

Recirculation of burned gas effects on NO_x emissions from natural gas-hydrogen-oxygen flames in a burner with separated jets

Sébastien Yon^{1,*}, Jean-Charles Sautet¹, Toufik Boushaki²

1: CORIA UMR 6614 – CNRS - Université et INSA de Rouen, Saint Etienne du Rouvray. France

2: ICARE – CNRS – Avenue de la Recherche Scientifique Orléans. France

* corresponding author: sebastien.yon@coria.fr

Abstract Strict regulations of NO_x emission standards have resulted in an optimized performance of combustion chambers. The present paper describes the effects of burned gas recirculation on NO_x emissions from hythane-oxygen flames in a separated jet burner. NO_x emissions from a burner with separated jets and the effects of the size of the burned gases in recirculation in such a burner have not yet been studied.

Burners with separated fuel and oxidizer jets permit a high dilution of reactants and a large recirculation zone of burned gases in the combustion chamber, which favors the decrease of nitrogen oxide emissions.

In this study, the oxy-fuel burner was equipped with two nozzles: the first nozzle supplied a hythane flow (a mixture of natural gas and hydrogen) and the second nozzle supplied pure oxygen. The hydrogen content in the fuel varied from 0% to 20% in volume. The influence of the distance between the nozzles (12 to 100 mm) and the global equivalence ratio of the mixture were analyzed.

Measurements of combustion product concentrations were carried out at the combustion chamber exit using a water-cooled probe and a NO_x analyzer. The velocity fields and the size of the recirculation zone were determined by the Particle Image Velocimetry technique (PIV) in the reacting flow.

The results showed that an increase in size of the recirculation zone leads to a decrease in NO_x emissions (up to 96%), and that NO_x emissions are inversely proportional to the measured surface area of the burned gas recirculation zone. Due to the reaction between recirculated CO₂ and thermal NO and its chemical effects, NO_x emissions decrease in lean combustion because of the destruction of NO.

In oxy-fuel combustion occurring in burners with two separated jets, NO_x emissions depend on the size and composition of the recirculation zone of the burned gases, which are central to the goal of limiting NO_x emissions at the exit of the fumes.

1. Introduction

The reduction of nitrogen oxide emissions has been extensively investigated in combustion to answer to the more stringent NO_x emission standards. Numerous theoretical and technological strategies have been studied to reduce NO_x emissions as the effect of the flame temperature (Nada and Aggarwal 2004), the influence of the flame strain (Kim and Yoon 2007), or the flame radiation (Røkke et al. 1992). Many studies dealing with NO_x emission have focused on diffusion flames (Hwang et al. 2008), however, this kind of flame can become extremely interesting for reduction of NO_x emissions if the combustion is linked up to recirculation of burned gases in the combustion chamber. Recent attempts using gas recirculation have shown the efficiency of the recirculation of combustion products to reduce NO_x emissions (Nada et al. 2008). With this technique, the burned gases are recirculated inside the combustion chamber and introduced into the flame, allowing to radically reduce NO_x emissions with compact equipment and a low cost (Shinomori et al. 2011).

A new generation of burner with separated injections of fuel and oxidizer presents attractive perspectives in term of limitation of nitrogen oxide emissions. In these burners, the dilution effect linked to the nozzles separation increases the residence time of the combustion products and favours the recirculation of burned gases (Boushaki et al. 2009). Previous works have proved that the separation of the nozzles is an important factor in the NO_x production. Indeed, as mentioned by Dugué and Riffart (2006), increasing distance between jets decreases drastically the NO_x

emissions. The distance between the nozzles influences the flow dynamic acting on the distance of the beginning of the mixing point (the region where the jets start to interact and mix together). The more the nozzles are moved away, the more the jets interact upstream in the flow favouring the burned gases dilution (Salentey 2002). Furthermore, studies in free turbulent jet have shown that the space between the nozzles increases the size and the force of recirculation zone (Zhakatakev 1994). Previous works have studied the behavior of flow from burner with two jets, with a focus on flame lift-off heights and mixing zones for a natural gas-H₂-O₂ flame and have shown that the hydrogen addition reduces the size of the recirculation zone and therefore improves the dilution of the flame by the combustion products (Yon and Sautet 2012). Mixture of natural gas and hydrogen called hythane has the advantage to sensibly modify the properties of the fuel preserving the distribution installation. Due to hydrogen properties in combustion, that is to say high molecular diffusivity, wide flammability limits, high flame speed and low ignition energy (Cozzi and Coghe 2006), hydrogen in the fuel allows to combustion systems to operate with lean fuel mixtures. The increase of flammability limits in presence of hydrogen offsets the harmful effects of a lean combustion such as local extinctions, energy losses by radiation as well as flame stretching (Briones et al. 2008) Furthermore, the hydrogen addition in the fuel changes the size of recirculation zone of combustion products without modify thermal power, global equivalence ratio, distance between the nozzles.

To limit NO_x emissions in burners with separated jets, the air is substituted by pure oxygen, so called oxy-combustion. In air combustion, nitrogen brings about a low yield of combustion and a high energetic consumption because the nitrogen contained in the air acts as an energy ballast. The total substitution of air by oxygen leads to the improvement of heat yield, the rise of adiabatic flame temperature (2200 K for CH₄-air, 3090 K in CH₄-oxy-combustion (Perthuis 1983)), the reduction of fuel consumption of 50%, and from an environmental point of view, the decrease of the nitrogen oxide formation (up to 95%) because of the decrease of nitrogen quantities in the oxidant (Genies 1996). However, the rise of the flame temperature favours the nitrogen oxide formation via NO thermal mechanism. In oxy-fuel burner, NO_x production is due to effects of air infiltration, presence of N₂ in fuel, flame radiation and aerodynamic straining (Sung and Law 1998).

The present study is conducted with a diffusion flame from a separated jet burner to gain an understanding of the influence of the recirculation of burned gases on NO_x emissions in terms of size and composition. Three parameters are used to modify the size of the burned gas recirculation zone: the distance between hythane and oxygen jets, the hydrogen volume fraction in natural gas, and the global equivalence ratio. NO_x emissions are studied according to these parameters. The advantage of the burned gas recirculation zone is its size, which has the ability to radically modify NO_x emissions without changing the characteristics of the combustion chamber.

