Effect of Ozone on Combustion Performance of BioMethane (SNG) and Low Calorific Gas

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Abstract

The EU is tightening the requirements of environmental protection due to influence of pollutants from combustion systems on climate change and prompt to increase the efficiency and usage of alternative fuels. One of alternatives is biomethane (SNG), which production via thermochemical processes is gaining more attention. Also, waste gases could be produced during methanation and clean combustion of such gases is needed. However, these gases could have a varied part of ballast gas and low calorific value, which leads to decreased flame stability and burning velocity. Besides, the combustion temperature could decrease as well and the formation of intermediate fuel oxidation products could increase.

To increase combustion efficiency, ozone-assisted combustion was examined in a flat flame burner and a low swirl burner burning biomethane (SNG) and low calorific value gases. An addition of ozone reduced the flame lift-off by 20-70% during combustion of SNG and waste gases. Regarding the post combustion emissions, the ozone enrichment of SNG combustion reduced CO emissions but slightly increased NOx concentrations at higher ϕ values. The tendency on NOx increase due to the ozone addition was determined with LCV gases as well, but the ozone effect on reducing the CO emissions was more intensive, CO emissions were decreased by 70-300 mg/m^3.

Keywords: SNG, methnation, waste gas, ozone, combustion, flame stability, NOx.

1. Introduction

Nowadays, the usage of fossil fuel is still increasing and 80% of world energy is produced by combustion. Besides, the use of natural gas is projected to increase by nearly 12% in

2020 [1]. In addition, alternative fuels are getting more attention due to the demand of a more flexible and reliable energy supply system. In nowadays, the biomethane production via methanation of gases obtained in the biomass gasification process [2,3] is gaining a lot of attention and biomethane (SNG) is one of alternatives foreseen to replace natural gas. Another alternatives for heat and electricity production are gases obtained from various industrial processes (blast furnace gases, syngas, tail gases and etc.). However, these gases have a lower calorific value compared to methane and depending on gas composition, these gases could affect the combustion process, negatively. For example, high amounts of incombustible gases like N_2, CO_2 and etc. could lead to a reduction in flame temperature which in turn affects flame stability and reduces burning velocity [4–6]. According to [7,8] the CO_2 in gas mixtures could reduce NO_x emissions but the CO emissions could increase by half. Moreover, the combustion process of mixtures with CO_2 could end in reduced combustion efficiency, an unstable flame and unburned gas emissions into the atmosphere.

These problems require innovative solutions to stabilize flame. Investigations of combustion processes by other authors reveal that the flame stability is improved by enrichment of oxygen, hydrogen or syngas [9–11], combustion in porous burners [12,13] or flameless combustion [14,15]. One more solution for it, is the use of plasma technologies (thermal or non-thermal plasma) to ensure the stability and efficiency of the combustion process [16,17]. Over the last decade, promising results were presented and flame stability improvement was studied using a repetitive pulsed plasma [18,19], a gliding arc plasma [20], a laser-induced plasma [21] and dielectric barrier discharge plasma [22–24]. As an example, study on the dielectric barrier discharge showed the improved flame stability of jet flames and flame speed was enhanced by about 125% [25]. The authors observed that the plasma enhanced the flame lift-off velocity by about 125%. Other work [26] investigated an effect of different plasma discharge repetition rates on the flammability. It was determined that the flammability limit is extended from $\varphi = 0.6$ to 0.53 increasing the repetition rate from 10 kHz to 50 kHz. Similar work was presented by Barbosa et al. [27]. It was also noted that the repetition frequency influences the effectiveness of the plasma. Authors observed a significant improvement in the flame stability and the flammability limit was extended from $\varphi = 0.4$ to 0.11 at 30 kHz repetition rate.

