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## Article

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## Research paper

# Experimental evaluation of CO, NO<sub>x</sub>, formaldehyde and acetaldehyde emission rates in a combustion chamber with OEC under acoustic excitation



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## ABSTRACT

This study experimentally evaluates the interaction of the oxygen enhanced combustion (OEC) technique with the pulsating combustion technique by the acoustic excitation of flames, the effects of these techniques on atmospheric emissions of CO, NO<sub>x</sub>, formaldehyde and acetaldehyde, and the temperature of exhaust gases in diffusive and confined natural gas flames. The results showed a general trend of reductions in the emissions of CO and NO<sub>x</sub> with the enrichment of the oxidant with O<sub>2</sub> and also under some conditions with an acoustically excited flame. The results showed that the acetaldehyde emissions decreased with the ratio of equivalence but increased in the presence of acoustic excitation. Formaldehyde emissions showed no significant trend. The results show that the simultaneous application of the OEC and acoustic excitation techniques in a controlled manner can reduce pollutant emissions and increase the efficiency of thermal combustion equipment.

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## 1. Introduction

Industrial burners are major pieces of equipment in which the thermal efficiency must be increased to reduce fuel consumption and the emissions of polluting gases that cause the greenhouse effect. Carbon monoxide (CO) and nitrogen oxides (NO<sub>x</sub>) are produced by burning fossil fuels and have been proven to be the causes of global warming and climate change. To reduce such damage to the environment, most developed countries have restricted pollutant emissions, which has resulted in increased studies on the use of clean fuels and techniques to increase process efficiency (Carvalho and Lacava, 2003).

Carbonyl compounds (CC) are another type of pollutant that must be controlled in the operation of industrial burners. Formaldehyde and acetaldehyde are carbonyl compounds that are emitted in high concentrations through the burning of fossil fuels (Guarieiro et al., 2008a, 2009; Chenyang et al., 2018). Aldehydes play an important role in the formation of ozone and photochemical oxidants in the atmosphere. The impact of these compounds on human health is problematic because they are

considered carcinogenic compounds by the International Agency for Research on Cancer (IARC). Such compounds can also be formed indirectly by the atmospheric photo-oxidation of volatile organic compounds (VOCs) emitted by anthropic and/or natural sources. Among the anthropogenic sources of CC emissions are industrial processes, waste incineration, and the burning of wood, fuels and forests (Rodríguez et al., 2012).

At the end of the 1960s, the Oxygen enhanced combustion (OEC) process, which is a controlled process that causes an increase of O<sub>2</sub> concentration in the conventional oxidizing current (atmospheric air), was developed on an industrial scale in the metallurgy of ferrous and non-ferrous materials (Baukal, 1998).

The choice of the enrichment level with O<sub>2</sub> ( $\Omega$ ) depends on several factors. High ( $\Omega > 0.9$ ) and medium ( $0.3 < \Omega < 0.9$ ) enrichment levels are only recommended for cases requiring high temperatures. The use of a low level of O<sub>2</sub> enrichment ( $\Omega < 0.3$ ) is highly recommended for retrofit applications because of the low costs involved in modifying existing systems and the low oxygen supply required (Santos et al., 2011). In most cases, burners can operate successfully using combustion air enriched with O<sub>2</sub> up to 28% by volume in the oxidizer without modifications in the equipment (Santos et al., 2009).

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## Nomenclature

### Alphabetic symbols

CO	Carbon monoxide
NO <sub>x</sub>	Nitrogen oxides
OEC	Oxygen enhanced combustion
CC	Carbonyl compounds
CP	Central Point
HPLC	High performance liquid chromatography
DOE	Design of experiments
R <sup>2</sup>	Determination coefficient
F <sub>q</sub>	Frecuence

### Greek symbols

$\Omega$	Enrichment level
$\phi$	Equivalence ratio

The application of OEC in combustion systems, such as industrial burners, has considerable benefits, such as increased productivity and efficiency of heat transfer processes, lower volumes of exhaust gas and reduced fuel consumption. This technique is widespread in high-temperature processes, such as steelworks, ceramic material production processes, and heating and drying processes of materials and waste incinerators, but it is still rarely used in power plants and in the oil and gas industry (Baukal, 1998).

However, the use of the OEC technique with burners may lead to increased formation of soot in certain regions of the flame (Santos et al., 2009). Because soot is an important factor in thermal radiation of the flames and the OEC has a tendency in the soot formation on the flames pyrolysis zone, its properly controlled interaction with the OEC may increase the thermal efficiency of the burners by increasing the heat transfer from the flames to the heating surfaces through thermal radiation. The OEC can also generate an increase in NO<sub>x</sub> emissions in oxygen-enriched systems. However, two competing factors operate: the increase in the NO<sub>x</sub> concentration with increasing temperature and the decrease in NO<sub>x</sub> formation due to the lower amount of nitrogen available in the oxidizing agent with higher levels of enrichment (Baukal, 1998; Santos et al., 2009).

OEC was used by Santos et al. (2016) to analyze formaldehyde and acetaldehyde emissions in a combustion chamber for acetylene burning, and oxidant enrichments of 2% and 4% were shown to reduce the emission rates of formaldehyde and acetaldehyde with OEC compared to the use of atmospheric air as an oxidizer. In this case, a greater reduction of formaldehyde emissions was observed (Santos et al., 2016).

Numerically and experimentally evaluated the performance of a scrap preheating furnace using natural gas as a fuel and an oxidizer enriched with oxygen at volumes of 21% to 100%. The results showed that the temperature in the oven increased sharply with the increasing mole fraction of oxygen in the oxidizing agent; in addition, the furnace's heating characteristic time decreased exponentially with the increase in thermal power (Jeongseog et al., 2015).

