Nonconventional Technologies Review
Romania, June, 2021

USE OF HYDROGEN AS A NONCONVENTIONAL ALTERNATIVE FUEL IN COMBUSTION PROCESSES WITH LOW NO_X EMISSIONS

Lucian Paunescu[1], Gheorghe Surugiu[2] and Marius Paul Munteanu[3]
[1] Bilmetal Industries SRL Popesti Leordeni-Ilfov, Romania, lucianpaunescu16@gmail.com
[2] State Inspection for the Control of Boilers, Pressure Recipients and Lifting Installations Bucharest, Romania, gsurugiu2000@yahoo.com
[3] Metallurgical Research Institute Bucharest, Romania, mmunteanu53@gmail.com

ABSTRACT: The paper presents an experimental oxyhydrogen burner with a thermal power of 31.5 kW (9 m^3N hydrogen/h) designed for industrial use at high temperature with NO_x emissions at a relatively low level. The method adopted for the design consisted in mixing hydrogen with oxygen in two successive stages, the concentration of oxygen in the last stage being reduced below 20%. By this method, the flame temperature was reduced to around 1800 ºC allowing the decrease of NO_x (measured as NO_2) emissions below 240 mg/m^3N, i.e. below the maximum allowed limit (300 mg/m^3N) in the case of natural gas combustion with air. The advantage of using this "clean" alternative fuel under the conditions of operation only with oxygen is removing the possibility of polluting the atmosphere with greenhouse gases (CO_2), the waste gases released being only water vapor.

KEYWORDS: burner, alternative fuel, hydrogen, oxygen, nitrogen oxides, high temperature.

1. INTRODUCTION

Nitrogen oxides (NO_x) are a group of oxides composed of seven compounds (N_2, NO, N_2O_2, N_2O_3, NO_2, N_2O_4 and N_2O_5), of which nitrogen dioxide (NO_2) is the most important form of NO_x in the atmosphere. It is not only an important air pollutant, but can also generate so-called "acid rain". According to US Environmental Protection Agency [1], the limit level of NO_2 concentration in the atmosphere for health protection was allowed at 0.053 parts per million (ppm), i.e. 0.1 mg/m^3.

The internal combustion engine of the cars contributes with approx. 50% of the global NO_x emission in the atmosphere and electric power plants contribute with about 20%. Additionally, large quantities of NO_x come from industrial thermal boilers, incinerators, gas turbines, Diesel engines, cement manufacturing, glass manufacturing, oil refining, nitric acid manufacturing, etc. [1].

NO_x formation in combustion processes occurs mainly in the following cases. The so-called "thermal NO_x" is formed depending on the molar concentration of nitrogen and oxygen and the temperature of the combustion process. It has been found that process temperatures above 1300 ºC generate much higher concentrations of NO_x compared to thermal processes at lower temperatures. Nitrogen-containing fuels in their composition (e.g. coal), called "fuel NO_x", generate these types of pollutant by oxidation.

Another type of air pollutant (mainly CO_2) results from combustion processes of fossil fuels (coal, hydrocarbons). In recent decades there has been a tendency to replace at least partially these types of conventional fuel with alternative nonconventional fuels. According to [2], the European Union defines alternative fuels as fuels or power sources that replace at least partially fossil oil sources, referring mainly to energy supply for the transport sector. The alternative fuels include: electricity, hydrogen, biofuels (biomass, algae-based fuels), synthetic and paraffinic fuels, biomethane in gaseous or liquefied form and liquefied petroleum gas.

In the world, the use of hydrogen as an energy source is focused to two application domains: the supply of fuel cells for electricity generation and the combustion process to generate heat. One of the solutions to eliminate greenhouse gas emissions (mainly CO_2) could be the adoption of hydrogen as an alternative nonconventional fuel. On the other hand, the significant reduction of the combustion air until its complete replacement with technical oxygen, eliminates (or diminishes) the possibility of NO_x formation.

The use of hydrogen as a non-polluting energy carrier has been applied since the 1970s in the United States by blending it (in relatively low concentrations between 5-15 vol.%) in the existing network of natural gas pipelines [3]. The addition of hydrogen to natural gas significantly reduces greenhouse gas emissions.

