Pollutant emissions analysis of a natural gas gliding arc plasma assisted combustion system under fuel-rich condition

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Abstract

The recently increasing interest in plasma assisted combustion is motivated by new possibilities for ignition and flame stabilization, in addition to pollutant emission reduction and control. In a chemical active environment the plasma generates radicals, excited chemical specimens and ions thus increasing the combustion process reaction rate.

In this work the effect of plasma assisted combustion on pollutant emissions of a premixed flame of natural gas-air is investigated by using a two gas analyzer systems to acquire a diversity of pollutant gases.

The plasma is created by using a gliding-arc discharge, chosen because of its properties: works at atmospheric pressure, generates high electron density plasma and has chemical selectivity. The reactor geometry limits the equivalence ratio range of stability between $\phi = 0.3 - 0.7$ and $\phi = 1.2 - 1.4$. Within these ranges the equivalence ratio of $\phi = 1.2$ was chosen for investigation. The variable in this study is the gliding arc electrical power source ranging between 220W and 370W.

Results show the behavior from fundamentals components of natural gas combustion, as CH_4, nitrous oxide components, carbon monoxide, carbon dioxide and others. It was seen that relations between gas temperature, input electrical power and chemical dynamics influence concentrations of pollutant gases presented at the acquired sample. Also, it is seen how this vortex gliding arc architecture works in combustion terms.

Introduction

In recent years, the increasing attention to pollutant emissions and the stringent international regulations on this subject is forcing combustion engineers to develop and apply new technologies which improve the efficiency of combustion processes while reducing pollutant emissions. Among them the catalysis of chemical reactions using electric discharges generating plasma in addition to the combustion process is becoming a very promising area of science and technology [1]–[4]. This area, called assisted plasma combustion, has possibilities of industrial applications, especially in the chemical industry and for energy generation. Example of applications in the chemical industry is the production of chemical products with high commercial value like syngas (H_2 + CO) and pure hydrogen [5]–[7]. In the energy production industry the use of the Rich-Quenching-Lean (RQL) concept is favored by the presence of an electrical discharge in the rich zone which avoids soot formation and provides enough energy for combustion to sustain itself even in very rich mixtures.

The type of discharge chosen for plasma assisted combustion depends on the nature of the combustion process under consideration. For the applications earlier described the discharge should occurs at atmospheric pressure, providing a high electron density and stimulates certain degree of freedoms to obtain chemical selectivity (like non-thermal plasma) (Sagás et al 2011). Some types of discharges, like glow discharges (operates at higher pressures) and thermal arcs (do not allow chemical selectivity) are excluded due to the required combustion process conditions. Thus, for this applications the suitable discharger are the dielectric barrier and the microwave discharges along with non-thermal arcs. In the last group, the gliding-arc discharge has gained interest for plasma assisted combustion due to its properties [5], [9].

They operate at atmospheric pressure, the discharge is obtained between divergent electrodes (the gap between electrodes increases in the gas flow direction), and can operate in different regimes: thermal, transitional and non-thermal regimes [10]. Especially important for plasma assisted combustion is the transitional regime, where the discharge starts like a thermal arc. The arc is pushed and elongated by the gas flow, thus increasing the discharge voltage and power until the discharge makes a transition to a non-thermal regime when the power reaches its maximum. When the voltage reaches the breakdown value, a new arc is created in the smallest gap between the electrodes, the previous arc extinguishes and the cycle restarts [5], [10], [11].

What is believed to be the major difference between conventional and plasma assisted processes is the presence of different chemical routes opened by species generated by electrical discharge "side effects" like dissociation, excitation and radical formation [12]. Certain species can form much more quickly in the presence of an electric discharge, increasing the process reaction rate. For processes involving hydrocarbons, high reaction rates is attractive due to the speed and efficiency with which the chains are broken,

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fragmenting fuel molecules into smaller (usually more reactive) molecules and accelerating the process.

For plasma assisted combustion the formation of radicals and ions is of great importance [13]. Among the mechanisms proposed, it is suggested that the reactions can be accelerated by the breakage of the methane molecules and formation of CH3, CH2, CH and O radicals, with formation of intermediate species such as CH2O and CH3O and subsequent formation of H2 and CO. Other models suggest the formation of acetylene and ethylene as intermediate species [13].

An important aspect of discharges in rich and very rich fuel mixture is the absence of soot reported in the literature [12]. So far this is attributed due to the highly acidic nature of the discharge in the presence of air. A highly oxidative environment leads to carbon oxidation, avoiding soot formation. Obviously, the above process depends on the concentration of water vapor.

Thus, we propose the use of plasma out of thermodynamic equilibrium, in particular the gliding arc discharges in cases of assisted combustion and partial oxidation of natural gas, with the aim of studying the fundamental aspects of the process, particularly as the electrical discharge chemically affects the combustion process through pollutant emission gas analysis.