Another technique that has been studied with the aim of improving the efficiency of thermal combustion equipment is “pulsating combustion”, which can be defined as a burning process in which some state variables can be changed with time, such as the temperature, pressure or other variables (Zinn, 1986).

In 1859, Rijke discovered that a strong sound was produced by a hot metal screen located in the lower half of an upright, open tube. Based on Rijke's observations, Lord Rayleigh was the first to qualitatively describe heat-driven acoustic oscillation in 1878. He described the balance between acoustic dissipation and acoustic excitation. The occurrence of amplification or attenuation of the acoustic pressure by the addition of heat can be defined by the “Rayleigh criterion” (Raun et al., 1993).

In his criterion, Rayleigh says that an amplification of the pressure oscillation occurs when thermal energy is added to the combustion process at the instant of maximum compression, or heat is withdrawn at the moment of maximum rarefaction and the opposite denotes attenuation at the pressure oscillation.

Rijke tubes are often used in the analysis of acoustic excitation phenomena. However, most of these studies are focused primarily on the oscillations generated in the process itself rather than the use of external excitation. Thus, researchers can only measure acoustic oscillation data that are created by the flame itself (Raun et al., 1993).

Researchers have studied the excitation effect of an external source on a flame in a cylindrical tube and have observed interesting flame patterns. The results indicated that the patterns and properties of the flames are sensitive to the frequencies and amplitudes of the acoustic excitation (Farhat et al., 2005; Papadopoulos et al., 2001).

The characteristics of diffusion flames under acoustic excitation have been widely studied in the combustion community. The ranges of acoustic frequencies and amplitudes can affect the dynamics of the flame. Several conditions of forced acoustic frequencies may induce unstable flame behavior. In a closed combustion chamber, this instability can increase due to the coupling between the heat release rate and the acoustic pressure (Shahi et al., 2014; Chao et al., 1996). However, at some excitation frequencies, forced acoustic excitation can be used to control combustion and improve thermal efficiency and pollutant emissions (Toong et al., 1965; Foo et al., 2017).

Researches discussed the effects of acoustically forcing lifted diffusion flames at a frequency of 200 Hz. They found that the acoustic excitation produced almost homogeneous turbulence, which provided efficient mixing and resulted in a better combustion regime (Demare and Baillet, 2004).

Acoustic excitation has been shown to be an interesting technique in combustion systems of fossil fuels, mainly in the generation of energy, because it offers several advantages over the conventional combustion process. One of the advantages is a higher mixing rate between the fuel and the oxidant, which increases the turbulence in the flame region and results in more efficient burning and consequently in a reduction in the amount of fuel used. In addition, it decreases the emissions of gaseous pollutants from partial oxidation and particulates, increases the heat transfer in the combustion chamber, and requires less capital investment than conventional processes (Carvalho et al., 1987).

More recently, other articles have studied the influence of acoustic excitation on the gaseous and liquid fuels combustion (Huang et al., 2012; Wang et al., 2013; Karimi, 2014; Ju et al., 2017; Saurabh and Paschereit, 2017; Foo et al., 2018a,b; Niegoda-jew et al., 2018; Weiss et al., 2018; Bennewitz et al., 2018; Zhang et al., 2019; Liu et al., 2018).

The improvements provided by using OEC as well as the beneficial effects generated by the acoustic excitation of flames indicate that the use of both methodologies may further optimize the combustion of gaseous fuels. Thus, the aim of this study was to analyze the NO<sub>x</sub>, CO, formaldehyde and acetaldehyde emissions from natural gas burning in a combustion chamber to explore the effects of the percentage of oxygen in the oxidant and the variation of sound intensity on the emission rates of these compounds.

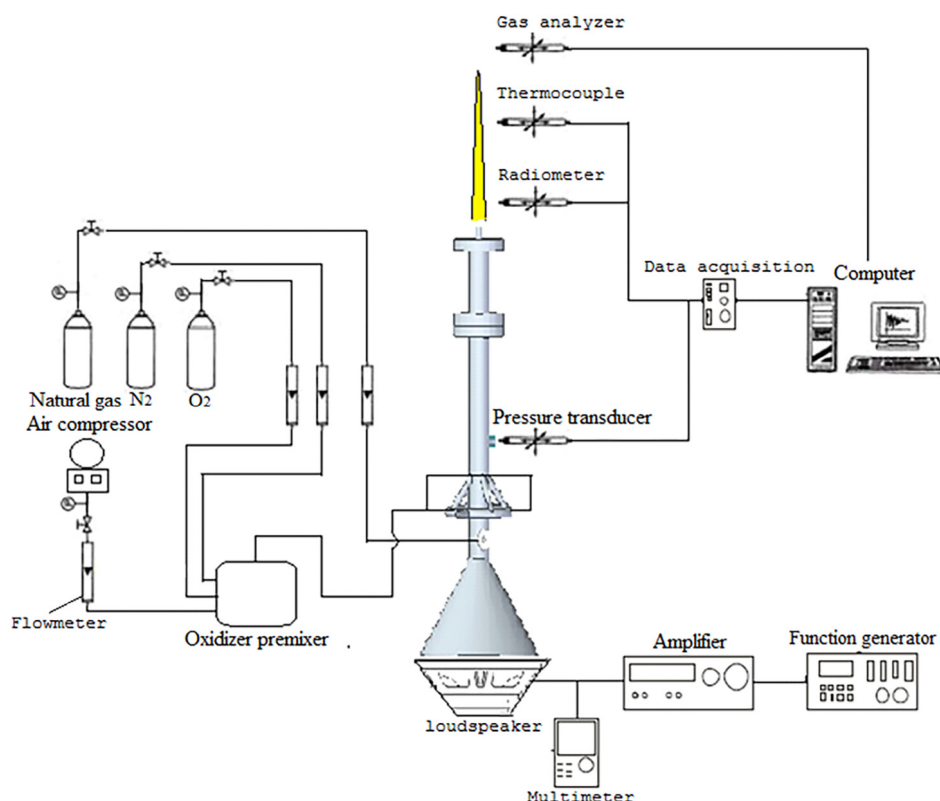


Fig. 1. Schematic diagram of the operation of the experimental prototype.