However, the literature review reveals that studies on the plasma assisted combustion processes are mainly oriented for the application in gas turbines (Kim et al., 2017). Besides, majority of studies focuses on the effects of ozone on flame propagation speeds and ignition characteristics, while the effect on flame stability and flammability limits is analysed insufficiently. Due to these reasons, this work analyses the use of non-thermal plasma for flame stability of the biomethane (SNG) and waste gases with low calorific value in the lean combustion regime and seeks to expand the knowledge of the ozone-assisted combustion. Also the ozone effect was investigated under oxygen-enriched conditions.

2. Experimental methodology

The experiments were performed using the experimental setup consisting of two different burners: a flat flame (A) and a low swirl burner (B), a quartz glass tube, a flame optical analysis system and an air and gas mixture supply system (Fig. 1).

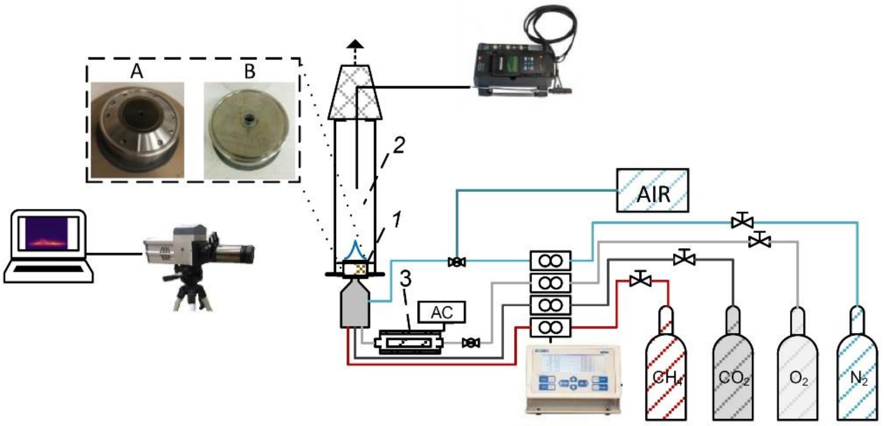

Figure 1. Schematic diagram of the experimental rig for combustion investigation: 1 – a burner of: A- a flat flame type; B – a low swirl type; 2 – a quartz tube; 3 – a DBD plasma reactor.

The combustion process was performed in the burner covered with the clean quartz glass tube of 73 cm height and 13 cm diameter used as a combustion chamber and a shield to prevent ambient air suction. For combustion experiments, biomethane (SNG) and low calorific value (LCV) gases were simulated using pure CH_4 (methane) and various mixtures of CH_4 and CO_2 (carbon dioxide) (Table I). The ozone-assisted combustion of SNG and LCV gas A was investigated in the flat flame burner. Meanwhile, the combustion of the LCV gases B and C with lower calorific value under oxygen enriched conditions (40 vol% of O_2 in N_2) and ozone addition was investigated in the low swirl burner. The mentioned gases and combustible air were supplied by separate inlets, which ensured a precise preparation of a premix. Each flow of gases was controlled by mass flow controllers BROOKS. During the combustion experiments, the equivalence ratio ϕ was changed in the range from 0.58 to 1.0.

In a case of ozone assisted combustion, oxygen and nitrogen were supplied separately. O_2 was used to produce ozone (14 mg/l) supplying it via a DBD plasma reactor (200 W) to the premixing chamber. To obtain the same composition of air, flows of N_2 and O_2 were controlled by mass controllers as well. In order to observe the combustion process improvement by the ozone introduction and to compare the obtained results, the overall flow speed of 0.14 m/s was sustained by supplying a constant 24 l/min flow of air/fuel mixtures at different fuel equivalence ratios φ.

Table 1. Composition of used gases.