Given this, it was realized an analysis of natural gas (NG) plasma assisted combustion in fuel rich condition ϕ = 1.2. For that, the main parameter analyzed was the input electrical power, ranging it in values from 220W to 370W. It was intended to see the behavior of hydrocarbons (Ethane, Propane and CH_4), nitrous oxide and derivations (NO, NO_2, NOx), carbon monoxide (CO) and carbon dioxide (CO_2). This enables to see characteristics of the chemical dynamics of the interaction of plasma discharges in natural gas plus air combustion.

Experimental setup and methodology

The experiments were conducted at the Laboratory of Combustion, Propulsion and Energy facility at the Technological Institute of Aeronautics, São José dos Campos, Brazil.

The experimental setup is composed of 3 parts: the plasma reactor, the exhaust gas analysis system and the test bench. In the following they are presented in details.

1.1. Plasma reactor

The architecture of the reactor, shown in Fig. 1, corresponds to a gliding arc in tornado (GAT reactor) [14]. It comprises a quartz tube, called discharge tube, of 22.5-mm internal diameter, 25.5-mm of external diameter and 62-mm length. A high-voltage tungsten electrode with a helical profile is fixed in a metallic holder positioned at one end of the discharge tube. At the other end of the discharge tube is located a brass grounded electrode which also works as a vortex chamber. More details on the gliding arc reactor can be found in [8].

The natural gas is injected in the vortex chamber where it acquires high tangential flow velocity. It flows until the end of the discharge tube with high swirl, while returning by the central axis of the reactor due to the half-spherical geometry of the reactor's end. Additionally, the profile of the helical electrode was made according to the gas flow lines in order to minimize the flow perturbation [8].

An alternate current 60-Hz transformer of 13800 V_{rms} is used as the electrical power source. The discharge current variation is allowed by a variable resistor. Moreover, an oscilloscope of 100MHz and a digital multitester were used for the discharge voltage monitoring and for the current monitoring, respectively.

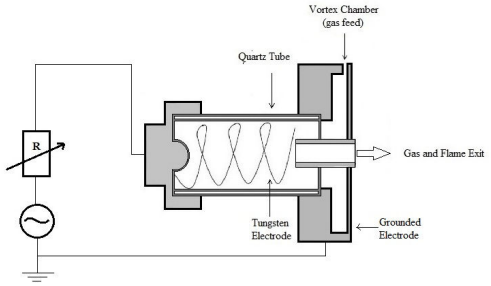

Figure 1. Plasma gliding arc reactor drawing (adapted from [8])

1.2. Exhaust gas analysis system

Two different gas analyzers are used to analyze of exhaust gas composition. One is a FTIR (Fourier Transform Infrared) gas analyzer, which provides real-time data with ppm or ppb sensitivity for multiple gas species in the rate of 1 sample per second (1 Hz). It uses an infrared optical system for acquiring the luminous intensity from components, converting this information in real-time concentration data.

The other is an analytical gas analyzer using multiple sensibility sensors for acquiring specific gases. It also has a ppm sensibility and its maximum acquisition rate is 0.25 Hz. All data are acquired via a wireless gateway to a computer attached into it.

Both gas analyzers use heated probes to avoid gas condensation of the sample gas and pumps are used to dredge the gas to the equipment's. Aiming to use both analyzers at the same time, a splitting gas device that interlinks the probes was developed. The pressure of both pumps was set to the same value for dredge the same quantity of gas. Thus, the analysis is made in the same condition and at the same time.

2.3. Test bench

The complete test bench, shown in Fig. 2, comprises the plasma reactor and the electrical source (described at topic 2.1), both FTIR and analytical gas analyzers (described at topic 2.2), an oscilloscope, a multitester, compressed air and gas supply, two mass flow meters, a post-reactor and two computers.

The chemical composition of the natural gas used in these experiments is presented in Table 1 [8].

Table 1. Natural gas composition [8]

Component	Name	Concentration
CH_4	methane	88.27%
C_2H_6	ethane	7.67%
C_3H_8	propane	1.55%
i-C_4H_{10}	i-butane	0.16%
n-C_4H_{10}	n-butane	0.29%
i-C_5H_{12}	i-pentane	0.08%
n-C_5H_{12}	n-pentane	0.065%
C_6H_{14}	hexane	0.075%
C_7H_6	heptane	0.01%
N_2	nitrogen	1.19%
CO_2	carbon dioxide	0.64%

The post-reactor consists in a metal chamber covered with a refractory material of cylindrical shape. The plasma reactor is coupled at one end of the post-reactor. Five holes on each side of the post-reactor allow access for the insertion of thermocouples and gas analyzers probes.

