EFFECT OF CO_2 DILUTION ON PREMIXED H_2/CO/CNG BLENDING SYNTHESIS GAS FLAMES

Ilker YILMAZ[1], Harun YILMAZ[2], Omer CAM[1], Mehmet Ali SOYTURK[1]
[1]Department of Airframes and Powerplants, Faculty of Aeronautics and Astronautics, Erciyes University, Kayseri, 38039 Turkey
[2]Department of Airframes and Powerplants, Civil Aviation College, Erzincan University, Erzincan, 24100, Turkey
Corresponding author: Ilker YILMAZ, e-mail: iyilmaz@erciyes.edu.tr

REFERENCE NO

COMB-05

Keywords:
Synthetic gas, Combustion, Emission, inert dilution.

ABSTRACT

In this study, effects of CO_2 dilution on combustion and emission behavior of premixed H_2/CO/CNG blending synthetic gas flames were experimentally investigated in a swirl stabilized, laboratory scale combustor. First, mixture of H_2/CO/CNG was tested as a baseline case (at a constant swirl number and equivalence ratio) and then varying amount of CO_2 (from 0 to 20% by volume, increased by 5%) was added to each gas mixture by keeping H_2/CO ratio at constant value of 1.0. Lastly, mixtures of H_2/CO/CNG/CO_2 were achieved and tested under the same physical and atmospheric conditions. Combustion behavior of such mixtures was evaluated with respect to measured axial temperature values. Moreover, emission behavior was analyzed by means of emitted CO, CO_2 and NO_x levels, which are detected by a flue gas analyzer. Results of this study revealed that thermal and radiation effect of CO2 dilution dominates temperature distribution, while chemical effect of inert dilution governs pollutant emissions.

1. INTRODUCTION

CO_2 dilution effects flame behavior by altering specific heat and adiabatic flame temperature (heat capacity of CO_2 is 1.68 times higher than that of N_2), chemical kinetics (high CO_2 levels increase backward reaction rates and thus reduce CO oxidation and H atom yield), radiation efficiency (since CO_2 is an infrared active molecule, CO_2 dilution results with additional heat loss) and diffusion process (CO_2 diffuses slower than N_2) [1-3]. Practically, CO_2 dilution is usually referred to obtain low flame temperatures and in turn low pollutant emissions [3]. Inert dilution can be applied to either fuel stream or air stream and mostly studied diluents are H_2O, CO_2 and N_2. There can be found a large literature investigation on the subject of inert dilution of both conventional and alternative fuels [5-9]. Since synthetic gases can contain diluents such as N_2, CO_2 and H_2O depending on the feedstock, gasifying agent and reactor atmosphere, effect of diluents on synthetic gas combustion and emission behavior is a key aspect in effective utilization of these gases.

Lee et al. performed combustion tests with a GE7EA gas turbine model combustor. They used mixtures of H_2/CO/N_2/CO_2/steam as fuel and evaluated the effects of inert constituents. For this purpose, they varied the amount of diluents and measured flame temperatures, NO_x and CO emissions, and observed flame shapes and flame instabilities. They reported that NO_x emission decreases with inert dilution and this decrement logarithmically correlates with diluent heat capacity. It was also shown that efficiency requirements (complete combustion, low CO emissions, and no flashback and blowout occurrence in the entire operating range of the combustor) can satisfactorily be met, indicating applicability of inert dilution for syngas turbine [10].

Tian et al. conducted experimental studies on N_2 diluted non premixed synthetic gas flames at different air humidification conditions to examine effectiveness of N_2 dilution on NO_x reduction. They also carried out OH-PLIF measurements to assess flame structure and concluded that as the amount of N_2 dilution increases, NO_x emissions reduce from 27 ppm to 12 ppm and reaction zone contracts in the case of dry air condition; when air is humidified (D_{steam}-flow rate of steam/flow rate of dry air = 25%), NO_x emissions further reduces to 10 ppm and emissions of CO remain low; further increment (D_{steam} = 50%)

slightly decreases NO_x emissions but largely increases CO emissions (incomplete combustion) [11].