## 2. Experimental apparatus

### 2.1. Experimental burner characteristics

The experimental device was designed to produce a confined diffusive flame of natural gas. The construction of the experimental burner was inspired by the combustion device developed by Ferreira (2001), which incorporated a sound system with an external source (loudspeaker). It also has the characteristics of the burner developed by Santos (2010), which was designed with an O<sub>2</sub> enrichment system in the combustion air stream and has data acquisition and interpretation devices for the analysis. The source of the acoustic field was an automotive Woofer type speaker with 12 inches diameter, which was responsible for the acoustic excitation imposed on the combustion. Enrichment of the oxidant was performed by injecting pure O<sub>2</sub> (99% purity) into the oxidant line, where it was mixed into the atmospheric air through the premixer. The enrichment premixer was designed to enrich the air going from the feed line to the fuel burning system with oxygen.

Fig. 1 shows a schematic diagram of the operation of the sound excitation system and the enrichment of the combustion air with O<sub>2</sub>. The figure shows the inlet lines for natural gas, oxygen, nitrogen and compressed atmospheric air generated from a compressor and the respective flowmeters and flow rate control valves. All of the oxidants pass through the premixer before entering the burner to homogenize the atmospheric air and oxygen. Fig. 1 also shows the sound amplification system, in which it is possible to vary the frequency and voltage, and shows how the data are acquired for the interpretation of the results.

Three controlled input factors were considered for the experiment: the voltage at the speaker, the level of oxidant enrichment (on the volumetric fraction), and the air/fuel equivalence factor. The voltage applied to the speaker is a specific characteristic of the technique of combustion excited by sound waves, the

oxidizer enrichment level ( $\Omega$ ) is a characteristic factor of the OEC technique, and the ratio of equivalence ( $\phi$ ) is a common variable in studies of combustion phenomenon that refers to the excess air involved in burning. The response variables to be analyzed are temperature, CO concentration and NO<sub>x</sub> concentration of the exhaust gases. An analysis of carbonyl compounds (CC) was also performed, specifically the formaldehyde and acetaldehyde in the exhaust gases.

### 2.2. Exhaust gases analysis

The exhaust gases were analyzed using a portable gas analyzer with measurement cells. The instrument was a TEMPSET 100 model manufactured by Telegan Monitoring Ltd<sup>®</sup>. The instrument allowed the measurement of carbon monoxide (CO) and nitrogen oxides (NO<sub>x</sub>, including nitric oxide (NO) and nitrogen dioxide (NO<sub>2</sub>)), and it included a thermocouple in the measuring probe that allowed temperature measurements of the exhaust gases. The measurements were performed to 1.1 m from the fuel nozzle in the combustion chamber. Fig. 2 shows a detail of the point of collection of the exhaust gases.

Besides these, formaldehyde and acetaldehyde were collected using impinger-type bubbler bottles in series containing 2, 4-dinitrophenylhydrazine (2, 4 DNPHi) derivative solution, for the formation of the respective hydrazones. A 2, 4 DNPHi solution with a concentration of 75 mg/L was prepared. A total of 25 mL of the solution was added to 1 mL of 1 N phosphoric acid for each impinger. The collection time was 5 min for each replicate at a controlled flow rate of 24 L/h and an ice bath temperature of approximately 6 °C (adapted from procedure reported by Guarieiro et al., 2008a).

The CC analysis were performed using a Shimadzu High Performance Liquid Chromatograph (HPLC) (model LC-20AT) equipped with a quaternary solvent pumping unit, an automatic injector (SIL-20AHT), a diode array detector (SPD-M20A), a degasser

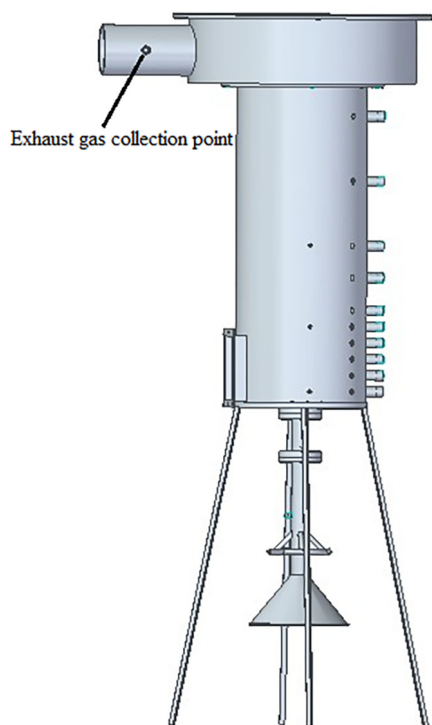


Fig. 2. Detail of the exhaust gas collection point.

(DGu-205), a furnace with a column (CTO-20A) and a controller interface (CBM-20A). The CC separations were carried out using a SHIMADZU® VP-ODS column (250 × 4.6 mm ID, particle size 4.6 μm) connected to a SHIMADZU® GVP-ODS pre-column (4.6 mm ID × 10 mm).