Gases	Composition
Biomethane (SNG)	CH_4-100%
Waste gas A	CH_4-40%/CO_2-60%
Waste gas B	CH_4-25%/CO_2-75%
Waste gas C	CH_4-20%/CO_2-80%

The combustion process was observed using an optical system for analysis of spatial distribution of the excited species OH* (282.9 nm), CH* (387.1 nm) and C_2* (514 nm) in the flame at atmospheric pressure. The flame images were captured using an ICCD (Intensified Charge Coupled Device) camera Andor iStar DH734. The diameter of the photocathode (intensifier) was 18 mm; a pixel size was 13 μm. The matrix contains 1024×1024 active pixels sensitive to the 200–800 nm wavelength range. The achieved resolution of the flame image was 6.6 pixels per millimetre. A single accumulation cycle time was set to 1.266 s. The optimal exposure was set to 0.04 s and 300 image acquisitions were accumulated into a single frame for each experiment.

Pollutant emissions (NO_x, CO) were measured by a flue gas analyser Testo 350XL. The probe was inserted at a 50 cm distance from the burner nozzle via the open end of the quartz tube for sampling (see Fig. 1).

3. Results and discussions

The combustion experiments using gases of different compositions (see Table 1) were performed in the flat flame burner and low swirl burner with ϕ ranging from 0.61 to 1. The first set of experiments was performed using the flat flame burner, and the second one – the low swirl burner.

3.1. Flame characteristics

Obtained biomethane and waste gas flame images through the bandpass filters at the emission wavelengths of OH*, C_2* and CH* radicals at fuel equivalence ratio $\phi = 0.66$ and 0.71are presented in Fig 2. According to images of OH* and CH*, the ozone addition to biomethane flame has neglible effect. Only an insignificant improvement in a flame shape was indicated by flame images obtained through bandpass filters at the emission wavelengths of C_2*. According to [28], the direct reaction between ozone and methane is immeasurably slow and only an improved oxidation of methane by O (1) is capable. A bit different results were observed with waste gases. During the waste gas A combustion without addition of ozone, the flame is lifted up and becomes unstable. According to images of C_2* (Fig. 3), the flame “butterfly” is formed in both cases. When the combustion is enhanced by ozone, the waste gas flame becomes more stable and the flame stretch is reduced. The observed emissions of OH*, C_2* and CH* radicals also shows increased intensity of these radicals. Based on [11] it indicates a higher flame temperature due to improved fuel oxidation process.

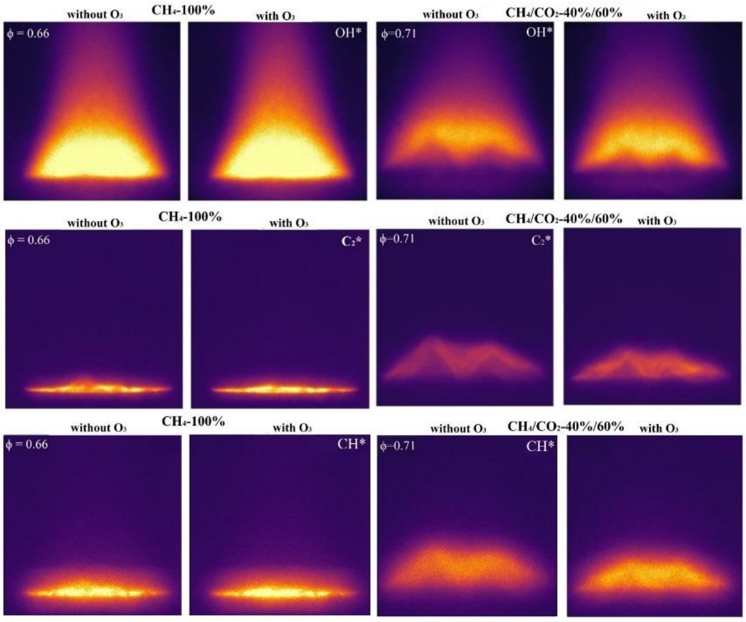

Figure 2. Flame images of SNG and waste gas A obtained through bandpass filters at the emission wavelengths of different radicals

Combustion of waste gas with lower amount of methane (≤ 30 vol% CH_4 in CO_2) was not possible in the flat flame burner and further experiments were performed in the low swirl burner supplying oxygen enriched air. Experiments were performed burning waste gases with composition of CH_4/CO_2-25%/75% and CH_4/CO_2-20%/80%. The obtained flame images of waste gases under oxygen enriched atmosphere and ozone addition are presented in Fig. 3.