Generally, the hydrogen production as a by-product of some industrial technological processes constitutes the main source of obtaining this gaseous fuel. The chemical industry is an important supplier of hydrogen, which is captured and introduced into the internal network of pipelines for its own ovens or for heating processes in neighboring areas as well as for bottling in pressure recipients intended for sale. Because the hydrogen has a lower calorific value (2.6-2.8 times) than the natural gas, it is used in mixed combustion installations together with natural gas, the main expected effect being to reduce the greenhouse gas emissions. Such a technical solution was applied by the team of authors of the current paper at the former Oltchim Rm.Valcea chemical plant [4, 5].

An electrolyzer patented in the United States in 2002 [6] was designed to produce a gaseous mixture containing hydrogen and oxygen in stoichiometric proportions called HHO. It has an internal structure different from the conventional molecular one, which gives it particular properties. Due to the purchase of the license for the application of HHO gas in Europe and Israel by the Romanian company Rokura Industrial Applications SRL Bucharest which allowed the use of the HHO gas generator, a team of researchers including authors of this paper have designed and tested a conventional natural gas-air burner, in which HHO gas was injected into the initiated flame. The results of these experiments published in the literature [7] and presented at an international conference [8] showed that the substitution of 30% natural gas with HHO gas led to the increase of combustion temperature from 1600 to 1890 ºC, so that the thermal effect to be similar with that of burning only of natural gas. The CO concentration in the waste gas decreased from 75 to 3 ppm.

Theoretically, it is known that in combustion processes the increase of the flame temperature at temperatures above 1300 ºC involves the NO_x formation. Except for combustion processes that take place in ovens with a controlled atmosphere (without nitrogen), in the other processes the flame produced by burning the fuel comes in contact with the atmospheric air existing in the oven. So, even if the oxidizer (combustion air) had a lower concentration of nitrogen by enriching it in oxygen, there are conditions for the NO_x formation in waste gas released in the oven, if the temperature of the combustion process is high.

Given the very high share (50%) of NO_x emissions caused by car or Diesel engines, Toyota Motor Corporation in collaboration with Chugai Ro Co. Ltd has developed for the first time in the world engines running on hydrogen (produced “in-situ”) and oxygen adopting the solution of developing hydrogen combustion in two stages to decrease the final flame temperature [9]. The concentration of oxygen in the last stage of combustion with hydrogen was thus reduced to 19% leading to a decrease in the flame temperature and a reduction of NOx emissions to the level allowed in the case of natural gas combustion (300 mg/m^3N) [10]. CO_2 emissions have been completely eliminated.

A Bunsen type burner for domestic applications has been designed and tested for operation with a stoichiometric mixture of hydrogen and oxygen [11]. Hydrogen was generated "in-situ" with an alkaline electrolyzer having a small production of maximum 3.5 L/min. The energy efficiency of the oxyhydogen burner was determined as a percentage of the thermal energy input transferred to a load in a given time interval and was 30% for a flow rate of 1.5 L/min and 76% corresponding to a flow rate of 3.5 L/min.

In 2020, FlammaTec Company designed and developed with a joint team of German and Czech researchers a series of four hydrogen/oxygen burners without carbon emissions, with a high efficiency of the combustion process, high-temperature combustion facility and flame stability. The thermal capacity of the burners was between 50-1500 kW (between 15-500 m^3N hydrogen/h). Hydrogen and oxygen supply pressures were between 20-140 mbar and 15-120 mbar, respectively [12].

Taking into account the previous experience in the field of hydrogen and oxygen-enriched air burners as well as the latest worldwide achievements on oxyhydrogen burners, the team of authors set out to make a hydrogen burner with a flow rate of 9 Nm^3/h using only oxygen as an oxidizer, without CO_2 emissions and with low NO_x emissions.