A mixture of ultrapure water (solvent A) and acetonitrile (solvent B) was used as the mobile phase (1.5 mL/min flow). The mobile phase gradient was 0 to 5 min (72% B); 5 to 10 min (72%–100% B); 17 to 23 min (100%–72% B); 23–25 min (72%). The wavelength used for the analyses were of 365 nm, with a total running time of 25 min and injected volume of the samples and standard solutions of 20 μL. The quantification of the CC was done using analytical standards of formaldehyde and acetaldehyde, in the form of their respective hydrazones, by means of external standardization.

### 2.3. Statistical analysis

The experiment was conducted following the Design of Experiments (DOE) methodology and was a factorial experiment (2<sup>3</sup> + center point) of two levels of each of the three controllable input factors. A statistical analysis was performed to identify the factors and combinations of factors that have greater effects on each variable response of the process as well as to obtain a model that represents the physical phenomenon being studied to allow the analysis and interpretation of the results.

The combination of each level of the three input factors plus a central point that was considered provided nine (9) different test conditions. Triplicates of each condition were performed, which led to a total of 27 trials, which were performed in a randomized way to reduce the possible interferences of the variables considered to be fixed.

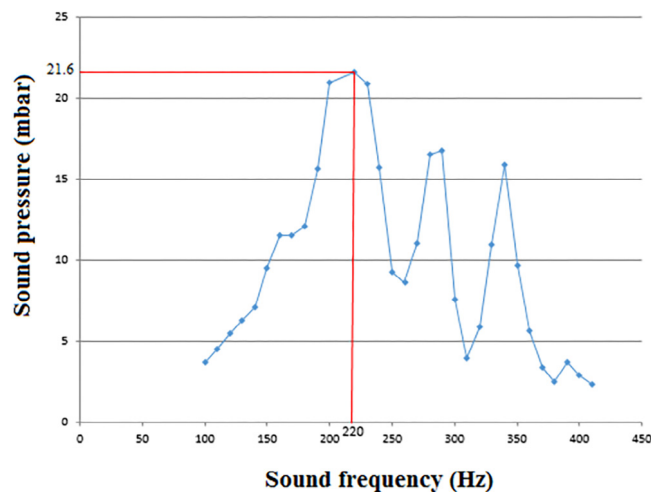


Fig. 3. Frequency vs. amplitude of the sound pressure.

## 3. Results and discussion

### 3.1. Characterization of the projected burner

For characterizing the limits of the voltage in the speaker, an initial test was performed without a flame (cold), in which a graph of the sound frequency vs. the sound pressure amplitude measured in the pressure transducer located in the air feed was made. The graph is shown in Fig. 3.

The initial test was performed by varying the frequency (Fq) at increments of 10 Hz, in the range from 100 to 400 Hz (values below 100 Hz and above 400 Hz did not have significant peaks of the sound pressure amplitude), and applying a constant voltage of 13 V to the speaker. The graph revealed three sound pressure peaks, which are the harmonics of the burner. The highest peak pressure of 21.6 mbar occurred at a sound frequency of 220 Hz, so this frequency was assumed to be fixed in the tests with sound excitation. The high voltage level (1) on the speaker was determined by conducting an initial hot test (all of the hot tests were performed after the flame had been lit for 1 h to heat the combustion chamber) and varying this factor to the point at which the flame detached from the edge of the burner. A maximum voltage of 5.3 V and a sound pressure of 8.8 mbar were found in the fuel-rich combustion conditions without oxygen enrichment, which is a critical condition for the flame to be more unstable; consequently, the zero sound excitation condition was considered to be the lowest level (0) for the voltage of the speaker.

The maximum O<sub>2</sub> level in the mixture was 30% ( $\Omega = 0.30$ ), which is the upper limit of the low enrichment in the OEC (Baukal, 1998). For low levels of enrichment, no major modifications to the equipment are required for the application of the OEC technique; consequently, the conditions of zero enrichment and 21% ( $\Omega = 0.21$ ) of O<sub>2</sub> from the atmospheric air were considered to be low levels of enrichment.

To characterize the limits of the ratio of equivalence ( $\phi$ ), lean combustion conditions ( $\phi = 0.7$ ) and rich combustion conditions ( $\phi = 1.3$ ) were considered for the lower and upper levels. This allowed the behavior in these two basic combustion conditions to be evaluated. These conditions were reached in the burner by varying the oxidant flow rate and maintaining a constant fuel flow rate. Nine conditions were tested for the burner with the maximum and minimum limits for each controllable variable including the center point (Table 1). All tests were done in triplicate, totaling 27 experiments.

**Table 1**  
Experimental levels employed for screening design.

Experiment	Equivalence ratio	Voltage range	Enrichment level
1	1.3	5.3	0.30
2	1.3	0	0.30
3	1.3	5.3	0.21
4	1.3	0	0.21
5	0.7	5.3	0.30
6	0.7	5.3	0.21
7	0.7	0	0.30
8	0.7	0	0.21
9 (CP)	1	2.65	0.25

CP = central point

**Table 2**  
Technical specification of the natural gas.

Characteristics	Natural gas
Chemical composition	88.82% CH <sub>4</sub>
	8.41% C <sub>2</sub> H <sub>6</sub>
	0.55% C <sub>3</sub> H <sub>8</sub>
	1.62% N <sub>2</sub>
	0.6% CO <sub>2</sub>
Specific mass ( $\rho$ )	0.62 kg/m <sup>3</sup>
Dynamic viscosity ( $\mu$ )	$0.10816 \times 10^{-3}$ P
Calorific value	9400 kcal/m <sup>3</sup>

The calculations of the stoichiometric variables were performed considering the design data of the experimental device and the technical characteristics of the fuel used, which was natural gas supplied by Bahiagás. The chemical composition and physical properties of the gas are shown in Table 2.