Li et al. simulated CO_2 and H_2O diluted syngas flames in partially premixed configuration using LES (Large Eddy Simulation)-LEM (Linear Eddy Model) technique. Results of their study revealed that fuel side CO_2 dilution considerably reduces flame temperature, thermal effect of CO2 dilution overweighs H_2O effect since chemical effect of CO_2 tends to decrease H and OH radical concentrations, vice versa for H_2O (H_2O dissociation). Zhang et al. both experimentally and numerically studied effects of inert dilution (N_2, CO_2) on evolution and quenching of hydrogen/carbon monoxide mixture synthetic gas flames. They measured extinction stretch rates in counter-flow flame configuration and achieved laminar flame speed values from literature. Numerical and experimental data showed a good consistency and showed that CO_2 has a more significant effect (decreasing effect), on flame evolution and quenching than N_2, thermal effect of inert dilution dominates this decrement, chemical effect of CO_2 dilution is more effective on decreasing extinction strain rate than laminar flame speed, and N_2 alters these properties only thermally [13]. Xu et al. investigated effects of substitution of N_2 (up to 30% in volume basis) in air (oxygen amount kept constant) with CO_2 and H_2O on the shape and structure of the H_2/CO diffusion flames. For temperature measurements and flame visualization (to detect flame height and radius), they used thermocouples and OH* - chemiluminescence method, respectively. They also modelled tested flames with a detailed chemical reaction mechanism developed for synthetic gas combustion. They reported that both CO2 and H_2O substitution of N_2 reduces maximum flame temperature (both thermal and radiative effect) and differently affects centerline temperature values. The chemical and diffusion effect of CO_2 (first increases, then decreases) and H_2O (vice versa) on flame temperature are different from each other. While the amount of OH radical increases via $H + O_2 = O + OH$ reaction with H_2O increment (shorter and narrower flames with high burning intensity arise), CO_2 suppresses these reactions (long and thick flames) [3]. Inert dilution also alters flame stability characteristics in an undesirable manner and this situation necessitates detailed investigation of stability aspect of diluent effect for reliable operation of synthetic gas fed burners. Li et al. built an experimental setup to study lean blowout limits of premixed synthetic gas flames at different diluent ratios. It was found that as the amount of diluent increases, lean blowout limits (equivalence ratio at which blowout occurs) increase. Besides the thermal effect of N_2 and CO_2 dilution, chemical kinetic effect of CO_2 poses several challenges by means of flame stability. Lastly, they proposed a correlation (utilizing Damköhler number and normalized flame temperature) to predict lean blowout limits of diluted synthetic gas flames and reported that such correlation can be used in a large scale of dilution [14].

In this study, effects of CO_2 dilution on combustion and emission behavior of premixed H_2/CO/CNG blending synthetic gas flames were investigated in a premixed laboratory scale swirl stabilized combustor. First, mixture of H_2/CO/CNG was tested as a baseline case then keeping H_2/CO ratio constant, respective mixture was diluted with 5% CO_2 by volume and tested at the same equivalence ratio (0.6), swirl number (0.2) and thermal power (3 kW). The amount of CO2 in gas mixture was gradually increased from 5 to 20% at the intervals of 5% and effect of CO_2 dilution on combustion and emission behavior was evaluated via examining axial temperature and radial (also axial) emission measurement values.

2. EXPERIMENTAL SETUP

2.1. Burner

In Fig. 1, designed and manufactured burner can be seen. This burner was designed to operate in premixed mode and at thermal powers up to 10 kW. It consists of an air/fuel inlet, two slots for photodiode and pressure sensor installation (not used in this study) and a swirl housing that contains swirl generator. The modular structure of burner allows swirl generator to be changed without disassembling burner. Operating pressure of the burner is 20 mbar.

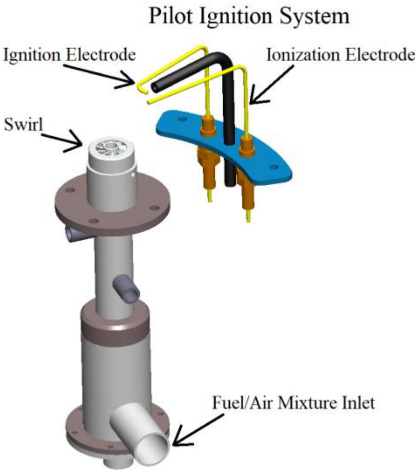


Fig. 1. Solid model of the burner.

Each constituent of synthetic gas mixture is supplied from a gas tank. Depending on the gas composition and thermal power, each gas is metered by a digital mass flow controller (MKS-GE50A). A six channel vacuum system controller (MKS-946 Series) governs digital mass flow controller and desired mass flow rate is adjusted through. Mass flow controllers require relatively high gas pressures to properly operate, this is why these equipment are installed right after pressure regulators (reduces pressure from 200-300 bar to 0-1.5 bar) on the gas tanks. Other pressure regulators and manometers are assembled to gas supply lines to further reduce pressure (since burner operate at 20 mbar) and monitor pressure value, respectively. Mechanical flow meters are mounted for control purposes.