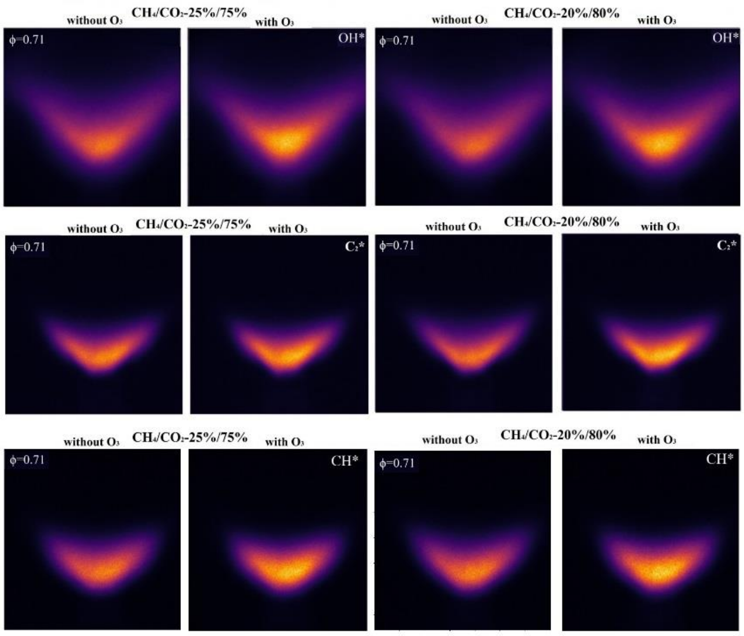

Figure 3. Flame images of waste gases obtained through bandpass filters at the emission wavelengths of different radicals

It shows that during waste gas combustion under oxygen enriched atmosphere, the flame is lifted up and stretched. Moreover, the flame lift-off increases increasing the CO_2 concentration in the waste gases. When the combustion is assisted by ozone, emission intensities of OH*, C_2* and CH* radicals increases and indicates the higher flame temperature (Fig. 3). However, the flame stability improvement by ozone addition is not noticeable from the obtained images. For this reason, to evaluate the ozone effect on flame stability, the flame lift-off height was determined by measuring the vertical distance at which the average intensity of C_2* radical emission is at its maximum.

3.2. *Lift-off height of ozone-assisted flames*

The evaluated heights of the maximum C_2* luminescence intensity with ϕ ranging from 0.58 to 1 for different gases are presented in Fig. 4. The effect of ozone addition to SNG (CH_4-100%) flame is insignificant, the flame lift-off is reduced only by 0.02 cm with ϕ ranging from 0.71 to 1.0. At leaner combustion regime (ϕ = 0.58 – 0.71), the ozone addition decreased the flame lift-off by 0.43–0.21 cm compared to the combustion case without ozone. A greater effect of ozone addition was observed during combustion of LCV gas with composition of CH_4/CO_2-40%/60% (see Fig. 4). As in the previous case, the most intensive effect of ozone-assisted combustion was determined in lean-combustion region (ϕ = 0.63) and the flame lift-off was reduced by 0.32 cm as well compared to the combustion case without ozone (see Fig. 4).

Increasing ϕ from 0.66 to 1, the flame lift-off decreased by 0.24–0.1 cm as well (see Fig. 4). Comparing SNG and waste gas combustion results, it was assumed that the higher effect of ozone on LCV gas combustion was achieved due to increased CO_2 concentration in the LCV gas and O_3 affected CO_2 resulting in CO, which increased the calorific value of these gases.