For the calculation of the fuel flow rate, the Reynolds number was considered to be  $Re = 4100$ , where the flame is in the transition zone between laminar to turbulent flow. So, the natural turbulence due to the fuel flow did not have an impact on the results. For this purpose, the fuel flow rate was kept constant (20.2 L/min) under all of the tested conditions, and we varied the flow rate of the oxidant to reach the desired conditions. Table 3 shows the parameters calculated by stoichiometric reactions between the fuel and oxidant.

### 3.2. CO and NOx emissions and exhaust temperature

Table 4 presents the values obtained for the response variables (CO, NOx and exhaust temperature) for the 27 tests performed (triplicates of the 9 conditions tested). The means and standard deviations of the response variables for each test condition are presented. An analysis of the data shows good uniformity, which is reflected in the acceptable standard deviations.

Individual analysis of each response variable provided the results presented in Table 5. The  $R^2$  values for the regressions of the data were satisfactory (above 88%). The highest coefficient of determination was obtained for CO (99.98%), and the lowest was obtained for exhaust temperature (88.25%). The  $R^2$  values indicate that the applied statistical regressions accurately represent the measured experimental data.

The Pareto plot of the standardized effects illustrates how the controllable parameters (ratio of equivalence, voltage signal and enrichment level) and their combinations influenced the values of the response variables (CO, NOx and exhaust temperature). Fig. 4 shows the Pareto graphs for the tests performed for each response.

For the CO emissions, all of the factors and their combinations had significant influences on the results. For the NOx emissions, only the enrichment level and the ratio of equivalence had strong influences, which is consistent with the results of Cardu and Baica (2005). These authors demonstrated a significant dependence of

the thermal NOx concentration on the atomic oxygen concentration and the flame temperature. The exhaust temperatures were influenced most by the ratio of equivalence and by its combination with the enrichment level.

The individual factors in the Pareto graphs show that of the three controllable variables, the input voltage signal on the speaker had the least influence on all of the response variables. This may have occurred because the voltage was limited to 5.3 V, which reflects a sound pressure of 8.8 mbar in the oxidant feed. Due to the low fuel combustion conditions ( $\phi = 1.3$ ) and the lack of oxygen enrichment ( $\Omega = 0.21$ ), the phenomenon known as “lifted flame” occurred at voltages above this limit. Operations under lifted flame conditions are undesirable in industrial burners due to the loss of efficiency and the instability of the flame, which can be extinguished. For the other tested conditions of low fuel combustion ( $\phi = 0.7$ ) or oxidant enrichment levels with O<sub>2</sub>, it was possible to achieve higher input voltage values in the speaker.

To determine the behaviors of the response variables with variations of the main factors, graphs of the main effects were generated for each response (Fig. 5).

The graphs of the main effects show a decrease in the CO concentration with increasing O<sub>2</sub> enrichment due to the greater availability of oxygen molecules for the formation of CO<sub>2</sub>, which contributes to complete combustion. The same result occurred with an increase of excess air in the combustion, which is represented by a reduction of the ratio of equivalence. Although the voltage signal in the speaker had a smaller influence on the response variables, it generally contributed to reductions of the emissions of both CO and NOx. This phenomenon was also observed by Kim et al. (2009) and was attributed to the fact that the sound excitations improve the mixing rates between the fuel and the oxidant.

In general, the results for the NOx concentrations were opposite to those of the CO concentrations, where there was an increase in NOx with increasing percentage of O<sub>2</sub> in the oxidant and a reduction with an increase in the ratio of equivalence. According to Cohen et al. (1996), this behavior occurs because the thermal NOx depends on the relation between the flame temperature and the ratio of equivalence. Thus, it is necessary to study these two pollutants together.

An expected reduction in the exhaust gas temperature occurred under the poor combustion conditions ( $\phi = 1.3$ ) due to the incomplete combustion conditions provided and evidenced by the high CO emissions, which are conditions that reduce the energy expenditure by the chemical reactions. A small increase in the average exhaust gas temperature was observed with the inclusion of the acoustic field, which can be explained by the increase in convective heat transfer at the gas outlet caused by the sound excitation. In addition, the temperature of the exhaust gases decreased when the oxidant was enriched with O<sub>2</sub>. According to Santos et al. (2009), this can occur when the application of OEC induces the formation of soot in the flame; because soot enhances heat transfer by radiation in the combustion chamber, less energy is provided by the exhaust gases.

Fig. 6 shows contour plots of the voltage applied to the speaker and the oxidizer enrichment level to examine the relationships between the response variables and the two main controllable input factors that characterize the combustion techniques. The graphs were generated from the calculated regression equations.

Fig. 6a shows reduction in CO concentrations for conditions without oxygen enrichment ( $\Omega = 0.21$ ) and with sound excitation, but we could not observe CO reductions caused by the acoustic field in regions with oxidant enrichment. This occurs because these regions already have low CO concentrations even without sound excitation.

**Table 3**

Parameters calculated to perform the experiments.