The experimental setup is composed by a burner functioning with hythane and pure oxygen, placed in bottom wall of the combustion chamber. The gas sampling is achieved by a water-cooled probe and NO_x (NO+NO₂) are detected by a gas analyzer using the chemiluminescence technique.

Through the Particle Image Velocimetry (PIV), the study of the jet aerodynamic allows to characterize the size of the recirculation zone and to understand the influence of recirculation of combustion products on NO_x emissions.

In a first part, NO_x emissions are studied according to the hydrogen volume fraction in the fuel for a fixed equivalence ratio corresponding to the stoichiometric condition. In a second part, NO_x emissions are investigated as a function of the equivalence ratio without hydrogen addition.

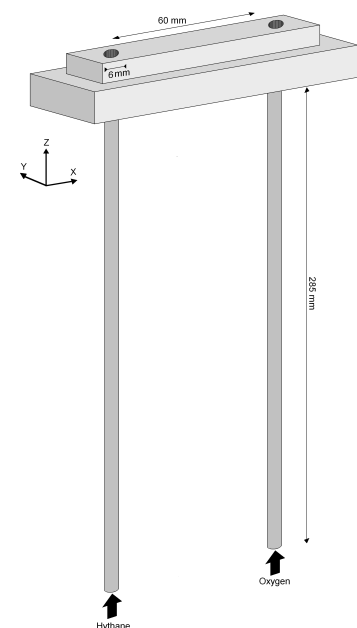


Fig. 1 Diagram of the burner with two separated jets.

2. Experimental section

2.1. Burner, flow control system and combustion chamber

The aim of this study is to investigate the effects of the dilution of the flame by the combustion products on NO_x emissions through the distance between the nozzles (from $D=12$ mm to $D=100$ mm), the hydrogen content in fuel (from $\alpha_{H_2}=0\%$ to $\alpha_{H_2}=20\%$) and the global equivalence ratio (from $\Phi=1$ to $\Phi=0.7$).

The burner depicted in Fig. 1 is made up of two non-ventilated jets, one of hythane (natural gas and hydrogen) and the other of pure oxygen. The separation distance between the nozzles (D) varies from 12 to 100 mm. The internal diameter of the nozzles is $d=6$ mm.

The natural gas has a density of 0.83 kg.m^{-3} and a volume composition of 85%CH₄, 9%C₂H₆, 3%C₃H₈, 2%N₂, 1%CO₂, and traces of higher hydrocarbon species. The hydrogen volume fraction in the fuel, $\alpha_{H_2} = \rho_{H_2} \dot{m}_{H_2} / (\rho_{H_2} \dot{m}_{H_2} + \rho_{NG} \dot{m}_{NG})$ is varied between 0% and 20%, with \dot{m} and ρ respectively the mass flow rate and the density and subscripts H₂ and NG representing hydrogen and natural gas. The oxygen has a purity of 99.5% and a density of 1.354 kg.m^{-3} (at 1 atm and 15 °C). The fuel flow rate (\dot{m}_{FUEL}) and the exit velocity of hythane (U_{FUEL}^0) depend on the hydrogen volume fraction in fuel blend ($\dot{m}_{FUEL} = \dot{m}_{NG} + \dot{m}_{H_2}$) and the equivalence ratio. For the pure natural gas configuration ($\alpha_{H_2}=0\%$) in stoichiometric proportion, thermal power $P=25$ kW, $\dot{m}_{FUEL} = 0.55 \text{ g.s}^{-1}$ and $U_{FUEL}^0 = 23.7 \text{ m.s}^{-1}$.

The flow rate and the exit velocity of oxygen are fixed whatever the configuration and correspond to the value calculated for a thermal power of 25 kW in stoichiometric proportions ($\Phi=1$), thus $\dot{m}_{O_2} = 1.954 \text{ g.s}^{-1}$ and $U_{O_2}^0 = 51.3 \text{ m.s}^{-1}$.

Table. 1 summarizes the parameters of this experimental study including natural gas and hydrogen flow rates (\dot{m}_{NG} and \dot{m}_{H_2}), the fuel exit velocity and Reynolds, as well as the initial velocity ratio r between fuel (natural gas + hydrogen) and oxygen.

Φ	P (kW)	Hythane Jet					Oxygen Jet			r $\frac{U_{FUEL}^0}{U_{O_2}^0}$
		α_{H_2}	\dot{m}_{NG} (g.s ⁻¹)	\dot{m}_{H_2} (g.s ⁻¹)	U_{FUEL}^0 (m.s ⁻¹)	Re	\dot{m}_{O_2} (g.s ⁻¹)	$U_{O_2}^0$ (m.s ⁻¹)	Re	
1	25	0%	0.5556	0	23.7	10761	1.954	51.3	21823	0.46
1	25	5%	0.5473	0.0031	24.5	10632	1.954	51.3	21823	0.48
1	25	10%	0.5384	0.0065	25.5	10498	1.954	51.3	21823	0.50
1	25	15%	0.5288	0.0101	26.5	10358	1.954	51.3	21823	0.52
1	25	20%	0.5184	0.0140	27.6	10212	1.954	51.3	21823	0.54
0.9	22.5	0%	0.5000	0	21.3	9684	1.954	51.3	21823	0.42
0.8	20	0%	0.4444	0	18.9	8608	1.954	51.3	21823	0.37
0.7	17.5	0%	0.3889	0	16.6	7532	1.954	51.3	21823	0.32

Table. 1 Parameters of the study. Inside diameter of the nozzles $d=6$ mm.

The regulation of natural gas flow is provided by a mass flow controller (TYLAN RDM 280) while the oxygen and the hydrogen flow rates are controlled by sonic throats related to pressure gauges. Flames from the oxy-burner take place in a combustion chamber, with a square cross section of $60 \times 60 \text{ cm}^2$ and 100 cm height, as shown in Fig. 2. To limit the wall solicitations and to keep a constant wall temperature of, the lateral walls are refractory-lined inside (12 mm of thickness) and water-cooled outside of the combustion chamber. The exhaust section is closed by a convergent

with a small square cross section opening of 12x12 cm² allowing the exhaust of flue gases. Six windows are made on every lateral wall in order to give optical accesses necessary to the Particle Image Velocimetry setup.