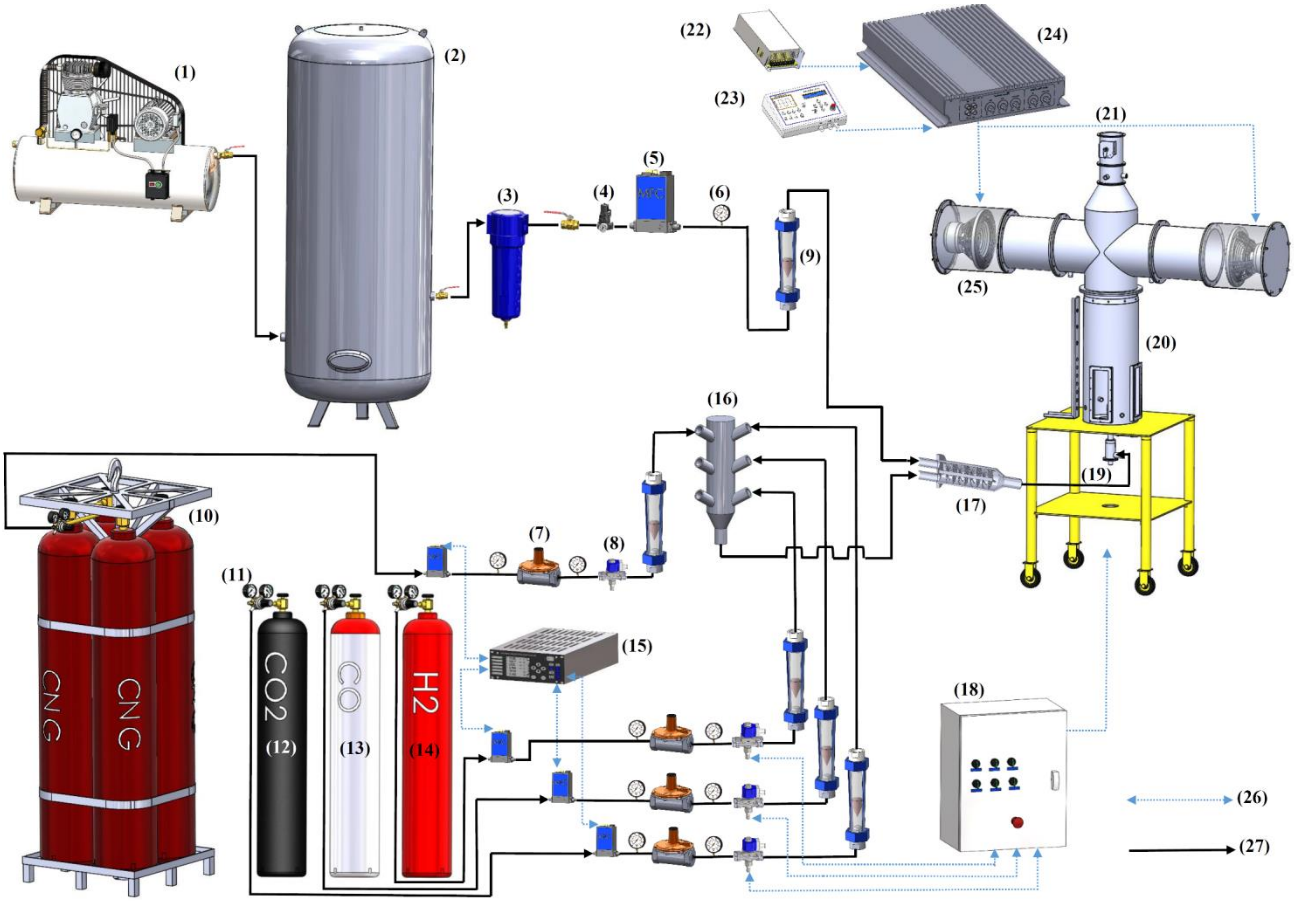


Fig. 2. Layout of the overall combustion system.

1. **Air compressor**
2. **External air tank**
3. **Filter (for steam and oil removal)**
4. **Pressure regulator**
5. **Mass flow controller**
6. **Manometer**
7. **Pressure regulator**
8. **Solenoid valve**
9. **Float type flowmeter**
10. **CNG tank**
11. **Pressure regulator**
12. CO_2 tank
13. CO tank
14. H_2 tank
15. Vacuum system controller
16. Gas collector
17. Fuel/air pre-mixer
18. Control Panel
19. Burner
20. Combustion chamber
21. Flue
22. Electrical connections
23. Gas supply lines

A piston type air compressor with 1500 m^3 storage capacity provides combustion air needed and the amount of air (equivalence ratio dependent) is regulated by a digital mass flow controller (MKS-1579A). Air supply line also consists of a manometer and a mechanical flowmeter. Both gas supply lines and air supply line are equipped with solenoid valves which opens or closes these lines depending on the flame presence, which is detected by an ionization electrode. The schematic view of the overall combustion system and experimental equipment are demonstrated in Fig. 2. All fuel gases and combustion air are delivered to a gas collector then fully mixed in a static pre-mixer, which is equipped with a mechanical flashback arrestor, before entering the burner.

The combustor has 1650 mm length, 330 mm outer and 320 mm inner diameter (5 mm wall thickness). Axial and radial temperature and emission measurements are performed via numerous slots located on the walls, side arms and chimney of the combustor. There is also a cooling slot in which air is fed through an external air turbine around the circumference of the combustor (not added to combustion air). Tempered glasses which are located in front and side of the combustor make flame optically accessible and give chance to change swirl vane without removing burner. All metal components of the combustor are made of the stainless steel.