Fuel flow = 20.2 ± 0.5 L/min		
Equivalence ratio ( $\phi = 1$ )	Atmospheric air flow (L/min)	Pure oxygen flow (L/min)
Enrichment level (25%) ( $\Omega = 0.25$ )	191.2 ± 0.5	10.2 ± 0.5
Equivalence ratio ( $\phi = 1.3$ )		
Enrichment level (21%) ( $\Omega = 0.21$ )	154.9 ± 0.5	0
Enrichment level (30%) ( $\Omega = 0.30$ )	137.3 ± 0.5	17.6 ± 0.5
Equivalence ratio ( $\phi = 0.7$ )		
Enrichment level (21%) ( $\Omega = 0.21$ )	287.7 ± 0.5	0
Enrichment level (30%) ( $\Omega = 0.30$ )	255.0 ± 0.5	32.7 ± 0.5

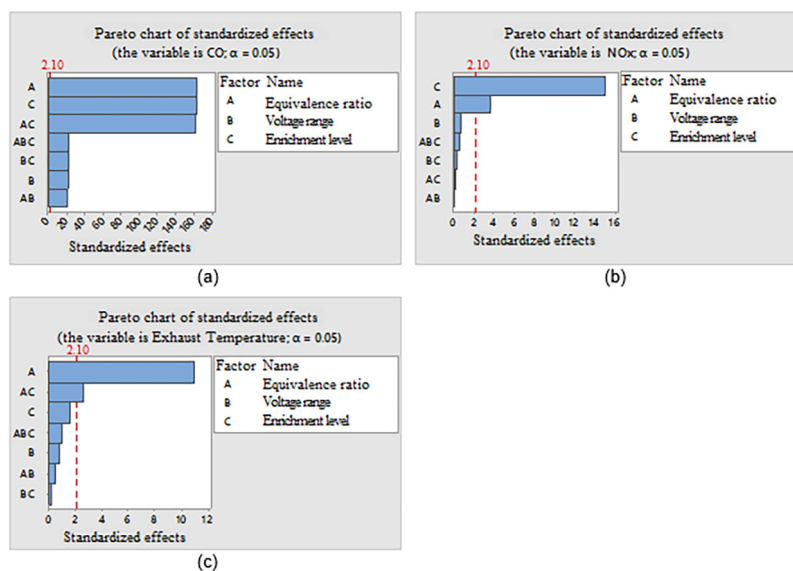
**Table 4**Results of the experiments: CO, NOx and exhaust temperature<sup>a</sup>

Test conditions	Equivalence ratio	Voltage range (V)	Enrichment level	CO (ppm)	NOx (ppm)	Exhaust temperature (°C)
1	1.3	5.3	0.30	6.3 ± 2.1	93 ± 7	207 ± 3
2	1.3	0	0.30	2.7 ± 0.5	99 ± 6	207 ± 4
3	1.3	5.3	0.21	484 ± 8.8	41 ± 7	215 ± 3
4	1.3	0	0.21	633 ± 2.5	40 ± 3	211 ± 1
5	0.7	5.3	0.30	1.3 ± 1.9	107 ± 12	228 ± 2
6	0.7	5.3	0.21	4.0 ± 2.9	53 ± 7	225 ± 3
7	0.7	0	0.30	2.3 ± 0.5	109 ± 11	226 ± 2
8	0.7	0	0.21	3.7 ± 1.2	56 ± 2	226 ± 1
9	1	2.65	0.25	5.0 ± 0.8	71 ± 2	220 ± 5

<sup>a</sup>Mean ± Standard deviation.**Table 5**

Regression equations for variable response of CO and NOx emissions and exhaust temperature.

Variable response	R <sup>2</sup> (%)	Regression equation
CO	99.98	$CO = -2438.4 + 3493.1\phi + 112.13V + 8134.6\Omega - 159.26\phi * V - 11642.0\phi * \Omega - 377.8V * \Omega + 535.8\phi * V * \Omega$
NOx	93.12	$NOx = -34.0 - 48.1\phi - 6.0V + 515\Omega + 6.9\phi * V + 105\phi * \Omega + 22.4V * \Omega - 28.0\phi * V * \Omega$
Exhaust temperature	88.25	$Exhaust\ temperature = 228.9 - 5.7\phi - 5.26V + 64.2\Omega + 5.87\phi * V - 86.4\phi * \Omega + 19.64V * \Omega - 21.0\phi * V * \Omega$

 $\phi$  = Equivalence ratio;  $\Omega$  = Enrichment level; V = Voltage range.**Fig. 4.** Pareto charts: (a) CO emission; (b) NOx emission; (c) exhaust temperature.

One of the main effects of the combined use of the OEC and pulsating flame techniques is the possibility of reducing NOx concentrations in  $O_2$  enriched flames with the use of sound excitations to the flame (Fig. 6b). This effect can be explained by the homogeneous turbulence throughout the flame allowing an equalization and a lower mean flame temperature, whereas the application of the OEC technique alone results in regions of

the flame with extreme temperature peaks that contribute to the formation of thermal NOx.

In general, an increase in the temperature of the exhaust gases was observed with the application of the acoustic field to the combustion (Fig. 6c), mainly in the zone of combustion that is rich in fuel. This is due to the increase in convective heat transfer at the gas outlet as well as increases in the rates of the chemical

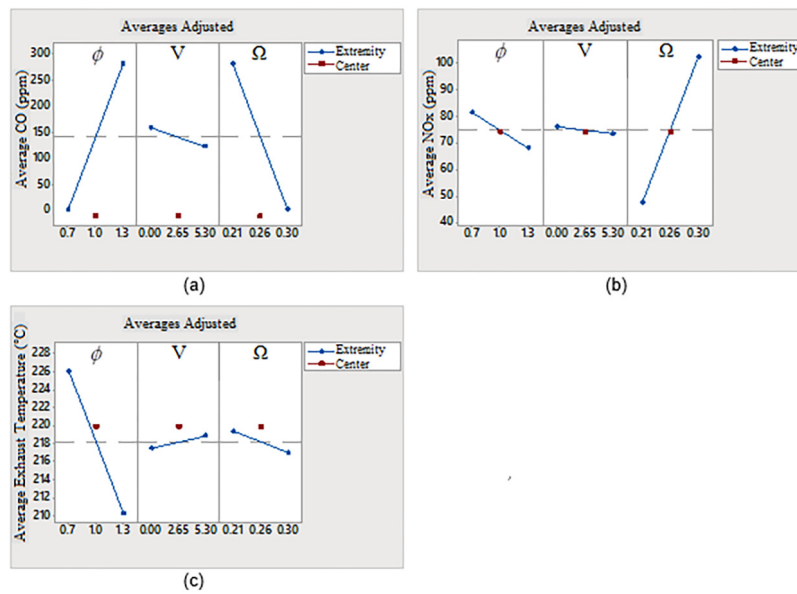


Fig. 5. Graphs of the main effects for (a) CO emission; (b) NOx emission; (c) exhaust temperature.