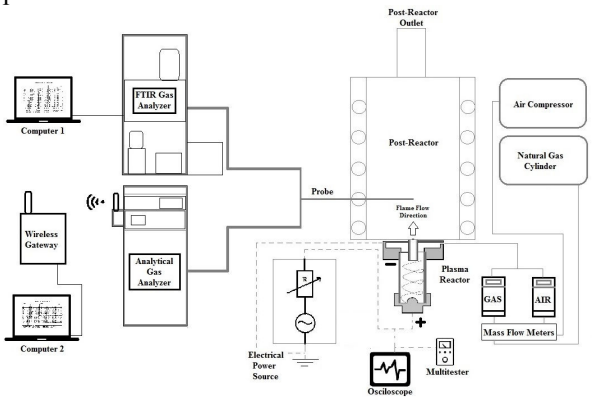

Figure 2. Test bench display

2.4. Methodology

In the present work the influence of plasma assisted combustion of premixed natural gas-air is investigated by means of exhaust gas analysis using the FTIR gas analyzer and the analytical Gas Analyser (described in section 2.2). The reactor architecture allows stabilized flames in the range of equivalence ratio between $\phi = 0.3 - 0.7$ and $\phi = 1.2 - 1.4$. Tests were carried out at constant equivalence ratio of $\phi = 1.2$ due to the interest in applying plasma assisted combustion in the rich region of RQL combustors. Previously research carried out by [23] measured the influence of the equivalence ratio and input power at poor equivalence ratio conditions. For reaching $\phi = 1.2$, it was used an air mass flow of 0.8 g/s and a gas mass flow of 0.056g/s.

Given this requirement, the main parameter analyzed was the variation of input power, since it influences the ionization and oxidation of the mixture [19]. The system was tested in a range of input power between 220W to 370W. The input power is determined by information of input voltage, provided by the oscilloscope, and input current, given by the multitester. The acquired data are stored in two computers, one for each analyzer.

Results and Discussions

In this section are presented and discussed the results from pollutant emissions detection by the exhaust gas analysis system using the FTIR analyzer and Analytical Gas Analyser. The following species are considered: methane (CH_4), Ethane, Propane, nitrogen oxides (NO, NO_2, NOx), carbon monoxide (CO), carbon dioxide (CO_2) and water (H_2O).

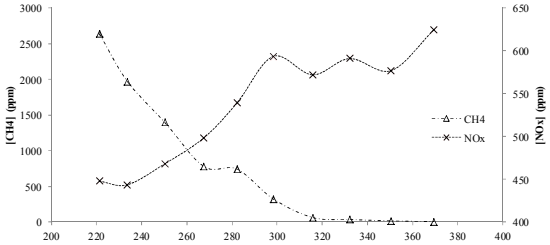

Figure 3. Behavior of [CH_4] and [NOx] with input electrical power in the exhaust gases

Figure 3 shows the behavior of CH_4 and NOx concentration in ppm, in the exhaust gas, while increasing the input electrical power. While increasing the electrical power the CH_4 concentration steeply decreases. Increasing the electrical power, the reactor nozzle temperature increases due to Joule effect. According to [18], both the gas temperature and the plasma density increase with input electrical power, allowing a higher conversion of CH_4 and higher hydrocarbons into H_2, H_2O, CO, and CO_2 and therefore a more efficient combustion [18] thus confirming our results.

The behavior of NOx concentration, as seen in Fig. 3, can be divided in three regions: the first region, between 225 W and 280 W with a steep increase, followed by a region of increase until 360 W and again a region of steep increase. According to the thermal NOx mechanism, NOx formation is proportionally correlated to temperature [15]. The steep increase of NOx concentration in the first region can be explained by the thermal NOx mechanism as a more complete combustion takes place ([CH_4] decreases) thus increasing the flame temperature. The smooth increase of NOx concentration may be explained again by thermal NOx mechanism but in this region the temperature increase is smaller.

Figure 4, shows the behavior of CH_4 and CO concentrations in the exhaust gases. The CO is present in high concentrations between 220 and 300W, where the residence time of the mixture in the plasma is lower due to lower power [16]. Increasing the power the concentration gets very low due to the higher dissociation and conversion tax.

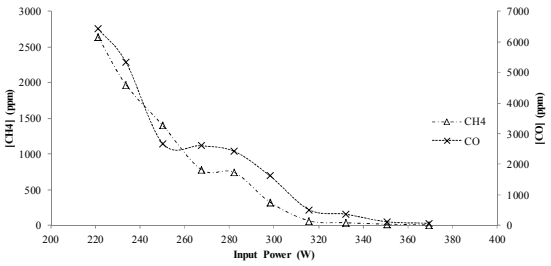

Figure 4. Behavior of [CH_4] and [CO] with input electrical power in the exhaust gases

Figure 5, shows the behavior of NOx, NO and NO_2 concentrations in the exhaust gases. The graphic shows an increase of NO concentration and a smooth NO_2 concentration increases. This occurs, again, because of the gas dissociation caused by a longer residence time of gas at the plasma. As NO is a more stable specimen, it increases as the power increase. The NOx, as it is a sum from NO and NO_2, it get an increase behavior.

