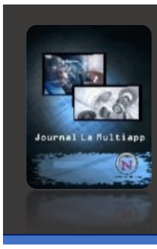

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Reducing GHG and NO$_x$ Pollutant Emissions by Hydrogen Burning with Technical Oxygen

Lucian Paunescu[1], Gheorghe Surugiu[2], Paul Dan Stanescu[3]

[1]Bilmetal Industries SRL Popesti-Leordeni, Ilfov, Romania
[2]State Inspection for the Control of Boilers, Pressure Recipients and Lifting Installation Bucharest, Romania
[3]Technical University of Civil Engineering, Department of Thermotechnics and Thermal Equipment Bucharest, Romania

**Corresponding Author: Lucian Paunescu*
Email: lucianpaunescu16@gmail.com

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Abstract

Traditional Ghanaian furniture designs are known as an exhibition of the splendor and elegance of decorative objects or decorative ornaments that carry profound philosophical and sacred meanings. The features found in Ghana's traditional Ghanaian design can be considered as objects for international players, with their own identity and not found elsewhere. This paper therefore seeks to identify and illuminate some of these Akan cultural objects in Ghana by referring to the Ashanti region. This paper also shows with examples how these design elements can be developed and maintained by integrating them into modern furniture and interior designs or as a straightforward or adaptive design.

Introduction

According to the IEA analysis carried out in 2019 (World Energy, 2019), the power provided by renewable energy in the period 2019-2024 is estimated to increase by 50 % mainly due to solar energy (60 % of the total growth). The increasing use of wind energy should contribute with 25 % of the planned 50 % increasing. The bioenergy capacity is also expected to increase by 4 % (especially in China, India and the EU). Instead, the power provided by hydropower stations will decrease, although it will still represent 10 % of the total increase in renewable capacity. Biofuels as a form of bioenergy can find their application in road transport and various forms of electricity and heat generation. Greenhouse gas (GHG) emissions containing mainly carbon dioxide (CO_2) supplied to the environment by the two mentioned areas by fossil fuel consumption had major increases and annual growth rates well above those of other sectors. For example, the fossil fuels used in transport (especially oil) represented in 2018 above 96 % of the total fuels. The transportation sector has generated about 15 % of the world's GHG emissions and about 23 % of the total GHG emissions from energy generation (World Energy, 2019; Sims et al., 2014). The GHG emissions in the period 1960-2018 in EU countries were generally below the average value recorded in 2018. The Czech Republic and Latvia had the lowest increase in GHG emissions (only 7 %). The highest increases were reached in Italy (229 Mt CO_2), Spain (219 Mt CO_2) and Poland (144 Mt CO_2). Only four European countries (Germany, Luxembourg, Sweden and the UK) had emissions in 2018 less than or equal to those of 1960 (Paraschiv & Paraschiv, 2020). On the other hand, the work (Greenhouse gas, 2021) shows that the road transport is responsible for more than 25 % of greenhouse gas emissions in the EU in 2010-2019 and for a major climate change. The

European Commission Fuel Quality Directive (FQD) has established that, by the nature of fuels used in road transport, the intensity of GHG emissions in the EU should be reduced by 6 % in 2020 compared to 2010. In 2019, Finland and Sweden have slightly exceeded the target for reducing the intensity set at 6 %, while the Netherlands was close to reaching it (5.8 %), because the three countries have high proportions of biofuels in the road transport (9; 7 and 22%, respectively). The GHG emissions of the three countries have values of 10.4; 15.2 and respectively, 14.1 g CO_2/MJ.

In the first half of the 20th century, biofuels were a suitable option for car engines, the bioethanol being commonly used in the United States and Europe (Jeswani et al., 2020). After World War II, the petroleum fuel was preferred due to its lower price. During the global hydrocarbon crisis of the 1970s, there was a tendency to return to biofuels. At the end of the millennium, due to rising oil prices, the interest in biofuels has increased again and in the last decade, with the increase of global warming through GHG emissions, the United States and Europe have developed programs to implement biofuels, especially in transportation road (European Commission, 2018).

According to Jeswani et al., 2020, biofuels can be classified into three groups. The first group would include products from food or animal feed crops by fermentation or distillation processes. The second group biofuels come from non-food raw materials (lignocellulosic plants, agricultural or forest residues, solid municipal waste, etc.). The biofuel produced from algae is included in the third group. Biofuels resulted by preparation (cultivation, collection, drying, milling/crushing, etc.) or processing (esterification, fermentation, gasification, hydrolysis, pyrolysis, distillation, purification, etc.) technique are: bioethanol, biodiesel, biogas, butanol, hydrogen, methanol, mixed alcohols, vegetable oil, glycerol, etc.). The using fields of biofuels are: road transport, energy generation and chemical industry.