2. ADOPTION OF THE TECHNICAL SOLUTION

The experimental oxyhydrogen burner was designed to obtain a flame completely free of carbon oxides (CO_2 and CO) and, at the same time, with low NO_x emissions (favored by the high temperature developed in the combustion zone). The first objective is theoretically achievable by use of hydrogen as a gaseous fuel. By the hydrogen burning, the only resulting gaseous product is water in the form of vapors and possibly oxygen, if the burner runs with excess oxygen compared to the stoichiometric requirement (0.5 m^3N oxygen/m^3N hydrogen). The chemical oxidation reaction of hydrogen is as follows:

$$H_2 + 0.5O_2 = H_2O \tag{1}$$

The second objective can be constructively achieved by ensuring a staged distribution of hydrogen in the combustion area of the burner. It has been found that a high concentration of oxygen when burning hydrogen generates a very intense combustion, greatly increasing the temperature of the flame. To avoid this undesirable phenomenon, the solution of a hydrogen-oxygen pre-combustion was adopted by a radial distribution of hydrogen from the supply pipe before the central nozzle. The radial jets coming out through orifices provided in the pipe wall must meet the oxygen flow distributed peripherally parallel to the axis of the burner. The contact of the two fluids generates a small-scale pre-combustion in the first part of the burner combustion chamber. The effect of this pre-combustion is the decrease of the oxygen concentration in the hydrogen-oxygen mixture. Thus, the main combustion that occurs in the second part of the combustion chamber enclosure, at the exit from the burner body, benefits from a much lower proportion of oxygen (according to [9], less than 20%). Thus, the oxyhydrogen flame temperature is significantly decreased from about 2660 ºC to 1800-1850 ºC contributing to the reduction of NO_x emissions.

The constructive and functional scheme of the experimental burner designed for operation with hydrogen and oxygen is shown in Figure 1.

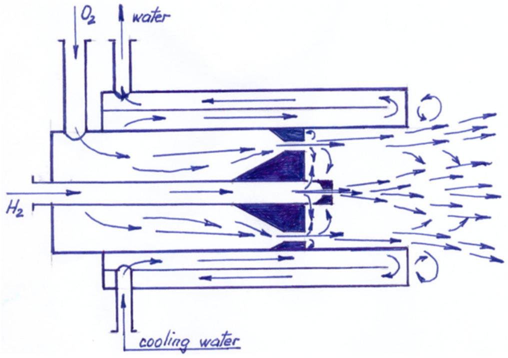


Figure 1. Constructive and functional scheme of the experimental burner

The main data used for designing the experimental burner were:

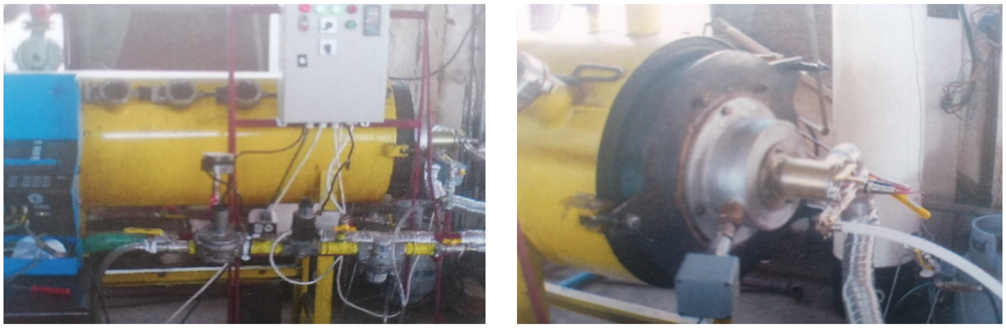
-nominal hydrogen flow rate:	9 m^3N/h
-nominal hydrogen pressure:	150 mbar
-nominal oxygen flow rate:	4.5 m^3N/h
-nominal oxygen pressure:	145 mbar
-hydrogen speed in the axial orifice:	250 m/s
-hydrogen speed in the radial orifices:	190 m/s
-oxygen speed in the peripheral axial orifices:	210 m/s
-waste gases speed at the exit of the burner:	130 m/s
-cooling water flow rate:	1 m^3/h
-cooling water speed:	1.5 m/s

3. EXPERIMENTAL METHODOLOGY

3.1 Description of the testing stand

The experimentation of the burner described above took place both at the Metallurgical Research Institute Bucharest in free space for checking the technical solution and visualizing the burner flame, as well as on the testing stand at the Technical University of Constructions Bucharest to determine its functional parameters.