To be able to ignite low calorific value synthetic gas, a pilot ignition system is integrated with the combustor. This system comprises a fuel tank (LPG), fuel supply line, solenoid valve, a radial air fan and a burner.
Other equipment not addressed here were installed to combustion system for dynamic and static (flashback, blowout) flame stability studies.

2.2. Measurement Equipment

Temperature measurements are performed with ceramic coated NiCr-Ni alloy K (Range -200–1200C°) and Pt%13Rh-Pt alloy B-type (Range 0–1800C°) thermocouples. Measured temperature values were corrected considering radiation losses and were found to be 3-37 K lower than actual values. Data obtained from these thermocouples are converted to quantifiable parametric information via a 28 channel data logger (Expert Key 200L) that has 100 kS/s sampling rate, and stored in a computer. Tuning of sampling rate of each channel, selection of channel input value (voltage, ampere etc.) and monitoring measured values during experiments are performed with ProfiSignal Go software.
Emission measurements are performed via a portable flue gas analyser (NOVA Plus RCU). Typical accuracy of the analyzer is: for O_2 ± 0.2%; for CO (in the range of 0-4000 ppm) ± 5% (or ±10ppm – whichever is higher), >4000 ppm ± 10%; for NO (in the range of 0-1000 ppm) 5% (or ±5ppm - whichever is higher), >1000 ppm 10%; for NO_2 (in the range of 0-200 ppm) 5% (or ±5ppm - whichever is higher), >200 ppm 10%; for SO_2 (in the range of 0-2000 ppm) 5% (or ±10ppm - whichever is higher), >2000 ppm 10%; for CO_2 ±0.3%. In addition to these measurements mentioned above, the amount of each gas is metered by digital mass flow controllers that have typical

accuracy of: for gas supply lines ± 1% of set point for 20 to 100% of full scale, ± 0.2% of set point for 0 to 20% of full scale; for air supply line ± 1% of full scale.

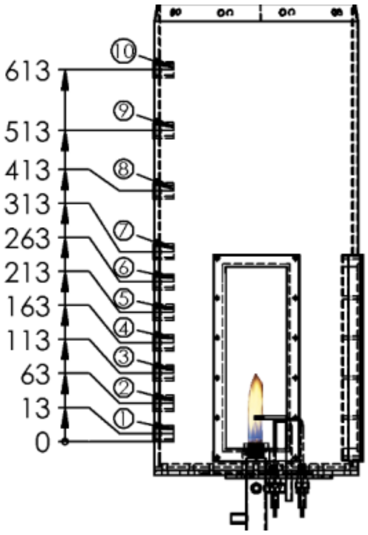


Fig. 3. Measuring ports.

2.3. Operating Conditions and Gas Compositions

Thermal power of the combustor was set as 3 kW. Mass flow rate of each gas and combustion air were determined depending on the thermal power and gas composition, and equivalence ratio, respectively. Geometric swirl number (0.2) and equivalence ratio (0.6) were kept constant. Fuel gases and air were premixed at room temperature in a pre-mixer before entering the burner. All experiments have been performed at the local atmospheric conditions of city of Kayseri, Turkey. Lastly, combustion takes place at 20 mbar (gauge pressure) and room temperature.

As a baseline case, mixture of H_2/CO/CNG with constant H_2/CO ratio (1.0) and CNG amount (by volume) was specified and combusted. CNG is added to each gas mixture for flame stability issues and to represent CH_4 (a common constituent of synthetic gas mixtures). Later on, each gas mixture was diluted with varying amount of CO_2 (from 0 to 20% by volume, increased by 5%) and tested under the same physical and atmospheric conditions to evaluate effect of inert dilution on synthetic gas combustion. Tested gas mixtures are tabulated in Table 1.

Table 1. Tested gas mixtures.

Syngas	**CNG**	**H_2**	**CO**	**CO_2**
SG 1	20	40	40	-
SG 2	20	37.5	37.5	5
SG 3	20	35	65	10
SG 4	20	32.5	32.5	15
SG 5	20	30	30	20

3. RESULTS AND DISCUSSIONS

Operability issues in combustion systems are primarily controlled by dynamic and static (blowout, flashback, and liftoff) behaviors of the flame. To guarantee a reliable operation and to be able to make synthetic gases applicable in existing burners with minimal structural modifications, synthetic gas combustion characteristics must be examined and elaborated. For instance, H_2 rich synthetic gas mixtures are associated with high flashback propensity, while CO rich synthetic gas mixtures generate unstable flames (because of the low H atom concentration). Furthermore, inert constituents of synthetic gases pose several challenges for flame propagation and extinction characteristics [15, 16]. Since fuel composition effect is out of scope of this study, we will only address effect of inert dilution on fundamental flame characteristics of synthetic gases such as flame temperature and emissions. In Fig. 4, axial temperature profiles of SG 1, SG 2, SG 3, SG 4 and SG5 mixtures at the equivalence ratio (ER) and swirl number (SN) of 0.6 and 0.2, respectively are illustrated.