The combustion chamber reaches the equilibrium temperature after 25 minutes and the chemical equilibrium after 40 minutes. The chemical equilibrium corresponds to the moment from which air contained in the chamber is totally consumed, insuring the only presence of burned gases inside the chamber.

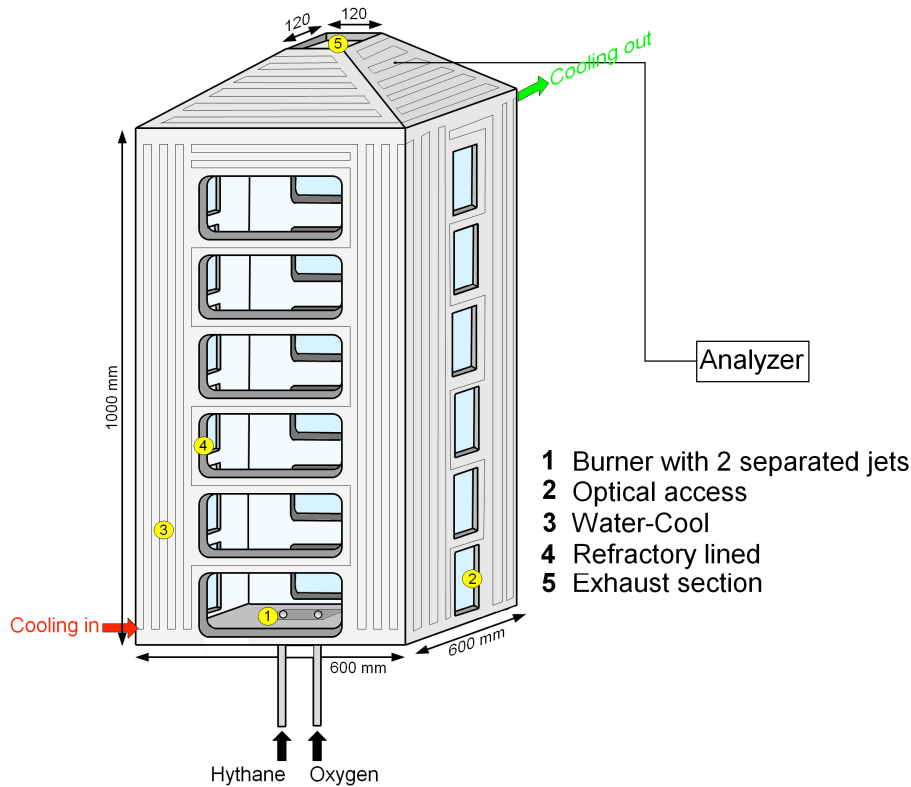


Fig. 2 Schematic of the combustion chamber.

2.2. Measurements of NO_x emissions

The sampling of combustion gases takes place at the exit of the combustion chamber, i.e. at the height $Z = 1240$ mm. Analyses are performed using a NO_x analyzer (HORIBA PG-250) related to a water cooled probe (1 mm in diameter) with a heated sample line transfer. The principle of nitrogen oxide measurements is based on chemiluminescence method. In this phenomenon, a part of the nitrogen monoxide (NO) contained in the sample gas reacts with the ozone (O₃) leading to the production of NO₂ according to equation 1:



A part of this generated NO₂ is in its excited state (NO₂^{*}) and returns to the normal states emitting a photon according to equation 2:



where h represents the Planck's constant and ν is the frequency of its associated electromagnetic wave.

The measurement of the photon flux emitted represents the NO concentration. The NO₂ is converted in NO with the Molybdenum-Carbon catalyst. The measurement of converted NO gives the NO₂ concentration. The measurement range of the NO_x analyser is between 0 ppm and 1000 ppm with repeatability equals to $\pm 0.5\%$ of full scale.

2.3. Velocity measurements

The Particle Image Velocimetry (P.I.V) is used to study the aerodynamic of the flow. This non-intrusive method permits to obtain 2-D images of the flow and to deduce the instantaneous two-dimensional velocity fields.

A double-pulsed Nd-Yag laser (Big Sky CFR200, Quantel) with a wavelength of 532 nm and a frequency of 10 Hz is used as light source (120 mJ/pulse and a pulse duration equals 8 ns). An optical system constituted by 3 consecutive lenses creates the laser sheet with a thickness of 500 μm and a height equals to 80 mm. A CCD camera with a dynamics range of 16 bits (2040*2040 pixel², Image Pro X Lavis) is oriented perpendicular to the laser sheet. A lens with a focal of +85 mm (1:14.1 Nikkor, Nikon) is placed perpendicular to the light source and collects the signal of Mie scattering emitted by the seeded particles (zirconium oxide (ZrO₂)) in the reacting flow. In front of the lens of the P.I.V setup, an interference filter (532 nm, 3 nm bandwidth) is used to reject the bright luminosity from the oxy-flame. Concerning the determination of the dynamic fields of the flow, the calculations of the cross-correlation images are carried out by the Davis software. The dimensions of interrogation windows are 64*64 pixels² with 50% covering, thus a grid step of 32 pixels.

To characterize the flow and to ameliorate the spatial resolution, it is necessary to realize measures according to four different heights (0 - 93.5 mm, 58 - 172 mm, 136 mm- 250 mm and 212-328 mm). Thus, the different pertinent zones of the flow can be studied in order to describe the recirculation zone, the fusion zone where the two jets begin to mix and interact and the combination zone where the flow tends to have similar behaviour to a single jet.

The processing of 500 image pairs enables to obtain 500 instantaneous velocity vector fields for every heights and configurations. The mean velocity field is obtained by averaging the 500 instantaneous fields.

Fig. 3 displays the mean two-dimensional velocity field and the characteristic zones of a flow with the two separated jet for the case of stoichiometric configuration, $\alpha_{H_2} = 20\%$ and $D=60$ mm. This study focuses on the recirculation zone of combustion products located between the two jets (see the red square in Fig.3).