The testing stand (Figure 2) has a thermal capacity of maximum 200 kW. It consists of a sealed horizontal cylindrical enclosure with an outer diameter of 500 mm and an inner diameter of 400 mm, cooled with water. The length of the stand is of maximum 1200 mm, being adjustable according to the capacity of the tested burner. Several visors are provided on the surface of the stand to visualize the characteristics of the flame. The waste gases are passed through a heat exchanger placed at the end of the stand. The cooling water circulates in a closed circuit.



a b

Figure 2. Pictures of the testing stand: a – the testing stand; b – burner mounting detail.

3.2 Method of experimentation

The measuring and control equipment related to the testing stand used during the experiments to perform the measurements of flow rates, pressures, temperatures and chemical composition of the waste gases was of the latest generation with a very good accuracy.

The method of experimentation was adopted to the concrete conditions offered by the testing stand at the Technical University of Constructions. Six thermal regimes of stabile operation of the burner between 5-9 m^{3}N/h were tested. The oxygen was used to ensure both the stoichiometric combustion of hydrogen in three of the six regimes, and the combustion with a small excess of oxygen in the other three thermal regimes. The hourly flows of hydrogen and oxygen and the pressures of these fluids at the inlet to the burner were read from the control panel of the equipment. Also, the parameter values of the cooling water were noted to determine the thermal energy quantity taken over by it and implicitly, in order to calculate the value of the waste gases temperature at the exit from the working enclosure of the testing stand. The flame length corresponding to each thermal regime was measured through visors provided on the enclosure wall. The chemical composition of waste gases was determined with the analyser existing as an integral part of the equipment, being measured the values of oxygen (vol.%), NO and NO$_x$ as NO$_2$ (ppm converted in mg/m^{3}N). The CO$_2$ and CO values (vol.%) continuously indicated their non-existence in the waste gas flow.

4. RESULTS AND DISCUSSION

4.1 Results

The experimental results obtained after testing the burner designed for operation with hydrogen and oxygen are presented in Table 1.

Table 1. Test results of the experimental oxyhydrogen burner

Hydrogen		**Oxygen**		**Waste gas composition**			**Flame tempe-rature °C**	**Flame length mm**	**Heat taken over by cooling water MJ/h**
Flow rate m^{3}N/h	Pressure mbar	Flow rate m^{3}N/h	Pressure mbar	O$_2$ %	NO mg/m^{3}N	NO$_x$ mg/m^{3}N			
9.0	150	4.5	145	-	236	242	1850	400	102.4
9.0	150	4.7	149	0.4	220	227	1835	420	101.3
7.0	105	3.5	80	-	196	205	1810	360	79.6
7.0	105	3.6	85	0.3	189	200	1800	380	78.8
5.0	60	2.5	50	-	160	184	1750	300	56.9
5.0	60	2.6	51	0.5	153	170	1730	330	56.0

Hydrogen and oxygen were used in gaseous form bottled under pressure in metal containers. Hydrogen was purchased from Linde Gaz Romania in 8.9 m^3 bottles at 200 bar, the technical oxygen being supplied by Artego in 6.35 m^3 steel containers at 150 bar.

According to the data in Table 1, the oxyhydrogen burner designed for use in industrial heating processes has an operating range under conditions of maximum flame stability between 5-9 m^{3}N/h. The pressure (at the entrance to the burner body) had values between 60-150 mbar. The hourly oxygen flow was between 2.5-4.5 m^{3}N/h with the supply pressure between 50-145 mbar.

Practically, three hourly flows of hydrogen were used: 5; 7 and 9 m^{3}N/h, the oxygen flows being at stoichiometric values (2.5; 3.5 and 4.5 m^{3}N/h, respectively) or at slightly higher values corresponding to excess oxygen coefficients of 1.03-1.04 (2.6; 3.6 and 4.7 m^{3}N/h, respectively).

Due to the application of the hydrogen distribution technique in the combustion zone with oxygen, the waste gases temperature at the exit of the burner had significantly lower values (between 1730-1850 ºC) compared to the maximum value of the oxyhydrogen flame temperature (2660 ºC) [14] . The effect of the temperature decrease on the NO$_x$ emissions value was their reduction to values between 170-242 mg/m^{3}N, i.e. below the maximum allowed limit for the emissions resulting from the combustion of natural gas with air [10].