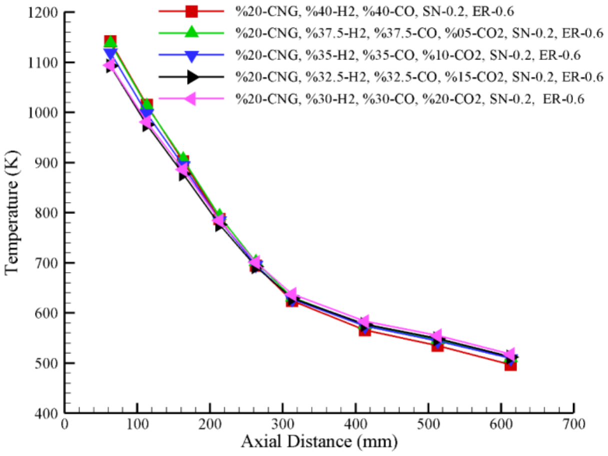


Fig. 4. Axial temperature distributions.

Temperature profiles of tested mixtures show a good agreement in terms of trend and value. At burner outlet, temperature values for SG 1, SG 2, SG 3, SG 4 and SG5 mixtures are 1142.02, 1138.15, 1119.3, 1093.14 and 1094.03 K, respectively. As the CO_2 amount in synthetic gas mixture increases, flame temperature decreases except for the SG 6 mixture (a small increase-less than 1 K). Thermal and radiation effect of CO_2 dilution causes flame temperature to decrease but this decrement is not that distinct. The difference between peak temperature values of SG 1 and SG 5 mixtures is 47.99 K. With increased axial distance, temperature values further decrease but between the axial positions of 200-300 mm away from burner outlet, temperature profiles become in line with each other.

At the outlet zone of the combustor, exit temperature values are 496.11, 510.25, 509.14, 511.84 and 517.25 K for SG 1, SG 2, SG 3, SG 4 and SG5 mixtures, respectively. This situation indicates that CO_2 amount positively affects exit temperature values. Reaction zone extends in axial and radial directions in the case of CO_2 addition (heating value and thus adiabatic flame temperature decrement reduces flame speed and reactivity). CO_2 presence increases rate of reaction of $CO_2 + H \rightarrow CO + H$ and in turn, H atom concentration decreases by suppressing main chain branching reaction of $H + O_2 \rightarrow O + OH$. As a consequence, concentrations of radicals such as O, H and OH decrease and longer flames with lower burning intensity occur. [15]. Higher exit temperature values of high CO_2 diluted mixtures (due to the longer flame lengths) are attributed to this phenomenon.

To analyze effect of inert dilution in more detail, temperature profiles of SG 1, SG3 and SG 5 mixtures are demonstrated in Fig. 5. It can clearly be inferred from this figure that temperature distribution gets higher value with decreasing CO_2 amount in beginning and mid sections of the combustor but towards the outlet of the combustor, this temperature decrement retards with increasing CO_2 amount, and SG 6 mixture forms the highest and most uniform temperature distribution.

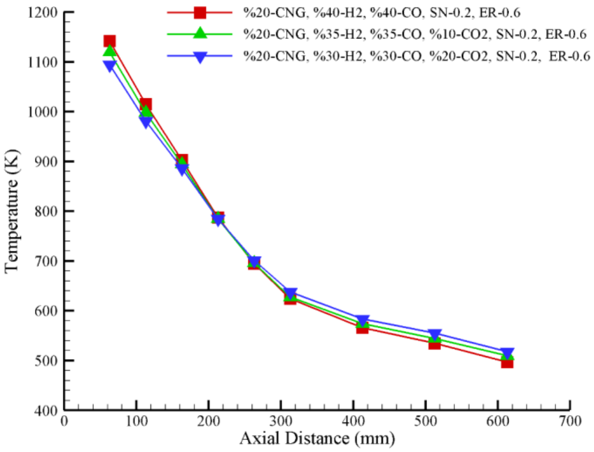
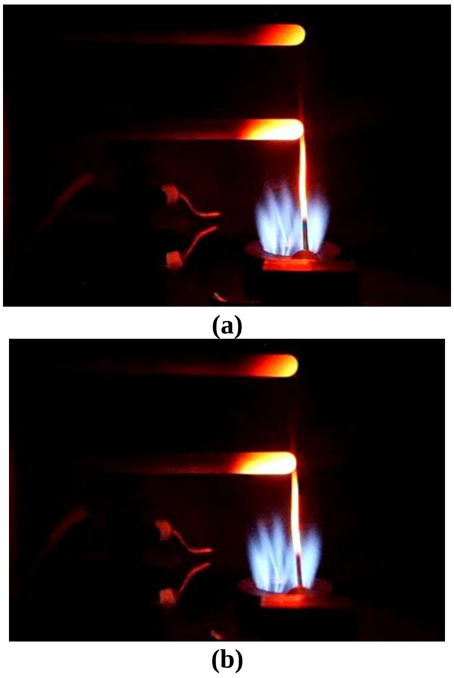


Fig. 5. Axial temperature distributions.