The present paper refers to methods of using one of the biofuels mentioned above: hydrogen as a by-product of the chemical industry, bottled under pressure and supplied on the market in containers. The using field of hydrogen is the generation of thermal energy. The advantage of using this biofuel type in combustion installations is that the waste gas is water avoiding the existence of oxide compounds of carbon (CO_2, CO). However, even in the case of choosing hydrogen as a substitute for fossil fuels, but using air as a source of oxygen for the combustion process, another type of environmental pollutant (nitrogen oxides with the general formula NO_x) is released into the atmosphere. This group of oxides, of which nitrogen dioxide (NO_2) is the most important and most dangerous, comes in the highest proportion (about 50 %) from the residual gases of the internal combustion engine of cars in traffic, but also from the waste gas emissions of thermal power plants (about 20 %). Also, industrial manufacturing the cement, glass, nitric acid, incinerators, industrial thermal boilers, gas turbine, Diesel engines, etc. are to blame for NO_x generation and atmosphere pollution. To protect the human health, the annual maximum values of NO_x (as NO_2) in the atmospheric air are limited by international environmental protection agencies to 0.04 mg/m^3. Hourly NO_2 limit value reaches 0.2 mg/m^3 (Nitrogen dioxide, 2014). Limiting the amount of NO_x emissions into the atmospheric air for a time, usually longer, should not be confused with limiting the same emissions of waste gas released by the combustion processes of fuels, captured for oxide analysis before their release into the atmosphere. These limits are much higher, around 200-300 mg/m^3 in the case of low calorific fuels (Appendix 1, 2020) and up to 350 mg/m^3 in the case of higher thermal power fuels.

Generally, the NO_x formation is much more intense in combustion processes carried out at high temperatures (above 1300 ºC). Technically, the reduction of NO_x concentration in waste gas is possible by the way in which the fuel-combustion air mixture is made (usually in several mixing stages) (Gaba & Paunescu, 1998) or by increasing the oxygen concentration in the combustion air from 21 % to very high values that can even reach 100 %.

The first use of hydrogen's ability to reduce GHG emissions by burning together with a fossil fuel (natural gas) was in the 1970s in the United States by injecting hydrogen (5-15 vol. %) into the natural gas pipeline network (Melaina et al. al., 2013). The result of the preparation of this gas mixture was a major reduction in GHG emissions from combustion.

An applying solution of combined combustion with air of natural gas and hydrogen in an industrial burning plant in the Romanian chemistry industry was tested by the authors of the current work (Paunescu et al., 2007 a). Considering that hydrogen was a by-product of the plant, the reduction of natural gas consumption by above 20 % and the decrease of GHG emissions in the technological plant represented significant economic effects.

An innovative method of producing a hydrogen-oxygen mixture in stoichiometric proportions (called HHO) was patented in the United States at the beginning of the new millennium (Klein, 2002). Due to its completely different internal structure from the conventional molecular structure, containing molecules, atoms, free ions, free OH groups, similar to the plasma state, this mixture activated by direct contact with a metallic or ceramic material or a hot gas flux can develop a much higher thermal energy than the theoretical one. The collaboration of the authors of this paper with the Romanian company Rokura International Applications Bucharest (which bought the license for HHO application) allowed the access to the HHO generator and technology. A natural gas burner modified by injecting HHO into the already formed flame offered the possibility of testing the influence of the addition of the hydrogen-oxygen mixture in the conventional natural gas and air flame. The experiment identified that replacing 30 % natural gas with HHO increased the flame temperature from 1600 to 1890 ºC, so that the thermal effect to be similar to the operation of the burner with only natural gas (Paunescu et al., 2005; Paunescu et al., 2007 b).

The Japanese corporation Toyota Motor in collaboration with Chugai Ro Co Ltd has developed for the first time in the world engines for cars that run on hydrogen produced "in situ" and oxygen. Also, oxy-hydrogen burners were performed for industrial heating furnaces (Toyota, 2018). The technical solution adopted the two-stage hydrogen combustion method, aiming to decrease the final flame temperature. Hydrogen is distributed from the central pipe through radial orifices, partially meeting the oxygen jets brought into the combustion chamber area through several axial orifices occupying an annular peripheral area, as well as through an axial central nozzle. The second mixing step between the axially distributed hydrogen and the rest of the annular oxygen jet takes place in a more distant space compared to the mixing zone in the case of burning in a single step. The proportion of oxygen in the second stage is diminished and cools the oxy-hydrogen flame, reducing the possibility of NO_x formation. GHG emissions are zero.

In 2020, the Czech company Flamma Tec has developed a range of four oxy-hydrogen burners without GHG emissions, the combustion process being carried out stably at high temperature (FT Hydrogen Burner, 2020). The thermal power of the burners varies between 50-1500 kW, with hourly hydrogen flow rates between 15-500 m^3N/h. The hydrogen supply pressure is between 20-140 mbar and the oxygen pressure is in the range 15-120 mbar.

The objective of the research included in the current paper is to design, achieve and test a 30 kW-experimental oxy-hydrogen burner, without GHG emissions and low NO_2 emissions, adopting the method of staged mixing of hydrogen and oxygen.