The appearance of the flame during the experiment was examined through the visors of the test stand. A partial image of the flame corresponding to the maximum thermal regime (with 9 m^{3}N hydrogen/h and 4.5 m^{3}N oxygen/h) is presented in Figure 3. The radiant aspect of the flame is obvious and is due to the water vapor in the waste gas composition.

A more eloquent image of the flame made for the same thermal regime was obtained during the preliminary experiment performed in free space (Figure 4). Thus, the dimensions of the flame could be more easily identified, which at the maximum regime reaches 400 mm as well as the positioning of the thermal zones. A lower temperature zone can be distinguished at the base of the flame corresponding to the pre-combustion stage and a maximum temperature zone corresponding to the main combustion stage. The frontal area is a buffer zone between the flame and the oxidizing environment, in which the temperature decreases.

Figure 3. Image of the burner flame (through the visor of the testing stand)

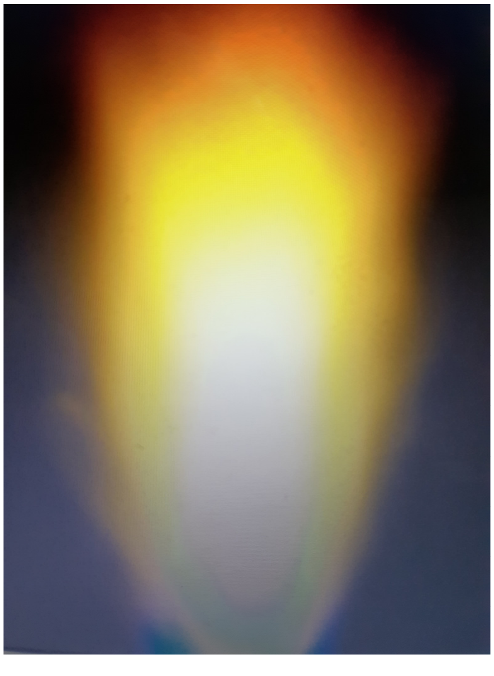


Figure 4. Picture of the burner flame (viewed during the burning in free space)

4.2 Discussion

Reducing NOx emissions from fuels combustion has been a concern of researchers around the world for 3-4 decades. Methods for reducing this pollutant have been identified and applied in fossil fuel combustion processes. The technique of mixing fossil gas and combustion air in stages is known, being one of the ways to reduce the flame temperature and implicitly, NO_x emissions [13]. However, its application to oxyhydrogen burners is a technical novelty. According to [9], this technique was first used in the world by Toyota Motor Corporation in collaboration with Chugai Ro Co. Ltd in 2018, for applications in the field of internal combustion engines of automobiles.

Oxyhydrogen burners made by FlammaTec [12] in 2020 are destined to industrial heating installations, the hydrogen flow rate range being between 15-500 m^3N/h, but there is no information in literature on the NO_x level in the released waste gas.

Therefore, the experiments performed by the authors have an originality character by the destination of the designed and tested equipment (industrial heating installations). In the perspective of increasing the thermal power of oxyhydrogen burners, a great disadvantage exists in Romania by closing the chemical plants (e.g. Oltchim Rm. Valcea) that could have provided a hydrogen supply at the necessary flow rates from the internal plant pipelines network.

5. CONCLUSION

The objective of the paper was to create an oxyhydrogen burner for industrial heating processes without greenhouse gas (CO_2) emissions and with relatively low NO_x emissions despite the very high temperature of the hydrogen combustion process with oxygen.

A technique for reducing the combustion process temperature was adopted in order to reduce NO_x emissions by ensuring two combustion stages between hydrogen and oxygen: a pre-combustion at the base of the combustion chamber of the burner and then the main combustion in the second part of the combustion chamber continued at the exit of the burner body. The effect of this distribution mode of fuel and oxygen is to reduce the concentration of oxygen in the fluids mixture below 20 vol.% and to decrease the flame temperature at the exit of the burner.

This technique was known as one of the ways to reduce NO_x emissions in the combustion process of fossil fuels with combustion air. Its application in the case of hydrogen and oxygen was made for the first time in the world by Toyota Motor Corporation in 2018 for internal combustion engines of automobiles.