To examine effect of inert dilution on flame shape, instantaneous flame images (with the same focal length and exposure time) of SG 1, SG 2, SG 3, SG 4 and SG5 mixtures were utilized and illustrated in Fig. 6. For all mixtures tested, flames firmly attach both inner and outer annulus of the burner, indicating that inert dilution do not significantly change flame anchoring position. As the CO_2 content in synthetic gas mixture increases, flames become more bluish, and elongate as a result of reduced reaction kinetics.



(a)

(b)

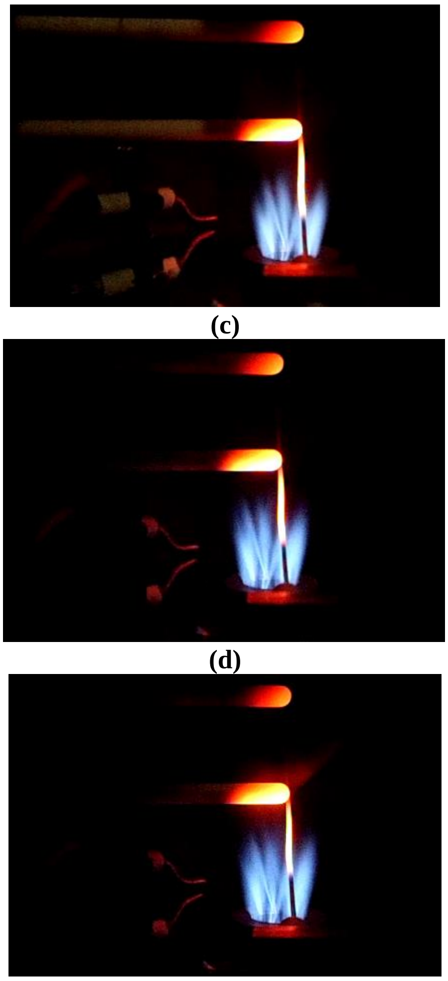


Fig. 6. Instantaneous flame images of.
a) SG 1, b) SG 2, c) SG 3, d) SG 4, e) SG 5 mixture.

Sampling of post combustion gases was conducted at 3 different radials (5, 10 and 16 mm away from combustor centerline) and axial (63, 163 and 263 mm away from burner outlet) positions for at least one minute. Due to the high temperature values in and near the flame region, species are highly reactive and chemical reactions still progress. As a consequence, we could not attain stable emission values in this region and emission readings was not started to be taken from centerline of the flame. In Fig. 7, CO_2 concentration profiles in percentage of total exhaust gases can be seen.

As mentioned before, mixture's reactivity reduces with CO_2 addition (hence burning rate and flame temperature). At the axial position of 63 mm, CO_2 emission of SG1 mixture is the highest due to the highest H_2 content (or in the absence of CO_2-high reactivity) and likewise other mixtures, CO_2 emissions decrease except for the SG 5 mixture as the CO_2 amount in synthetic gas mixture increases. This opposite trend of such mixture is because of the highest CO_2 content.