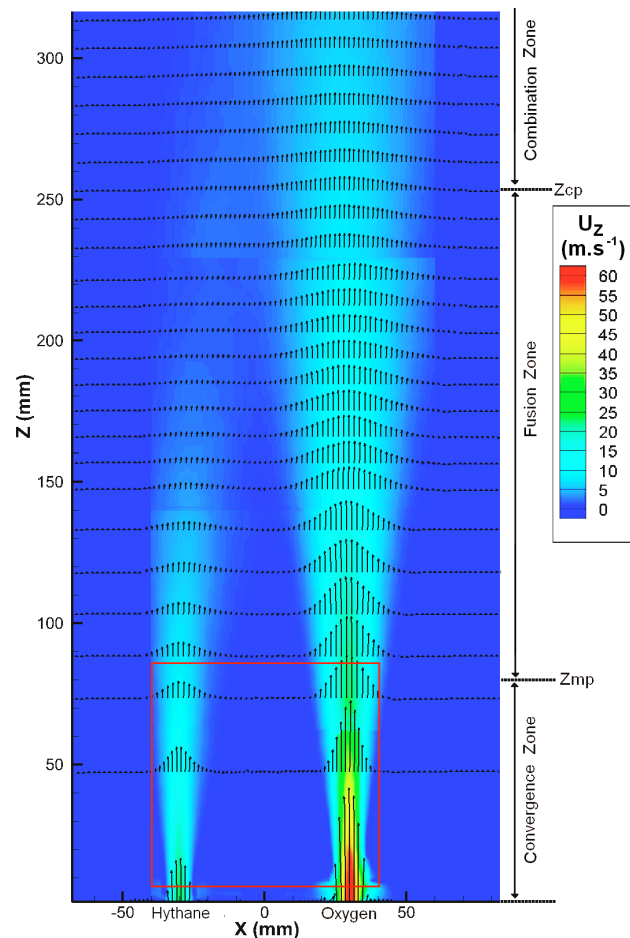


Fig. 3 Mean two dimensional velocity field and the characteristic zones of a flow with separated jets (case: $\Phi=1$, $\alpha_{H_2}= 20\%$ and $D=60$ mm). The red square displays the location of the refined P.I.V in the recirculation zone.

3. Results and discussion

3.1. Determination of size of recirculation zone

From the Particle Image Velocimetry in reacting flow, the size of recirculation zone is studied. The recirculation zone is defined between the burner exit ($Z = 0$ mm) and a characteristic height called the mixing point Z_{mp} . The Z_{mp} point is located as the height where the velocity between the two jets stops to be negative, equivalent to the end of the burner gases recirculation. After the mixing point Z_{mp} , the two jets start to mix and interact together corresponding to the beginning of the fusion zone of the jets.

The shape of the recirculation zone resembles a trapezium, where the base of the trapezium is formed by the distance between the nozzles, D , minus the internal diameter of the nozzles; Z_{mp} is the height of the trapezium, and the second base of the trapezium is the distance between the jets at the Z_{mp} height, as shown in Figure 4. The motions observed in the YZ-plane had no influence on the flow behavior. The PIV measurements in the YZ-plane indicated that the U_z mean velocity was near zero, except in the jet-plane ($Y=0$). In the jet-plane, the U_z mean velocity followed the same tendency as in the XZ-plane. Furthermore, the jet interaction zone in the YZ-plane was very thin and independent of the configuration. The PIV measurements in the YZ-plane showed that the recirculation zone was only influenced by 2D motions in the XZ-plane.

In order to study the velocity fields in the recirculation zone where the velocities are low (less than 1 m.s^{-1}), it is necessary to have important temporal and spatial resolutions. Table. 2 summarizes the different magnifications and time intervals Δt between two images of P.I.V corresponding to each burner configuration.

	$D=12 \text{ mm}$	$D=24 \text{ mm}$	$D=40 \text{ mm}$	$D=60 \text{ mm}$	$D=80 \text{ mm}$	$D=100 \text{ mm}$
Magnification (mm.pixel^{-1})	0.0316	0.0328	0.0364	0.0392	0.0419	0.0456
Δt (μs)	150	150	200	200	250	250

Table. 2 Magnification and time interval Δt of P.I.V with the distance between the nozzles.

Fig. 4 displays the two-dimensional velocity fields refined on the recirculation zone. When the temporal and spatial resolutions increase, the information on the velocities of the natural gas and oxygen jets is not pertinent, however, the velocity field between the two jets is more accurate. The correlation between the distance between the nozzles D and the size of the recirculation zone

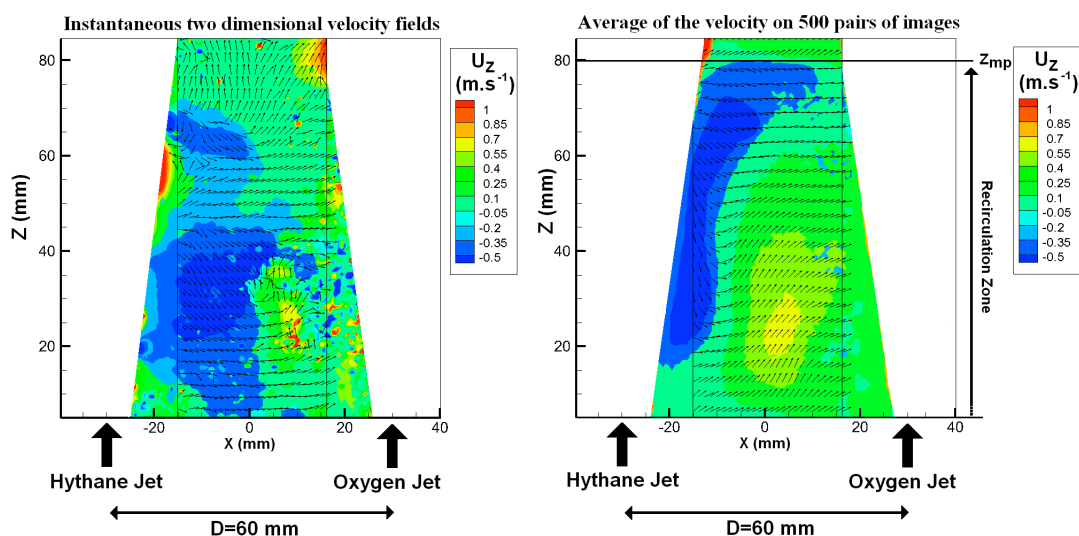


Fig. 4 Instantaneous and average two-dimensional velocity fields focusing on the recirculation zones of the flow ($\Phi=0.7$, $\alpha_{H_2}= 20\%$ and $D=60$ mm).

S_{ZR} is depicted in Fig. 5 without and with hydrogen addition.

