion instabilities.

The fuel and air are injected independently, each through six ports, perpendicular to the burner axis at the entrance section. The length of the annular section, from fuel/air injection ports to the annulus exit is twenty annulus widths. The premixing of fuel and air is performed in the burner annulus. The quality of mixing of fuel and air was checked and found that the differences between actual and perfect premixing were better than \pm 5% [27]. A flat-vane, axial flow swirler, with vane angle of 45° is mounted in the annulus, at 20 mm before the burner exit.

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The CH₄ and C₃H₈ were used as basic fuels. Biogas, landfill gas and lower calorific value natural gases were simulated by addition of CO₂ into CH₄. Fuels and CO₂ were supplied from pressurized cylinders, while the air was supplied by a fan. The uncertainty of flow rate measurements for CH₄, C₃H₈, and CO₂ was \pm 5% (rotameters manufactured by Dwyer Instruments, Inc.), while the air-flow rate uncertainty was \pm 3% (orifice flow meter; BS 1042 fluid-flow in closed

conduits). The quality of CH_4 and C_3H_8 was 99.5%, The experiments were performed at constant nominal thermal power of 4 kW, the corresponding Reynolds number was 8500-12000, depending on fuel type and λ . The CO₂ content and air-to-fuel equivalence ratio were systematically and independently varied, as shown in tab. 1.

Fable 1. Fuel composition	and	air-to-fuel
equivalence ratios		

Fuels	CO ₂ content in fuel-CO ₂ mixture, [%]	Air to fuel equivalence ratio, λ
Methane and propane	0, 10, 20, 30, 40	1.0, 1.1, 1.2, 1.3, 1.4

Flame imaging and image processing

A Nikon D80 digital camera with Nikkor AF f 4-5.6D ED 70-300 mm telephoto lens fitted with a narrow band optical filter was used for the CH^* emission imaging of flame. An optical filter was chosen to fit the CH^* chemiluminescence emission which is centered at 430 nm. Technical details of the imaging system are given in tab. 2.

Table 2. Camera and optical inter details				
Camera	Nikon D80; DSLR			
CCD sensor	APS-C 10.8 MP; 23.6 mm × 15.8 mm Nikon DX format			
Resolution	3.872×2.592 (10.2 effective megapixels)			
Telephoto lens	Nikkor AF f 4-5.6D ED, zoom 70-300 mm			
Optical filter	Ealing 35-3300, 430.0 ±2.0 nm, transparency 51%			
	attenuation of other wave lengths >99%			

Table 2. Camera and optical filter details

Preliminary investigation and analysis were taken to set the CCD sensor sensitivity, camera exposure time and aperture, keeping in mind that the entire flame should be in focus, which is controlled by the camera aperture setting. The lower CCD sensor sensitivity produces lower noise in images while longer camera exposure improves signal to noise ratio. The final settings of the CCD sensor sensitivity, camera exposure time and aperture were ISO 200, 3 seconds and f8, respectively [28].

The preliminary investigation showed that the exposure time of 2 ms was fast enough to get the images that revealed turbulence-flame structures, as can be seen in fig. 2(a). The exposure time of 3 seconds produced images without noticeable turbulence-flame structures fig. 2(b).

In order to get images which CH^{*}emission intensities can be compared, the highest value of the acquired light intensity of the investigated flames, was set 250 on the grey

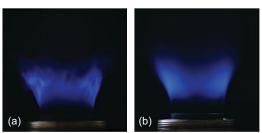


Figure 2. Flame images taken without optical filter; exposure time 2 ms (a) and 3 seconds (b).

scale level of 0-255. In practice this corresponded to the C_3H_8 -air stoichiometric flame. Consequently, the maxima of the acquired CH^{*} emission intensities for all other cases: C_3H_8/CO_2 ,

 CH_4/CO_2 mixtures, and $\lambda > 1.0$, were less than 250. The effective format of images was 249 pixels per line (*x*-co-ordinate) and 204 pixels per field (*z*-co-ordinate).

The acquired luminescence images of flames were processed by in house developed software [21]. To get an impression of the CH^* intensity distribution on the 0-255 scale, is shown in fig. 3.