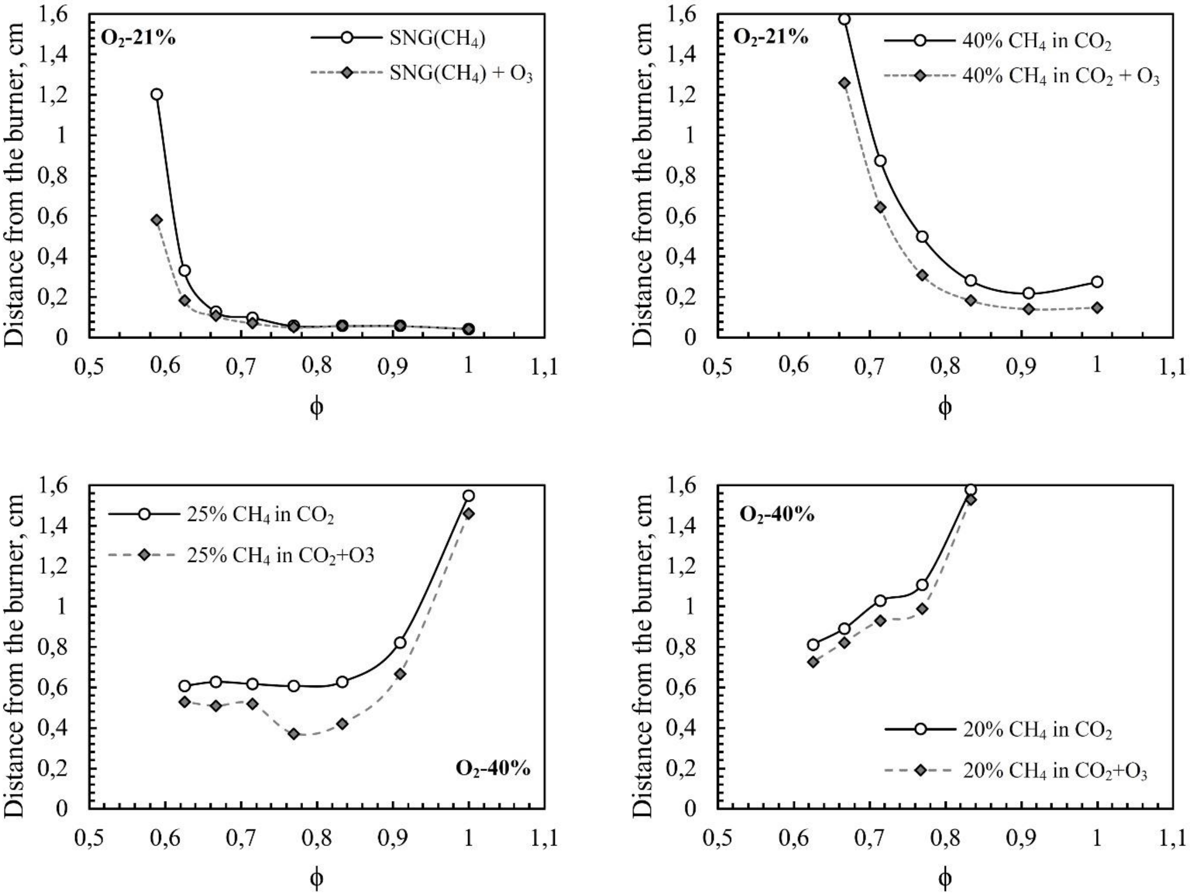

Figure 4. Vertical flame position during combustion without and with ozone addition versus ϕ

Further experiments were performed under oxygen-enriched conditions combusting LCV gases with composition of CH_4/CO_2-25%/75% and CH_4/CO_2-20%/80%. According to determined flame lift-off heights, the supply of oxygen-enriched air (40 vol% O_2 in N_2) improved flame stability of LCV gases ranging ϕ from 0.625 to 0.9. At higher fuel equivalence ratio the flame became unstable and lifted up. Even though the combustion was performed with oxygen-enriched air, the ozone addition improved the flame stability and the flame lift-off was reduced by 9-33% and by 10-11% respectively for CH_4/CO_2-25%/75% and CH_4/CO_2-20%/80%. It was considered that an increased O_2 concentration in air led to higher production of ozone, which in turn improved the combustion of LCV gases.