The figure clearly shows an increase of the recirculation surface with the separation distance of nozzles D . When D becomes more important, the hythane jet and the oxygen jet interact more downstream, the height of the mixing point Z_{mp} increases and the surface of the recirculation zone grows.

It is noted that for pure natural gas configuration ($\alpha_{H_2} = 0\%$), when D increases from 12 mm to 100 mm, the size of the recirculation zone increases by a factor of 400 (from 22 mm² to 8900 mm²). The increase of the recirculation zone is less important when the fuel jet moves away from the oxygen jet. The hydrogen volume fraction also influences the size of the recirculation zone, in particular for large distances between the nozzles. In the case of $D=100$ mm, when the hydrogen volume fraction passes from $\alpha_{H_2} = 0\%$ (pure natural gas) to $\alpha_{H_2} = 20\%$, the size of the recirculation zone reduces by 12%.

The high molecular diffusivity of hydrogen favours the mixing between the jets, which allows an early fusion of the jets. From a thermal point of views, the combustibility and the heat release rates of hydrogen favour the reaction and the expansion of the burned gases quickly, which reduce the size of the recirculation (Kim et al. 2009).

3.2. Temperature of the burned gases

In order to study the influence of temperature on NO_x emissions, it is important to measure the temperature in the combustion chamber. By reason of the highly different structure of the flame according to the configuration (D and α_{H_2}), the common criterion chosen to follow the evolution of the temperature as a function of the study parameters, is the temperature of the fumes. This temperature of combustion products are performed inside the convergent section of the combustion chamber ($Z = 1240$ mm) using K thermocouples. Results are illustrated in Fig. 6 as a function of the distance between the nozzles D .

The increase of the distance between the nozzles induces a decrease of temperature of burned gases. In fact, in the case of the pure natural gas

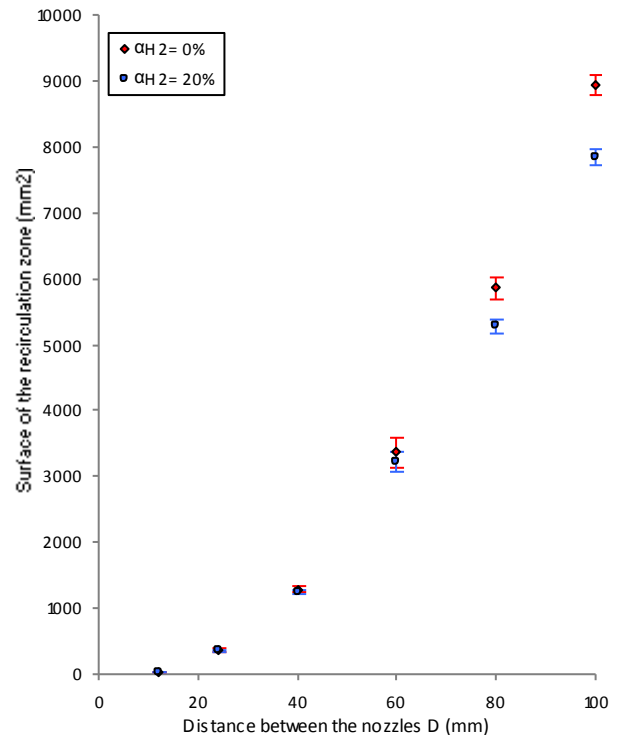


Fig. 5 Surface of the recirculation zone as a function of the distance between $\alpha_{H_2} = 0\%$ and $\alpha_{H_2} = 20\%$.

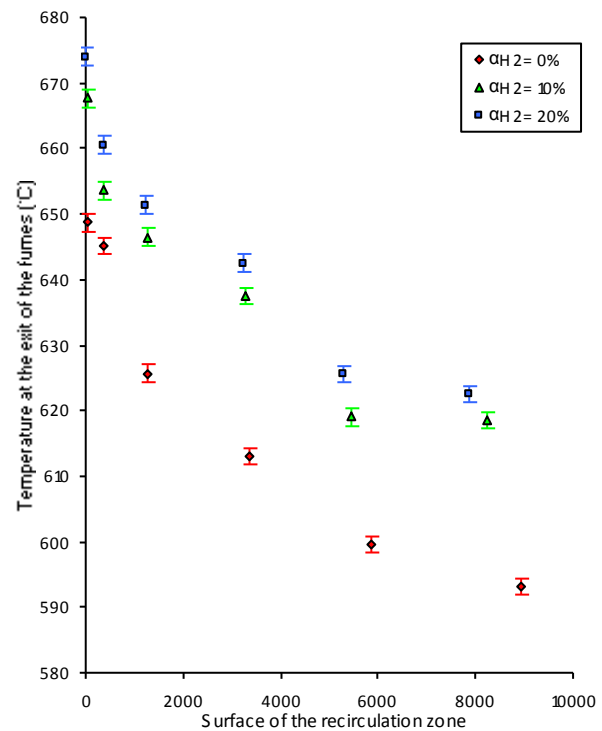


Fig. 6 Temperature of the burned gases at the chamber exit ($Z = 1240$ mm) versus the distance between the nozzles D for the hydrogen content in the fuel 0%, 10% and 20%.

($\alpha_{H_2}=0\%$), the temperature of fumes passes from 649 °C for $D=12$ mm to 593°C for $D=100$ mm.

The increase of the recirculation zone related to the increase of the distance between the nozzles promotes the flame dilution by combustion products reducing the flame temperature.

Fig. 6 shows that the hydrogen addition slightly increases the temperature of fumes. Note that whatever the distance between the jets, the variation of temperature of fumes is approximately similar. For the configuration of $D=12$ mm, adding 20% of hydrogen, the temperature of fumes increase from 649°C to 674°C, i.e. 25°C more than in the flame of pure natural gas. The increase of temperature of fumes is due to the increase of flame temperature with the hydrogen enrichment. Moreover with the hydrogen addition, the reduction of the size of recirculation zone limits the dilution of the flame by combustion products which limit the temperature of fumes reduction (Zhang et al. 2009).