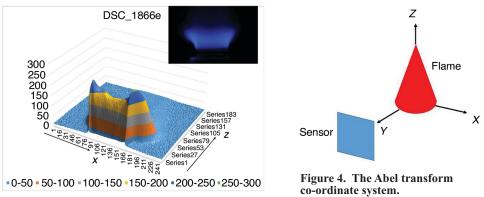


Figure 3. Illustration of acquired CH* chemiluminescence; the values are given in arbitrary units (*a.u.*); C₃H₈-air flame, $\lambda = 1.0$, CO₂ = 0%

The Abel deconvolution was done numerically using in-house developed software to be published in 2019. The flame was considered axisymmetric with a local CH^{*} luminescence intensity i = i(x, y, z), as a function of the radial distance $r, r^2 = x^2 + y^2$, and the co-ordinate z, fig. 4. One should note that the beginning of the co-ordinate x differs in the two co-ordinate systems, as shown in figs. 3 and 4.

The flames are assumed axisymmetric, therefore, one half of an image represents the flame as a whole. In the first step of the image analysis, the image zone with the highest values of grey level of CH* emission intensity is assumed the flame zone [7, 8]. In the next step, the Abel deconvolution is applied to find the grey level distribution in the flame zone.

Results and discussion

The photographs of CH^* emission intensity are used for the qualitative and quantitative interpretation of flame geometry and location. The flame location is assumed a zone with the local maxima of CH^* emission intensity [7]. As most of the original photographs were too dim for a naked eye, they are shown here in their inverted forms.

It is important to stress that the combustion took place under unconfined conditions which affects the flame due to the entrainment of the cold ambient air into the flame zone which increases the local content of air in the fuel-air mixture, decreases the flame temperature and increases the required time for chemical reactions. As a consequence, the flame location is shifted downstream. Besides, the flame may become prone to instabilities and flame blow-off can occur, as well. The increased mean velocity of the injected mixture of fuel and air, with the increase of nominal λ , further adds to the air entrainment effects due to the increased fuel-air flow momentum and turbulence intensity. Geometry, dimensions and location of flame depend on burner geometry, swirl number, Reynolds number, turbulence, fuel type, air-to-fuel equivalence ratio, laminar and turbulent flame speeds, confinement and other parameters [4, 11-13]. A swirl induces recirculating annular vortex, entrains fresh air, as mentioned before, but also re-circulates the combustion products with the incoming fresh fuel-air mixture, thus increasing the stability of

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flame. The re-circulating flow, produced by the swirl and the center bluff-body, stabilizes the flame in an annular vortex flow. In addition, the expansion of high temperature combustion products tends to shift the flame in outward direction further adding to flame geometry.

The acquired CH^{*} emission images of C_3H_8 flames, as a function of λ and CO_2 , are shown in fig. 5. The intensity of CH^{*} emission is presented in the arbitrary units (a.u.). When the air-tofuel equivalence ratio increases from 1.0 to 1.4, the mean exit velocity of C_3H_8 -air mixture increases 1.4 times, the adiabatic flame temperature decreases from 2257 to 1920 K and the laminar flame speed decreases from 0.41 to 0.26 m/s [13]. The flame becomes narrow and shifted downstream, as can be seen in figs. 5(a)-5(b), and 5(c)-5(d). The effect of CO₂ dilution, for $\lambda =$ constant, is shown in figs. 5(a)-5(c) and 5(b)-5(d). As can be seen, the effect of CO₂ on flame geometry and location is small because even the maximum $CO_2 = 40\%$ dilution increases the mean velocity of fuel-CO₂-air mixture less than 2%, while the stoichiometric, adiabatic flame temperature decreases from 2257 to 2211 K, or 2.0% [13], resulting in the flame which is marginally shifted downstream.