3.3. *Ozone effect on CO and NOx emissions*

The acquired emissions of NO_x and CO at different ϕ are presented in Fig. 5 and Fig. 6, respectively. During SNG combustion, the addition of ozone reduced NO_x emissions by 6–2 mg/m^3 at ϕ values of 0.63-0.83 and further increase of ϕ led to near equal NOx concentrations. During ozone-assisted combustion of LCV gas A, the NO_x emissions reduction was higher only ranging ϕ from 0.8 to 1.0 compared to the previous case. However, results obtained in the low swirl burners showed insignificant or even negative effect of the ozone addition on reduction of NO_x emissions. NOx emissions were nearly identical during combustion of CH_4/CO_2-25%/75% with ozone and without. Besides, the obtained emissions were highest compared to other combustion cases. This could be caused due to oxygen-enriched air which increases the flame temperature and thermal NOx formation intensifies. Though, comparable small emissions were determined combusting LCV gas C (CH_4/CO_2-20%/80%), but the ozone addition led to increased emissions by 4-8 mg/m^3.

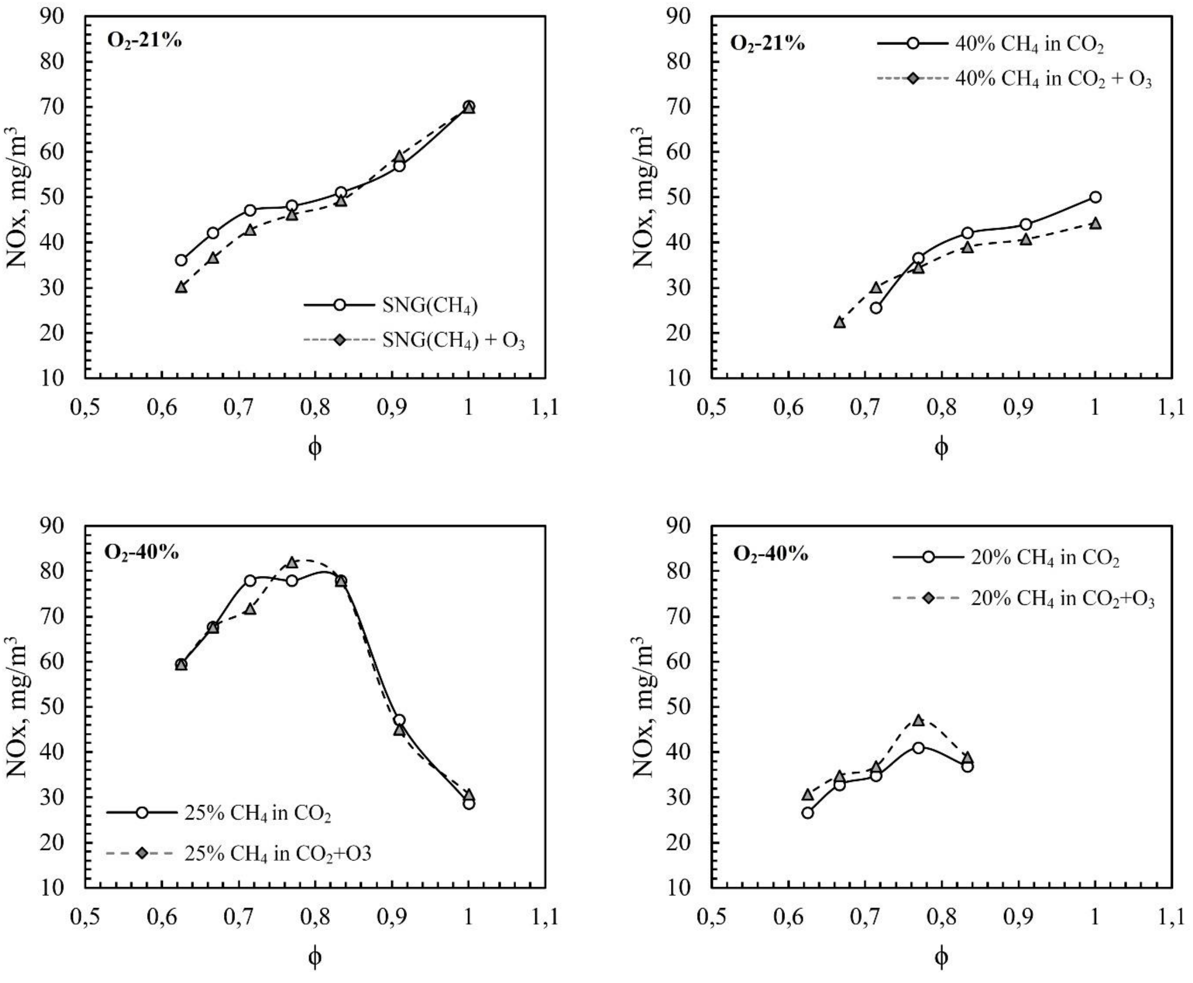

Figure 5. NO_x emissions at different fuel equivalence ratios ϕ

More encouraging results were observed analysing CO emissions. During ozone-assisted combustion of SNG, CO was reduced by approximately 50 mg/m^3 and 400 mg/m^3 for values ϕ=0.66 and 0.63 (see Fig. 6). In the case of waste gases A, the O_3 effect on CO was obtained similar and CO emissions decreased by 80-20 mg/m^3 with ϕ ranging from 0.71 to 1.0, respectively (Fig. 6). The tendency of CO emissions was a bit different performing combustion experiments in the low swirl burner under oxygen enriched conditions. During the waste gas B combustion, CO emissions were decreased by 20-40 mg/m^3 increasing ϕ from 0.625 to 0.833 due to the ozone addition. At higher ϕ values incomplete combustion occurred and CO emissions increased drastically and the ozone effect was insignificant. In case of waste gas C, CO emissions were reduced by 70-200 mg/m^3 and by 200-300 mg/m^3 respectively changing ϕ from 0.625 to 0.66 and from 0.714 to 0.77 due to the ozone addition.