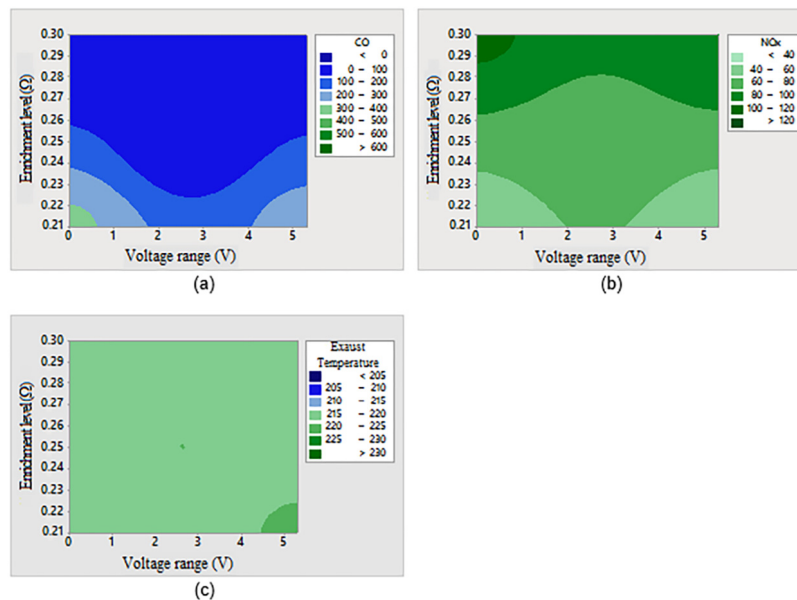


Fig. 6. Contour graphs of enrichment level vs. voltage for (a) CO emission; (b) NOx emission; (c) exhaust temperature.

reactions caused by the increase in the average turbulence in the flame region, which is characteristic of the pulsating combustion technique according to [Carvalho et al. \(1987\)](#).

### 3.3. Formaldehyde and acetaldehyde emissions

Fig. 7 shows the concentrations of formaldehyde and acetaldehyde at the outlet of the combustion chamber for each test condition (Table 1). Acetaldehyde was emitted in greater quantities in all of the experiments. The most acetaldehyde was emitted in experiment 6 (using bigger voltage range and smaller equivalence ratio and enrichment level), and the greatest emissions of formaldehyde occurred in experiment 1 (using bigger equivalence ratio and smaller voltage range and enrichment level).

An analysis of the experiments in which the equivalence ratio was modified showed a reduction in the emission rates of acetaldehyde with increasing ratio of equivalence. This was also

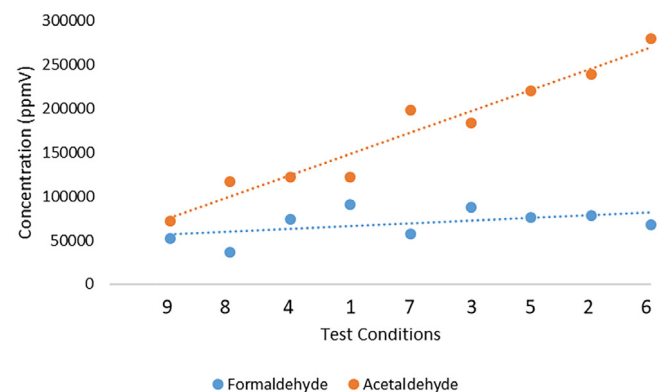


Fig. 7. Concentrations of the aldehydes (ppmV) in according to the collection parameters in each test condition (as listed on Table 1).