3.3. NO_x emissions

Due to the high temperature in oxy-fuel combustion, the predominant NO_x formation is due to the thermal NO mechanism (Fenimore 1979). The NO_x formation in the present system is due to the presence of nitrogen in the fuel (2% in volume) and air leaks. Fig. 7 shows the variation of NO_x emissions as a function of the distance between the nozzles D for the hydrogen volume fraction in the fuel $\alpha_{H_2}=0\%$, $\alpha_{H_2}=10\%$ and $\alpha_{H_2}=20\%$. The effect of the separation distance between the jets on NO_x emissions is significant of all cases of the hydrogen content in the fuel as shown in the figure. NO_x emissions noticeably decrease when the distance D increases. For the pure natural gas case ($\alpha_{H_2}=0\%$), the NO_x rate decreases from 690 ppm to 25 ppm. Two parts of NO_x rate distribution with D are observed: a very high decay between $D=12$ mm and $D=40$ mm, then a slow decay between $D=40$ mm and $D=100$ mm. The variation in NO_x rate in the first part is 600 ppm (from 690 ppm to 90 ppm), while this variation is about 65 ppm (from 90 ppm to 25 ppm) in the second part in the case of $\alpha_{H_2}=0\%$.

When the spacing between the jets increases, the recirculation rates of reactants by combustion products increase and the temperature decreases limiting NO_x formation by thermal NO mechanism. As regards the hydrogen effect, the results display that NO_x emissions increases with the increase of the hydrogen volume fraction in the fuel. This increase can be explained by the increase of flame temperature which favors in particular the formation of thermal NO. The high diffusivity of hydrogen can reduce the size of the recirculation zone leading to a decrease of the flame dilution by the combustion products. This limits the temperature reduction and consequently the NO_x rate reduction is limited.

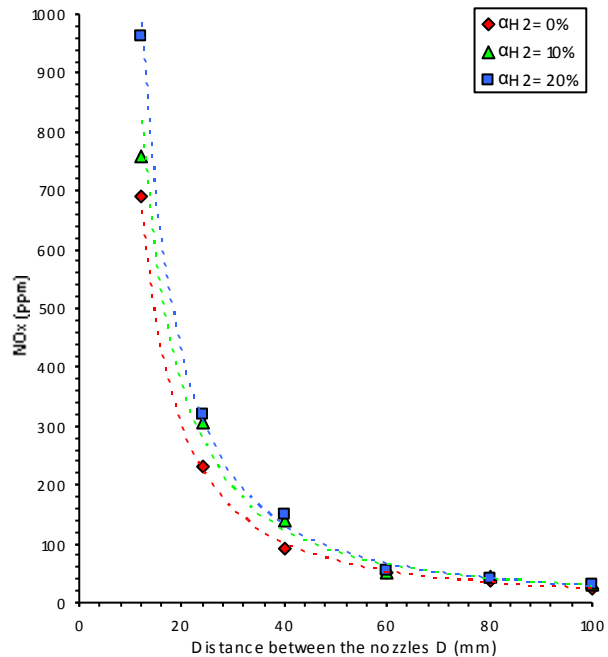


Fig. 7 NO_x emissions as a function of the distance between the nozzles D for different hydrogen volume fraction

3.4. NO_x concentration according to the size of the recirculation zone

This section deals with the link between NO_x emissions and the size of recirculation zone. Fig. 8 shows NO_x rates with the recirculation surface of burned gases for different hydrogen volume fraction α_{H_2} . The results show high NO_x emissions for small recirculation size and weak NO_x emissions for large surface of burned gas recirculation regardless the hydrogen volume fraction. It can be noted that NO_x emissions is linked to a size of the recirculation zone following a hyperbolic decrease. This tendency leads to study the inverse ratio of NO_x emissions as a function of the surface of recirculation zone (Fig. 9). In the case of burner with two separated jets, NO_x emissions at the exit of the fumes are inversely proportional to the surface of recirculation zone of burned gases ($1/NO_x \propto S_{ZR}$). This tendency allows to deduce NO_x emissions from size of recirculation zone and proves the efficiency of the separation of the nozzles to reduce NO_x concentrations.

Size of the recirculation of burned gases and its impacts on flame temperature is essential to reduce NO_x emissions by Zeldovich mechanism.

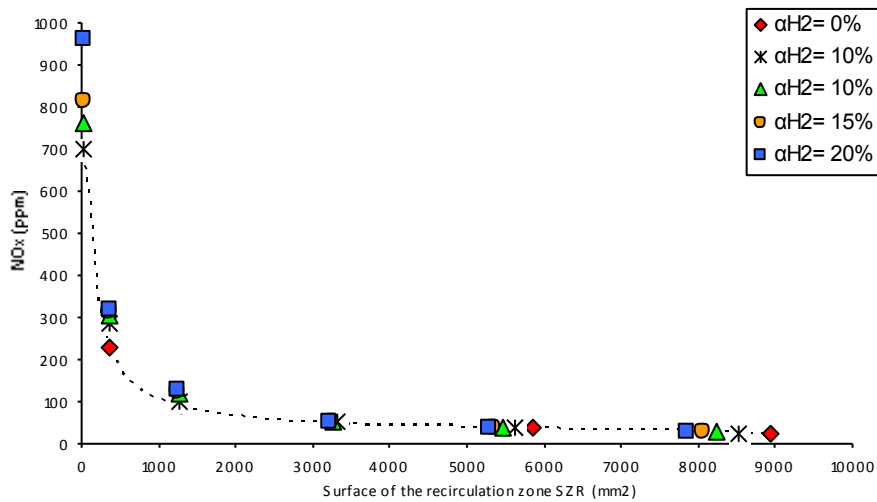


Fig. 8 NO_x emissions as a function of the surface of recirculation zone S_{ZR} for different hydrogen volume fraction α_{H_2} .