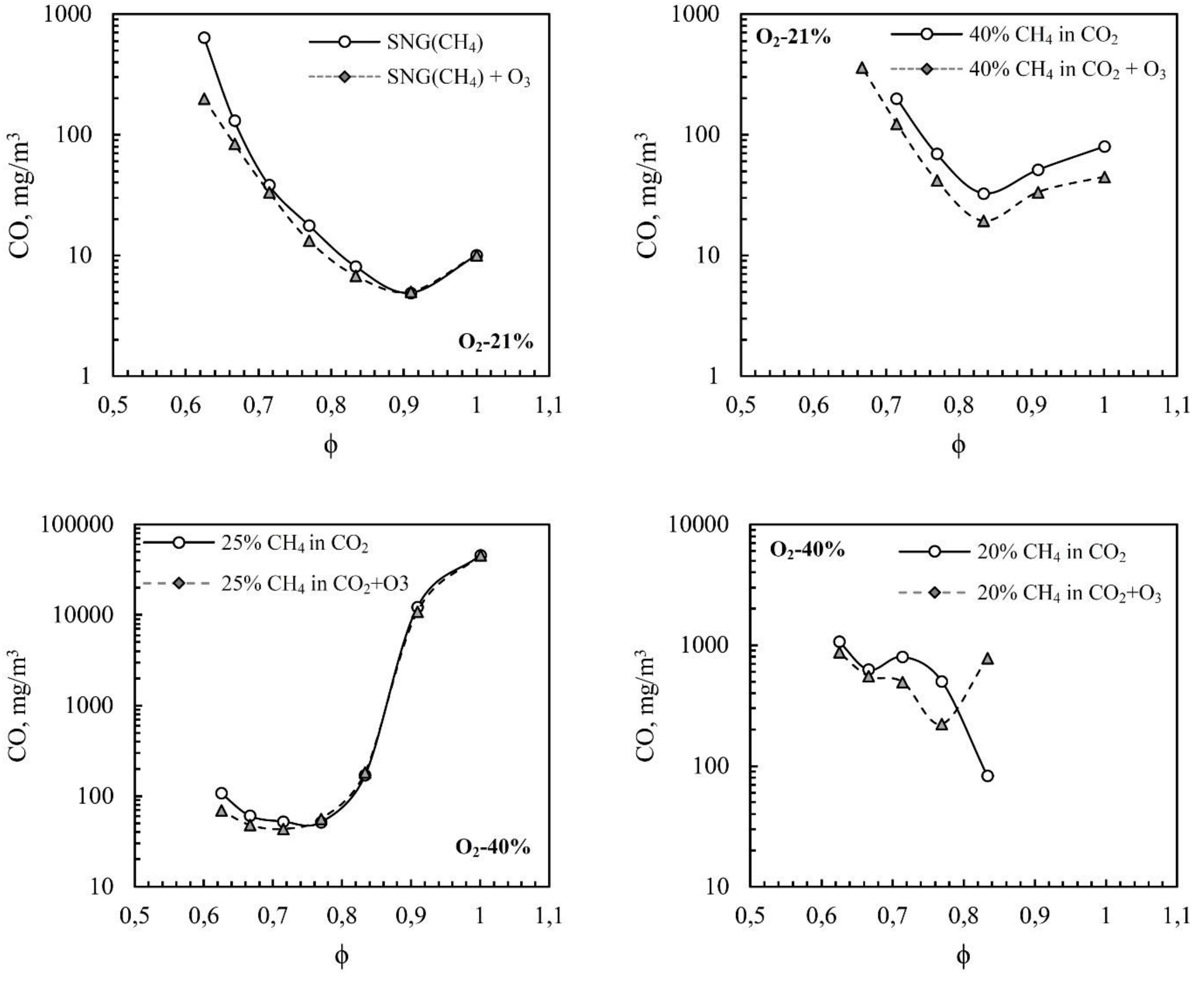

Figure 6. CO emissions at different fuel equivalence ratios ϕ

4. Conclusions

The effect of ozone on combustion efficiency was investigated in two burners (the flat flame and low swirl burner) in wide range of fuel equivalence ratios using different compositions of gases (SNG, waste gases with low calorific value). The obtained results showed promising results on reducing the flame lift-off for low calorific value gases. An addition of O_3 reduced the flame lift-off by 20-40% and by 40-70% in the case of SNG and in the case of LCV gases, respectively. The ozone effect was weaker on LCV gases with CH_4 concentration of 20-25 vol% supplying oxygen-enriched air and the flame lift-off was reduced only from 33% to 9%. The ozone enrichment of SNG combustion reduced NOx and CO emissions by 2-7 mg/m^3 and by 200-4 mg/m^3 increasing ϕ from 0.625 to 0.833, respectively, but NOx concentrations increased by 2 mg/m^3 at higher ϕ values. The tendency on NOx increase due to the ozone addition was determined with LCV gases as well, but the ozone effect on reducing the CO emissions was more intensive. The highest effect on CO decrease was observed during ozone assisted combustion of CH_4-20%/CO_2-80% mixture. CO emissions decreased by 70-300 mg/m^3.

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