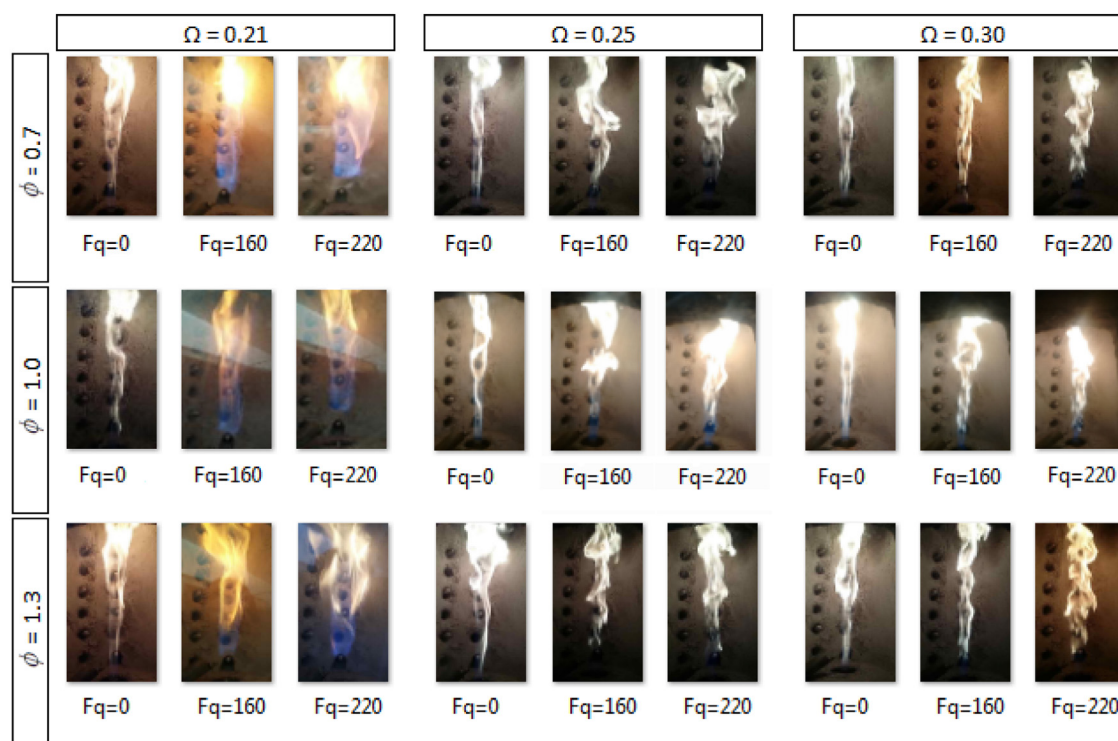


Fig. 8. Images of the flames under different test conditions.

observed by Santos et al. (2016) and occurs because the lower concentration of oxidant available in the oxidant/fuel mixture reduces the rate of aldehyde formation by direct oxidation of hydrocarbons.

We observed that there was not a clear decreasing trend of the emissions of aldehydes as a function of the amplitude.  $O_2$  enrichment did not cause an increase in the emissions of aldehydes as occurred in Santos et al. (2016).

Acoustic excitation increased the acetaldehyde emissions regardless of the equivalence ratio and enrichment level used (Fig. 7). This result may be related to the changes in the aerodynamic characteristics of the oxidant flow with the excitation due to the increased turbulence, which generates a preferential path for the formation of that compound. This trend was not verified in the formation of formaldehyde, which may have been because some aspects of the reaction had greater effects than the turbulence aspects on the formation of this compound.

The formaldehyde/acetaldehyde ratio was considered as a function of the three parameters studied. The ratio varied between 0.2 and 0.7 with a mean of 0.45. This mean ratio of less than 1.0 indicates that the acetaldehyde emissions were higher than the formaldehyde emissions, for all conditions studied.

Natural gas is composed mainly of methane; consequently, its oxidation will generate formaldehyde with a carbonyl functional group. Formaldehyde/acetaldehyde ratios below 1.0 are likely caused by decreases in the formation and emissions of formaldehyde under these combustion conditions. This provides an environmental benefit because this compound is highly toxic when present in the atmosphere and participates in the formation of ozone and photochemical oxidants (Guarieiro et al., 2008a).

#### 3.4. Dynamic aspects of the flames

Fig. 8 shows images of the flames with various combinations of the input variables. The voltage was set to the maximum possible, and the sound frequency (Fq) was varied. For conditions without

oxygen enrichment ( $\Omega = 0.21$ ), there was an increase in the instability of the flame with an increase in the sound frequency. It did not remain attached to the base of the burner, which is a condition known as a “lifted flame” that often leads to the flame dying out.

When the mixture was enriched with oxygen, the attachment of the flame significantly improved when the flame was excited with sound waves. This was possibly due to the OEC technique, which increases the flammability limits of the fuel/oxidant mixture. Additionally, we observed characteristics that include the increase in the apparent turbulence of the flame with the introduction of the acoustic field, the bluish color in some regions of the flame, and the absence of soot. In rich conditions ( $\phi = 1.3$ ) and without  $O_2$  enrichment, the mechanism of soot formation through oxidative pyrolysis was not potentiated, giving a bluish appearance to the flame root. In the case of lean conditions ( $\phi = 0.7$ ) with  $O_2$  enrichment, the mechanism of oxidative pyrolysis was higher in this case, producing a greater amount of soot and, therefore, the flame with a luminous appearance.

#### 4. Conclusions

In this study, the flame without enrichment in fuel-rich combustion was more unstable in the presence of forced excitation, which greatly limited the range of voltages used in the experiment, and the instability was controlled by enriching the oxidant with  $O_2$ . Instability in diffused flames can lead to a loss of efficiency and can even cause the flame to die out.

The results of the experiment were consistent with low variations between the obtained values. The modeling of the response variables obtained  $R^2$  values higher than 88%, which indicates good reliability of the models.

A general reduction in CO emissions was observed with the enrichment of the oxidant with  $O_2$  and in some conditions with an acoustically excited flame. The  $NO_x$  concentrations decreased in flames enriched with  $O_2$  when sound excitations were applied

to the flame. The NO<sub>x</sub> concentration varied inversely with that of CO; under the operating conditions, a smaller emission of carbon monoxide were associated with a biggest emission of nitrogen oxide. This behavior reveals the importance of determining operating conditions for combustion devices that result in the optimum emissions of pollutants.

The concentrations of formaldehyde and acetaldehyde under the tested conditions did not decrease with the application of the acoustic excitation associated with the enrichment of O<sub>2</sub>. However, the acetaldehyde emissions decreased when the equivalence ratio increased.

The study of the combined effects of the two techniques presents a promising opportunity to increase the efficiency and reduce the emissions of pollutants from existing industrial facilities due to the minor changes required to implement the techniques. Besides that, this study suggest that the replacement of the installed burners is not necessary when these minor changes are applied.

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