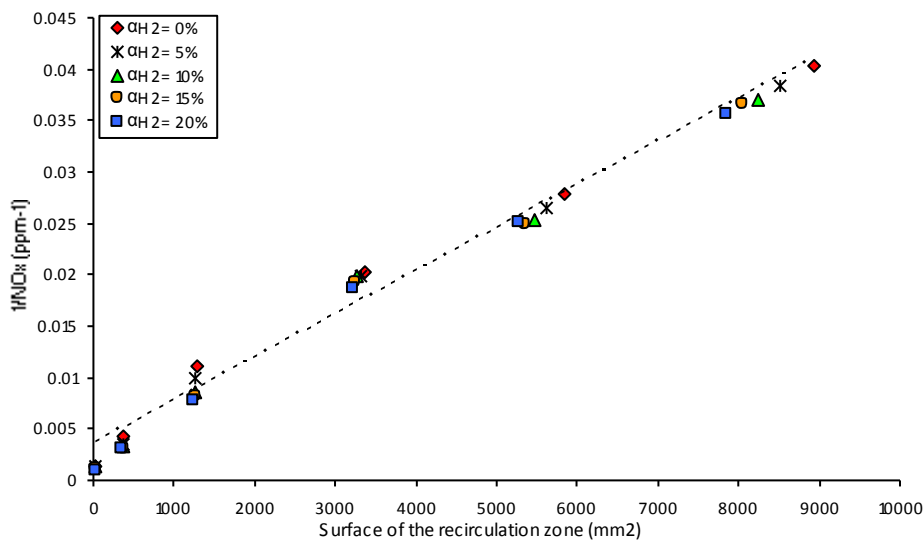


Fig. 9 Inverse of NO_x emissions according the surface of the recirculation zone for different hydrogen volume fraction α_{H_2} .

3.5. NO_x emissions according to the equivalence ratio

The study of NO_x emissions as a function of the size of the recirculation zone for different hydrogen volume fraction (from α_{H_2} = 0% to 20%) shows that NO_x emissions deeply depend to recirculated combustion products. In oxy-fuel combustion in diffusion flame, Park et al. (2009) show that NO_x emissions are linked to recirculated CO₂ in the flame. They focus on the role of the recirculated CO₂ and its chemical effects on the formation and destruction of NO_x through Fenimore (prompt NO) and Zeldovich (thermal NO) mechanisms. In oxy-fuel combustion, the recirculated CO₂ modifies oxidation reaction pathways and affect NO_x emissions behaviour in both mechanisms of thermal NO and prompt NO. The interruption of formation and destruction of NO_x depends to the behaviour of the recirculated CO₂.

From an experimental point of view, the unpredictability of air infiltration, the presence of nitrogen in the fuel, the size of recirculation zone and the aerodynamic straining of the flow make difficult the study of NO_x emissions.

The parameter retained to vary the amount of recirculated CO₂ in oxy-fuel combustion is the global equivalence ratio Φ . From the equation of combustion (equation 3) between the natural gas and pure oxygen, the variation of the global equivalence ratio Φ modifies the CO₂ rate in combustion products.

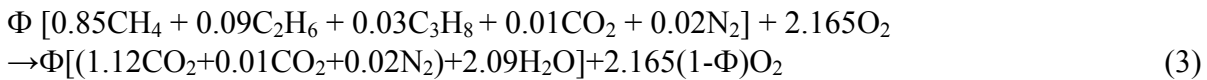


Table. 3 summarizes the results of temperature of fumes T , the size of the recirculation zone S_{ZR} and NO_x emissions according to the global equivalence (from $\Phi=0.7$ to $\Phi=1$) without hydrogen addition (α_{H_2} = 0%).

Φ	D=12 mm			D=24 mm			D=40 mm			D=60 mm			D=80 mm			D=100 mm		
	T (°C)	S_{ZR} (mm ²)	NO_x (ppm)	T (°C)	S_{ZR} (mm ²)	NO_x (ppm)	T (°C)	S_{ZR} (mm ²)	NO_x (ppm)	T (°C)	S_{ZR} (mm ²)	NO_x (ppm)	T (°C)	S_{ZR} (mm ²)	NO_x (ppm)	T (°C)	S_{ZR} (mm ²)	NO_x (ppm)
1	649	23	690	645	364	230	626	1287	91	613	3359	49	596	5854	36	593	8933	25
0.9	624	22	726	618	358	265	613	1260	99	606	3323	52	591	5759	48	587	8669	32
0.8	588	20	752	584	346	288	579	1224	120	562	3255	58	552	5581	55	548	8367	37
0.7	560	19	787	562	332	310	537	1175	138	515	3187	78	509	5312	58	507	7872	41

Table. 3 Temperature of fumes T , the size of the recirculation zone S_{ZR} and NO_x emissions according to the equivalence ratio (from $\Phi=0.7$ to $\Phi=1$) without hydrogen addition (α_{H_2} = 0%).

The decrease of the fuel flow rate enables to fuel jet to interact more in upstream in the flow with oxygen jet. The increase of the velocity difference between the two jet leads to an increase of the diversion of fuel jet toward the oxidizer jet: jets mix more upstream and reduce the size of the recirculation S_{ZR} . The decrease of the global equivalence ratio leads to a fall of the flame temperature. This decrease is appearing through the reduction of the combustion products temperature as shown in Table. 3.

For example for the configuration with $D=12$ mm, the temperature of the fumes passes from 649°C for $\Phi=1$ to $T=560^\circ\text{C}$ for $\Phi=0.7$.

Figure 10 illustrates NO_x emissions with the equivalence ratio for different distances between the jets. As shown below the increase of the spacing between the jets decreases the NO_x rate. The decrease of global equivalence ratio leads to an increase of NO_x emissions. Note that this effect is opposite to the size of the recirculation zone. For a distance between the nozzle $D=40$ mm, NO_x emissions increases from 91 ppm for $\Phi=1$ to 140 ppm for $\Phi=0.7$. This tendency of NO_x emissions is due to destruction and formation of NO_x through the thermal and Fenimore mechanisms.

Due to oxygen excess in lean combustion, the mole fraction of CO_2 in combustion products decreases when the global equivalence ratio Φ decreases. This reduction of CO_2 emissions in a lean flame induces a decrease in the amount of recirculated CO_2 which chemically reduces the effects on nitrogen oxide formations. In diffusion oxy-flames with recirculation of burned gases, prompt NO and thermal NO react with CO_2 and decrease NO_x emissions (Park et al. 2009). The decrease of the amount of CO_2 in recirculation zone leads to a rise of NO_x emissions. In the burner with separated jets, it is found that NO_x emissions are highly depend on the size and the composition of recirculation zone of burned gases.

4. Conclusions

The effects of recirculation of burned gases on NO_x emissions in natural gas-hydrogen-oxygen flames from a burner with separated jets have been experimentally investigated. The oxy-flame is used from a burner composed with two nozzles and develops inside a combustion chamber. The measurements of NO_x concentrations are carried out at the exit of the combustion chamber using a water-cooled probe and NO_x analyser.