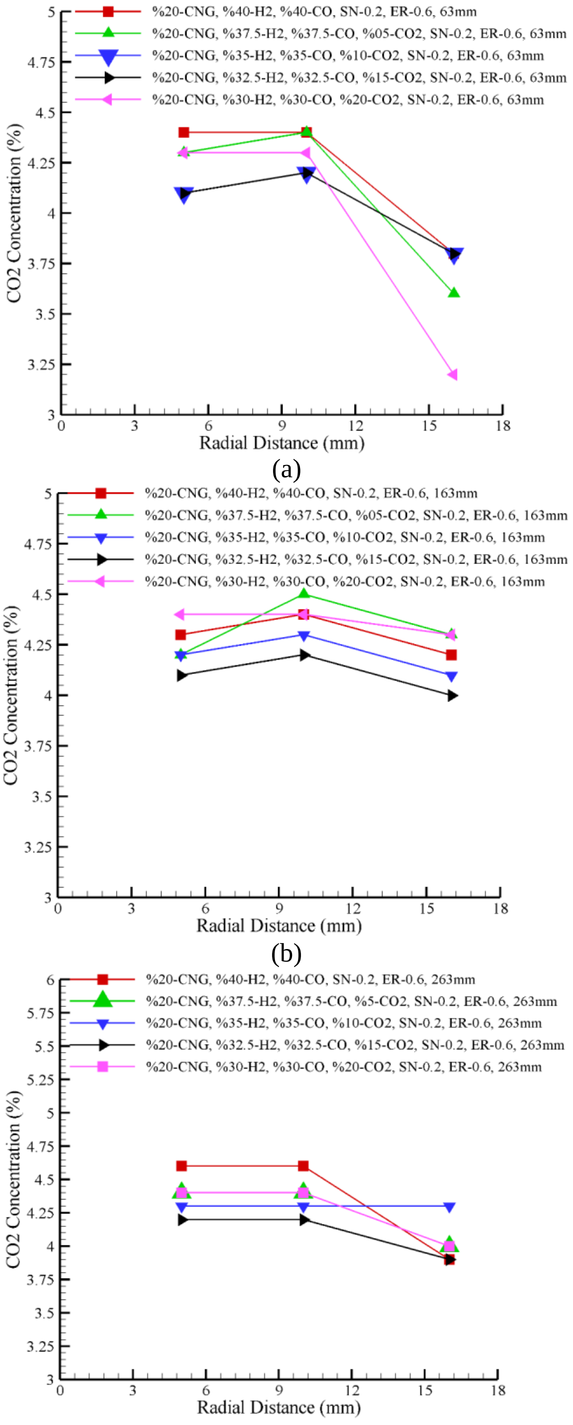
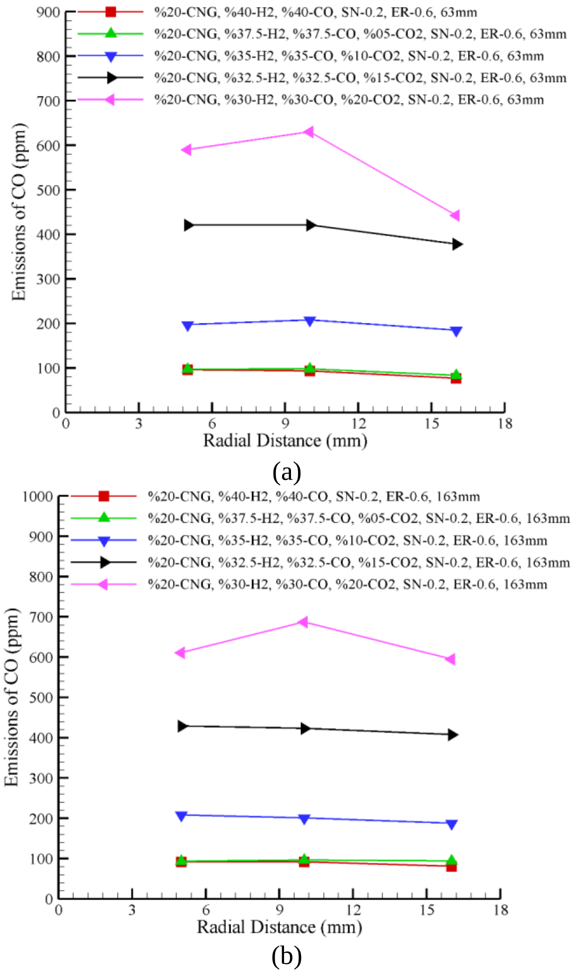


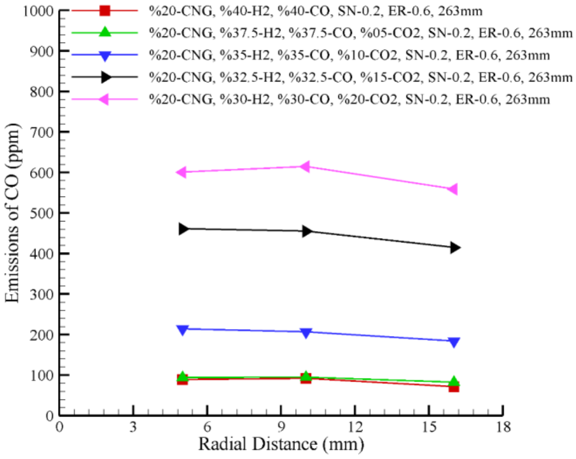
Fig. 7. CO2 concentration profiles.
Axial positions: a) 63 mm, b)163 mm, c) 263 mm.

At the axial positions of 63 and 163 mm, CO_2 emissions of most mixtures barely increases between radial positions of 5 and 10 mm. At 263 mm, emission values do not change in the same radial range. However, CO_2 emissions decrease with further increment in radial distance (from 10 to 16 mm) except for SG 3 mixture at 263 mm. Lastly, it can be concluded that CO_2 emission levels of tested mixtures slightly alter with axial position. CO_2 emissions either stabilize, decrease or increase but this decrement or increment is not more than 0.2%.



(a)

(b)



(c)

Fig. 8. CO profiles. Axial positions a) 63 mm, b)163 mm, c) 263 mm.

CO is an incomplete combustion product and oxidation rate of CO to CO_2 improves at high temperatures. Since all experiments were conducted at fuel lean equivalence ratio (0.6), emission levels of CO is relatively high and were found to significantly increase with increasing CO_2 amount, (Fig. 8) i.e. with decreased reactivity, irrespective of the axial or radial distance. Besides, CO profile of SG 1 mixture is nearly in line with that of SG 2 mixture at all radial and axial distances tested.

Because measured NOx values were not more than 2-5 ppm, it was not addressed in the extent of this paper.