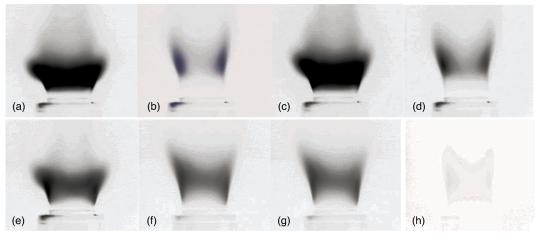


Figure 5. Effect of CO₂ and λ on CH^{*} emission of C₃H₈ (1st row) and CH₄ (2nd row) flames; (a), (e) $\lambda = 1.0$, CO₂% = 0; (b), (f) $\lambda = 1.4$, CO₂% = 0; (c), (g) $\lambda = 1.0$, CO₂% = 40; (d), (h) $\lambda = 1.4$, CO₂ = 40%

In order to illustrate the flame shapes and locations in a visually more comparable way, the halves of images are presented together, as shown in fig. 6.

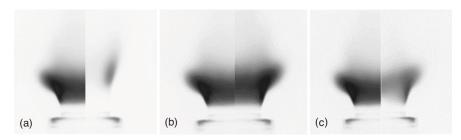


Figure 6. (a) Effect of λ on CH^{*} emission of C₃H₈ flame; CO₂ = 0%, λ = 1.0 (left half of image) and λ = 1.4 (right half); (b) effect of CO₂ on CH^{*} emission of C₃H₈ and C₃H₈-CO₂ flame, λ = 1.0, CO₂ = 0% (left half) and CO₂ = 40% (right half); (c) effect of fuel type on CH^{*} emission, λ = 1.0 and CO₂ = 0%, C₃H₈-air (left half) and Ch₄-air flames (right half)

The CH^{*} emission of C_3H_8 is about two times greater than that of CH₄ due to the favorable molecule structure of C_3H_8 , regarding the production of C_2H , the key radical for CH^{*} chemiluminescence emission.

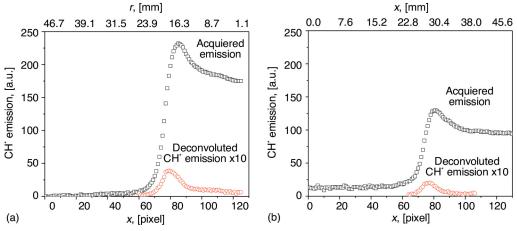


Figure 7. The CH^{*} emission and Abel deconvoluted curves; C_3H_8 (a), CH₄ (b); $\lambda = 1.0$, CO₂ = 0%

Concerning the CH^{*}emission intensity a typical example is shown in fig. 7. One should note that in order to make the curves visually comparable, the true values of the Abel deconvoluted curves are multiplied by 10. The deconvoluted CH^{*} curve peak area, fig. 7, represents a flame zone where the main chemical reactions take place. It is worth noting that the location of the flame zone is between the inflection point and the peak of the CH^{*} emission curves.

The effects of fuel type, air-to-fuel equivalence ratio and CO_2 content on CH^* emission are compared and analyzed. The deconvoluted CH^* emissions of CH_4 and C_3H_8 in the radial direction, as a function of CO_2 content and r, are presented in fig. 8. The results show that the CH^* emission intensity of C_3H_8 flame is about twice the intensity of CH_4 flame. This is because the main reaction path for CH^* emission is via C_2H radical, the main CH^* precursor, which concentration is higher in C_3H_8 than in CH_4 flames. The flame zone of CH_4 is narrower compared to the C_3H_8 flame zone. The effect of CO_2 dilution is small; the intensity maxima are marginally

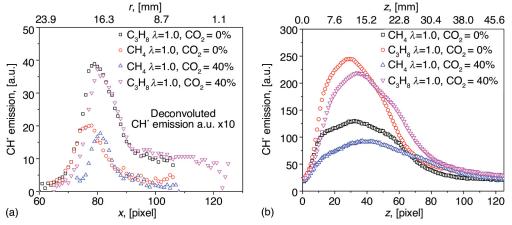
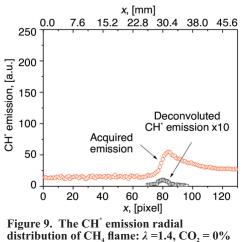


Figure 8. The CH^{*} distribution in radial direction (a) and in axial direction (b), as a function of fuel type and CO₂ content, at stoichiometric conditions.

lower and slightly shifted, about 1 mm, towards the axis. This is expected as the CO_2 dilution slightly decreases the adiabatic flame temperature.