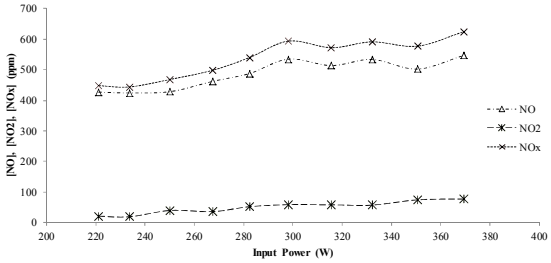

Figure 5. Behavior of nitrogen oxides [NOx], [NO] and [NO_2] with input electrical power in the exhaust gases

A relation between gas temperature, input power and electrode gap can be discussed. According to [16] and [17], a higher gap between electrodes reduces electrode temperature and increase the volume of reaction, thus increasing the conversion tax and the residence time of gas in the region of plasma influence. But, a bigger gap requires a higher power to break the dielectric resistance of the environment fluid. This produces a higher plasma density leading to a more complete burn of the mixture. Thus, an increase of the released thermal energy will occur, increasing the gas temperature and, though thermal conduction, the electrode temperature.

Figure 6 shows the behavior of concentration from methane, propane and ethane. As is seen, concentration of propane and ethane is smaller due to it concentration in the NG, which are found in smaller concentrations in it composition. Both CH_4 and the other hydrocarbons showed have the same behavior, following a bigger dissociation as the input power is increased.

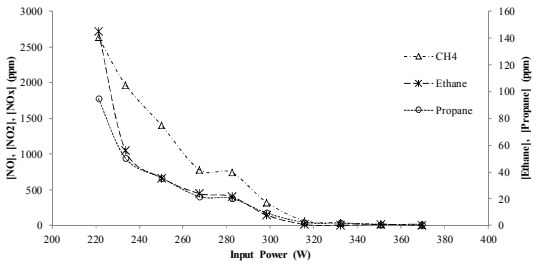

Figure 6. [CH_4], [Ethane] and [`Propane] concentrations in exhaust gases

Figure 7 shows the concentration behavior of CO, CO_2 and H_2O in exhaust gases. CO_2 concentrations show a steep increasing in the region between 300 W and 380 Was result of a more complete combustion due to higher electrical power. For a further increase of the power, CO_2 concentrations maintain an almost stable concentration. But, CO concentration decreases, result of a more complete burning of fuel, following the same behavior from the hydrocarbons previously analyzed. The H_2O follow a behavior similar to the CO, but it decrease is related to chemical balance.

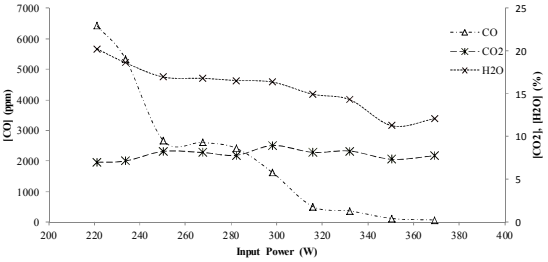

Figure 7. Behavior of [CO], [CO_2] and [H_2O] with input electrical power in the exhaust gases

Conclusions

In this study a gliding arc discharge for plasma assisted combustion of NG at rich-fuel condition was studied using the input electrical power as main parameter for analysis of the pollutant emissions concentration.

It is highlighted that the chemical dynamics of this type of plasma reactor is related to the reactor temperature, relating it to the dissociation level, conversion tax, elements selectivity and residence time.

Increasing the input electrical power especially in the range between 350 W and 390 W a more complete combustion is observed. The Ethane, Propane and CH_4 concentrations decrease in this region while CO_2 and concentrations steeply increase. The temperature increase due to a more complete combustion leads to an increase of NOx concentration. Moreover, a further increase in power doesn't change significantly the concentrations of this species except for NO which smoothly increases while reactor temperature increases.

From this, it is observed a great concentration of hydrocarbons at the exhaust of the reactor and an increase in NO concentration. This process opens a new route for application for production of syngas from NG and use in RQL combustion chamber, due to it balance between emissions. A control of the emissions can be done using a control of input power. This way a better balance of emissions can be reached. As seen the conciliator pollutant emission point is around 280W and 300W. For future applications, this architecture plasma reactor can be used for emission control in combustion chambers, in general.

For a better characterization of the combustion phenomena in the reactor, next steps of the research is to use emission spectroscopy for evaluate the formation of radicals in the reactor's combustion chamber, where the plasma occur, and in the flame. Also it is intended to

use optical high speed techniques to watch the formation of the flame and plasma.

Acknowledment
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