The experiments took place at the Technical University of Constructions Bucharest on a testing stand as well as at the Metallurgical Research Institute Bucharest in conditions of combustion in free space. Hydrogen and oxygen were used in gaseous form bottled under pressure in metal containers.

Six thermal regimes of stabile operation of the burner between 5-9 m^3N/h were tested, the oxygen/hydrogen ratio being stoichiometric or with a low oxygen excess coefficient (1.03-1.04).

The experimental results showed the total lack of greenhouse gases (CO_2) in the waste gas composition and NO_x emissions between 170-242 mg/m^3N (below the maximum allowed limit for these emissions resulting from the combustion of natural gas with air). The waste gas temperature at the exit from the burner had values between 1730-1850 ºC due to the application of combustion technique in stages of the two fluids.

6. REFERENCES

1. *Nitrogen oxides (NO_x), why and how they are controlled,* Clean Air Technology Center, US Environmental Protection Agency, North Carolina, United States, November (1999).
2. *Directive 2014/94/EU of the European Parliament and of the Council of 22 October 2014 on the deployment of alternative fuels infrastructure,* Brussels, (2014).
3. Melaina, M.W., Antonia, O., Penev, M., Blending hydrogen into natural gas pipeline networks: A review of key issues, Technical Report, National Renewable Energy Laboratory Denver, United States, March (2013).
4. Paunescu, L., Surugiu, G., Dica, C., Iorga, G., Necula, H., Use of hydrogen in combustion installation for chemical industry, Proceedings of the *8th IASTED International Conference on Power and Energy Systems* (EuroPES 2008), pp. 608-013, Corfu, Greece, June 23-25, (2008).
5. Paunescu, L., Surugiu, G., Dica, C., Darie, G., Panaitescu, R., Reducing the concentration of greenhouse gases by partial replacement of hydrocarbons with hydrogen in industrial installations, Proceedings of the *Conference of Excellence-Favorable premise for the development of the Romanian research space,* Brasov, Romania, October 24-26, (2007).
6. Klein, D., Hydrogen generator for uses in a vehicle fuel system, US6866756B2, (2002).
7. Paunescu, L., Surugiu, G., Dica, C., Stanescu, P.D., Iorga, G., Necula, H., Ivan, I., Aspects regarding the use of Klein gas (HHO) in combustion processes, *Termotehnica,* Vol. 32, No. 1-2, pp. 24-28, (2005).
8. Paunescu, L., Surugiu, G., Melinte, I., Dica, C., Stanescu, P.D., Iorga, G., Use in combustion processes for a new type of gaseous fuel based on hydrogen, Proceedings of the *2nd IASME/WSEAS International Conference on Energy & Environmental* (EE07), pp. 99-103, Portorose, Slovenia, May 15-17, (2007).
9. Toyota develops general-purpuse hydrogen burner for industrial use; NO_x emissions below natural gas burner levels, zero CO_2, *Green Car Congress Energy, Technology, Issues and Policies Sustainable Mobility,* November 09, (2018). https://www.greencarcongress.com/2018/11/20181109-toyotah2.html
10. Emission limit values: Combustion plants in EU and USA. https://www2.dmu.dk>docs>ELV_combustion
11. Moreno-Soriano, R., Soriano-Moranchal, F., Flores-Herrera, L.A., Sandoval-Pineda, J.M., Gonzales-Huerta Guadalupe, R., Thermal efficiency of oxyhydrogen gas burner, *Energy,* Vol. 13, pp. 1-11, (2020). http://www.doi.org/10.3390/en13205526
12. FT Hydrogen CF™ Burner-Carbon free, 2020. https://www.flammatec.com/services-products/burners/gaseous-fuels/hydrogen-carbon-free
13. Gaba, A., Paunescu, L., Combustion installation with high output and low NO_x emissions, Proceedings of the *National Energy Conference* CNE'98, pp. 118-123, Neptun-Olimp, Romania, June 14-18, (1998).
14. Care este temperatura tipica a flacarii pentru diferiti combustibili? December 07, 2019. https://www.greelane.com/ro/%C8%99tiin%C8%9B%C4%83-tehnologie-math/%C5%9Ftiin%C5A3%C4%83/flame-temperatures-table-607307