The CH^{*} emission peaks for all flames in the radial direction, as presented in figs. 8, are located in the 2 mm thick zone, with marginal shift of the CO₂ diluted flames towards the flame axis. The presence of CO_2 shifts CH^{*} peaks about 3 mm downstream. It is interesting to note that the peaks of CH^{*} emission can be located with the precision of order of millimeters.

As already mentioned and again confirmed in fig. 8 the CH^{*} emission intensity is greater for C_3H_8 vs. CH₄ flames due to chemical structure of C_3H_8 , which favors production of C_2H , the main precursor of CH^{*}. The increase of λ from 1.0 to 1.4 decreases



the CH^{*} emission to about one half of its value for $\lambda = 1.0$. The radial distribution of CH^{*} emission of lean CH₄-air flame is shown in fig. 9.

The effect of λ on flame location in axial direction is pronounced, on the contrary to the effect of CO₂, which is small, as can be seen in fig. 10.

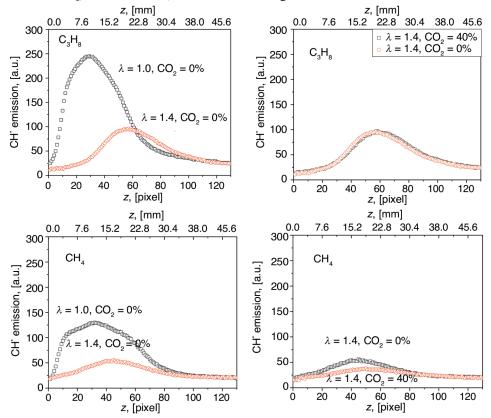


Figure 10. The CH^{*} axial distribution of C_3H_8 and CH₄ flames as a function of λ and CO₂.

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In conclusion, regarding the present investigation, there have been numerous, published investigations based on flame chemiluminescence, but rather limited number on biogas and hydrocarbon gases diluted with CO_2 . Mostly, chemiluminescence emission has been acquired and measured using specialized spectroscopic equipment, but quite small number using a commercial CCD camera. When it comes to the premixed, turbulent swirling flames of biogas or CH₄ diluted with CO_2 , the number of published papers is very limited. Shi *et al.* [25] and Lafay *et al.* [26] performed investigations similar with the present one, but with very important differences. The authors [25] investigated confined, not premixed flame of CH₄ and pure O_2 , or O_2 and CO_2 mixtures without Abel deconvolution. Instead they took the images in line with the combustor axis, at upstream and downstream positions. Investigation of Lafay *et al.* [26] can be considered most similar with the present one, but also with important differences. The authors used a simulated biogas to analyze the velocity field, flame structure, flame speed and the instability frequencies, but under the confined flame conditions. They found that the flame structure was directly linked to the laminar flame speed with the bifurcated flame, located in the corner and in the internal (close to the combustor axis) re-circulation zones.

Conclusion

The effects of fuel type, the CO₂ dilution (simulated biogas) and air-to-fuel equivalence ratio on the flame geometry and location, for the swirling, lean, premixed flame under unconfined conditions, have been investigated, using a commercial CCD camera and a CH^{*} chemiluminescence imaging technique. The C₃H₆ vs. CH, showed about two times stronger CH^{*} emission intensity. Air to fuel equivalence ratio had strong effect on the flame shape and location, while the effect of CO₂ was of minor importance. Both, CH₄ and C₃H₈ were similarly affected with the presence of CO₂. There was strong effect of air to fuel equivalence ratio on the CH^{*} intensity, flame geometry and location. The increase of air-to-fuel equivalence ratio decreased the CH^{*} intensity, made the flame mostly narrower and shifted downstream. For the stoichiometric mixtures of both fuels, the CO₂ decreased the CH^{*} emission intensity to some extent and slightly shifted the flame downstream. For lean mixtures of both fuels ($\lambda = 1.4$) the effect of even the highest concentration, $CO_2 = 40\%$, was practically insignificant. The flames were slightly narrower and shifted marginally downstream. The results showed, that using a commercial CCD camera, fitted with an optical filter, the CH^{*} chemiluminescence imaging technique was suitable method for the flame geometry and location analysis of the turbulent, swirling, premixed, unconfined flame of C_3H_8 and CH_4 , diluted with CO_3 .

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