4. CONCLUSIONS

In this study, effects of inert dilution (CO_2) on combustion and emission behavior of premixed CNG/H_2/CO mixture flames was experimentally investigated in a swirl stabilized combustor. CO_2 amount in gas mixtures was gradually increased from 5 to 20 % (by volume) at intervals of 5% and effects of dilution were analyzed by examining temperature, CO and CO_2 concentration profiles.

Results derived from this study are:

- Radiation effect of CO_2 dilution causes flame temperature to decrease but this decrement is not more than 48 K.

- Conversion rate of CO to CO_2 favors at high temperatures and with more reactive mixtures.

- Since inert dilution do not significantly change temperature distribution throughout the combustor for all mixtures tested, it is concluded that chemical effect of inert dilution dominates pollutant emissions.

Acknowledgements
We would like to thank the Scientific and Technological Research Council of Turkey (TÜBİTAK-MAG-215M821) for its financial support.

References

[1] Williams, T. C., Shaddix, C. R., Schefer, R. W., Effect of syngas composition and CO_2-diluted oxygen on performance of a premixed swirl-stabilized combustor, *Combustion Science and Technology*, Vol. 180, No. 1, 2007, pp. 64-88.

[2] Lieuwen, T., Yang, V., Yetter, R., Synthesis gas combustion: fundamentals and applications, CRC Press, 2009

[3] Xu, H., Liu, F., Sun, S., Zhao, Y., Meng, S., Tang, W., Effects of H_2O and CO_2 diluted oxidizer on the structure and shape of laminar coflow syngas diffusion flames, *Combustion and Flame*, Vol. 177, 2017, pp. 67-78.

[4] Wang, S., Ji, C., Zhang, B., Cong, X., Liu, X., Effect of CO_2 dilution on combustion and emissions characteristics of the hydrogen-enriched gasoline engine, *Energy*, Vol. 96, 2016, pp. 118-126.

[5] Zhang, B., Shen, X., Pang, L., Effects of argon/nitrogen dilution on explosion and combustion characteristics of dimethyl ether–air mixtures, *Fuel*, Vol. 159, 2015, pp. 646-652.

[6] Wang, Z., Zuo, H., Liu, Z., Li, W., Dou, H., Impact of N 2 dilution on combustion and emissions in a spark ignition CNG engine, *Energy Conversion and Management*, Vol. 85, 2014, pp. 354-360.

[7] Mitu, M., Giurcan, V., Razus, D., Oancea, D., Inert gas influence on the laminar burning velocity of methane-air mixtures, *Journal of hazardous materials*, Vol. 321, 2017, pp. 440-448.

[8] Duan, J., Liu, F., Laminar combustion characteristics and mechanism of hydrogen/air mixture diluted with N_2+ H_2O, *International Journal of Hydrogen Energy*, Vol. 42, No. 7, 2017, pp. 4501-4507.

[9] Allouis, C., Chiariello, F., Effect on O 2 enrichment and CO 2 dilution on rapeseed oil combustion in a stationary burner, *Experimental Thermal and Fluid Science*, Vol. 73, 2016, pp. 49-55.

[10] Lee, M. C., Seo, S. B., Yoon, J., Kim, M., Yoon, Y., Experimental study on the effect of N_2, CO_2, and steam dilution on the combustion performance of H_2 and CO synthetic gas in an industrial gas turbine, *Fuel*, Vol. 102, 2012, pp. 431-438.

[11] Tian, Y., Zang, S., Ge, B., Experimental investigation on the combustion performance of N_2 dilution in syngas non-premix combustion in humid air conditions, *Applied Thermal Engineering*, Vol. 107, 2016, pp. 560-564.

[12] Li, S., Li, S., Mira, D., Zhu, M., Jiang, X. Investigation of dilution effects on partially premixed swirling syngas flames using a LES-LEM approach, *Journal of the Energy Institute*, 2017.

[13] Zhang, Y., Shen, W., Zhang, H., Wu, Y., Lu, J., Effects of inert dilution on the propagation and extinction of lean premixed syngas/air flames, *Fuel*, Vol. 157, 2015, pp. 115-121.

[14] Li, S., Zhang, X., Zhong, D., Weng, F., Li, S., Zhu, M., Effects of inert dilution on the lean blowout characteristics of syngas flames, *International Journal of Hydrogen Energy*, Vol. 41, No. 21, 2016, pp. 9075-9086.

[15] Samiran, N. A., Ng, J. H., Jaafar, M. N. M., Valera-Medina, A., Chong, C. T., Swirl stability and emission characteristics of CO-enriched syngas/air flame in a premixed swirl burner, *Process Safety and Environmental Protection*, 2017.

[16] Valera-Medina, A., Vigueras-Zuniga, M. O., Baej, H., Syred, N., Chong, C. T., Bowen, P. J., Outlet geometrical impacts on blowoff effects when using various syngas mixtures in swirling flows, *Applied Energy, 2017.*