The increase of the distance between the nozzles raises the size of the recirculation zone and favours the dilution of reactants. The dilution by combustion products leads to a decrease in NO_x emissions up to 96%. Without hydrogen addition, NO_x rate decreases from 690 ppm for $D=12$ mm to 25 ppm for $D=100$ mm. The dilution of reactants by combustion products decreases the flame temperature and the NO_x formation particularly through thermal mechanism.

The high molecular diffusivity and combustibility of hydrogen favour the mixing between the jets and reduce the size of the recirculation zone. The dilution of oxy-flame by combustion products is less important with hydrogen addition and flame temperature increases. In the case of $D=12$ mm, NO_x emissions raises up to 39% when the hydrogen in the fuel is added by 20%. In oxy-fuel combustion with non-premixed flame, NO_x emissions are inversely proportional to the size of the recirculation zone of combustion product.

With recirculated CO_2 , the global equivalence ratio has a significant effect on NO_x emissions

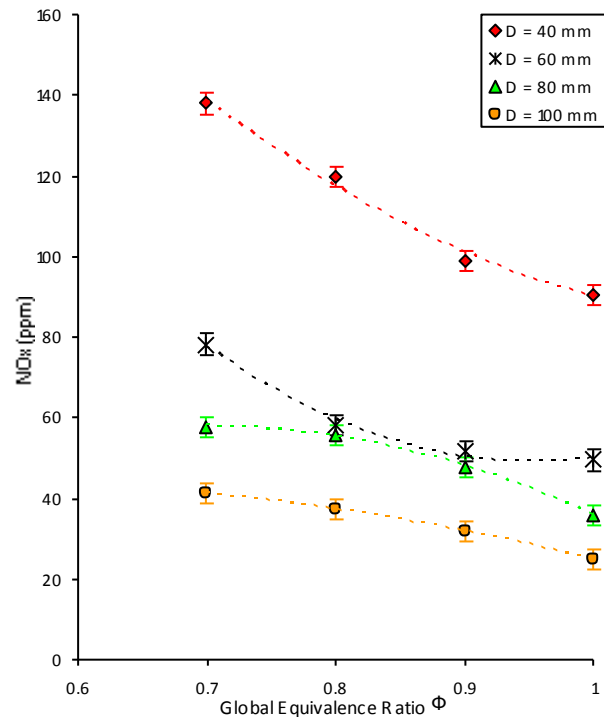


Fig. 10 NO_x emissions as a function of global equivalence ratio Φ for different distance between the nozzle D .

because recirculated CO₂ reacts with prompt NO and thermal NO and reduces NO_x emissions. The decrease of recirculated CO₂ linked to a lean combustion limits the reaction between NO and CO₂ and NO_x emissions in lean combustion are more important than for a stoichiometric configuration. In oxy-fuel combustion with burner with two separated jets, NO_x emissions depend on the size and composition of the recirculation of burned gases, which are central to limit NO_x emissions at the exit of the fumes.

References:

- Boushaki T, Sautet JC, Labegorre B (2009) Control of flames by radial jet actuators in oxy-fuel burners. *Combust. Flame* 156: 2043-2055
- Briones AM, Aggarwal SK, Katta VR (2008) Effects of hydrogen enrichment on the propagation characteristics of methane-air triple flames *Combust. Flame* 153:367-386
- Cozzi F, Coghe A (2006) Behavior of hydrogen-enriched non-premixed swirled natural gas flames *Int. J. Hydrogen Energy* 31:669-677
- Dugué J, Riffart S (2006) Recent developments in oxy-combustion and potential applications in refinery furnaces 7th European Conference on Industrial Furnaces and Boilers, INFUB7 18–21
- Fenimore CP (2009). In. *Proc. Seventeenth symposium (international) on combustion*. PA: Combust. Inst. 661-669
- Genies B (1996) *A.D.E.M.E* 153-196
- Hwang CH, Lee S, Lee CE (2008) The effect of turbulence intensity of ambient air flow on NO_x emissions in H₂/air nonpremixed jet flames. *Int. J. Hydrogen Energy* 33:832-841
- Kim M, Yoon M (2007) Flame residence time and strain rate in turbulent hydrogen non-premixed jet flames with coaxial air. *Proc. Combust. Inst.* 31:1609-1616
- Kim SH, Arghode VK, Linck MB, Gupta AK (2009) Hydrogen addition effects in a confined swirl-stabilized methane-air flame *Int. J. Hydrogen Energy* 34:1054-1062
- Naha S, Aggarwal, SK (2004) Fuel effects on NO_x emissions in partially premixed flames. *Combust. Flame* 139: 90-105
- Park J, Park JS, Kim H.P, Kim JS, Kim SC, Choi JG, Cho HC, Cho KW, Park HS (2007) NO behaviour in oxy-fuel combustion recirculated with carbon dioxide *Energy Fuels* 21:121-129
- Perthuis E (1983) in: *La Combustion Industrielle*, Technip, Paris
- Røkke NA, Hustad JE, Sønju OK, Williams FA (1992) Scaling of nitric oxide emissions from buoyancy-dominated hydrocarbon turbulent-jet diffusion flames. *Proc. Combust. Inst.* 24:385-393
- Salentey L (2002) Ph-D thesis, University of Rouen
- Shinomori K, Katou K, Shimokuri D, Ishizuka S (2011) NO_x emission characteristics and aerodynamic structure of a self-recirculation type burner for small boilers. *Proc. Combust. Inst.* 33:2735-2742
- Sung CJ, Law CK (1998) Dominant chemistry and physical factors affecting NO formation and control in oxy-fuel burning *Proc. Combust. Inst.* 27:1411-1418
- Yon S, Sautet JC (2012) Flame lift-off height, velocity flow and mixing of hythane in oxy-combustion in a burner with two separated jets *Appl. Therm. Eng.* 32:83-92
- Zhakatakev (2004) *Circulatory flow in the inner jet zone of a system of free turbulent jet* T.A. Plenum Publishing Corporation
- Zhang Y, Wu J, Ishizuka S (2009) Hydrogen addition effect on laminar burning velocity, flame temperature and flame stability of a planar and a curved CH₄-H₂-air premixed flame *Int. J. Hydrogen Energy* 34:519-527