Methods

In the current economic and environmental conditions, the use of hydrogen as an alternative energy source is an important advantage. It can partially or completely replace fossil fuels such as natural gas, gasoline, gas-oil, kerosene, etc., simultaneously eliminating GHG emissions from waste gas. Also, hydrogen could meet the requirements of power supply in situations where the use of energy storage would be difficult (e.g. fuel for heavy vehicles over long distances, off-road vehicles, electricity or fuel storage for damage to the power supply network or for land and air transport, etc.) (Lewis, 2021). Several ways of making hydrogen are known, starting with the well-known electrolysis of water and including various industrial technological processes in which this biofuel results as a by-product (Hydrogen production, 2021).

The design of the oxy-hydrogen burner took into account the previous experience of the authors in making conventional burners operating with fossil fuels (mainly natural gas) and oxygen-non-enriched or oxygen-enriched combustion air in various proportions up to 100 %. Methods to reduce NOx emissions by distributing the fuel in two mixing stages with the oxygen aimed at reducing the final flame temperature, were applied by the authors in previous works and more recently (in 2018) by Toyota Motor Corporation in the case of oxy-hydrogen burner.

NOx emissions from combustion processes are mainly in the form of NO. It is generated in the limit of oxygen available in the air at temperatures above 1300 ºC. The NO formation reactions according to Zeldovich (Technical Bulletin NO_x, 1999) are:

$$N_2 + O = NO + N \tag{1}$$

$$N + O_2 = NO + O \tag{2}$$

$$N + OH = NO + H \tag{3}$$

With the exception of NO from soils, NO is generated by human activity. NO_2 is present in the atmosphere and in the acid rain. According to some researchers, NO_2 is a suitable surrogate for NO_x because NO is rapidly converted to NO_2. So called “thermal NO_x” has its concentration controlled by the molar nitrogen and oxygen concentrations and the combustion temperature (above 1300 ºC) (Technical Bulletin NO_x, 1999). Practically, the use of an amount as close as possible stoichiometrically to the amount of fuel with which it reacts is ideal for minimal NO_x emissions. Also, lowering the flame temperature by self-cooling to the lower limit favors reducing the level of NO_x emissions.

The combustion reaction of hydrogen with oxygen is well known:

$$2H_2 + O_2 = 2H_2O \tag{4}$$

The flammability range of hydrogen in oxygen in closed spaces (propagation in tube) at the pressure of 1 bar and ambient temperature is extremely wide, between 4.1-94 %. A reduction in pressure below 1 bar leads to an increase in the minimum flammability limit of hydrogen in oxygen. The lower pressure observed for ignition is 0.5 mbar for a hydrogen concentration of 50 vol. % (Safety standard, 1997).

The reaction (4) is the simple, well-known variant, by which the water vapors result as a single combustion product. According to Willbourn et al., 1946, research conducted in this field since the first half of the 20th century showed that a chain of reactions would take place after about 550 ºC including three explosion limits, which are reached at a very low pressure (first limit), a pressure of about 10 mbar (the second limit) and pressures of 50-70 mbar (the third limit). Initially, by the reaction of molecular hydrogen (H_2), hydrogen atoms (H) or OH radicals with molecular oxygen (O_2) or oxygen atoms (O), the final reaction being:

$$OH + H_2 = H_2O + H \tag{5}$$

At the second explosion limit, the reaction (6/7) could take place with the formation of either H_2O_2 and H (6) or H_2O and OH (7).

$$HO_2 + H_2 = H_2O_2 + H \tag{6}$$

$$HO_2 + H_2 = H_2O + OH \tag{7}$$

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The constructive and functional scheme of the experimental oxy-hydrogen burner is shown in Figure 1.

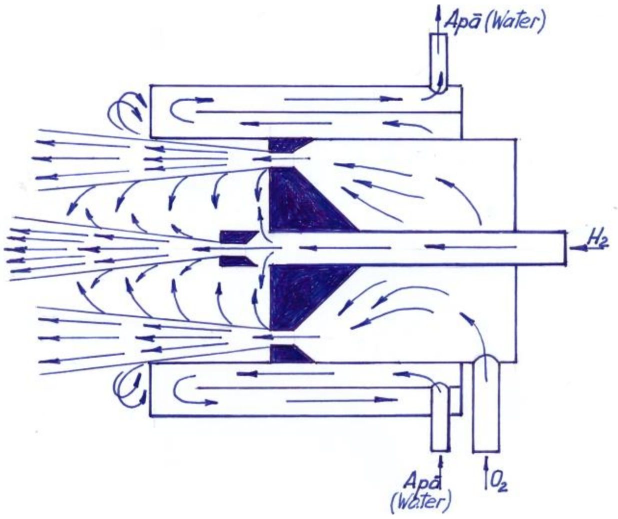

Figure 1.Constructive and functional scheme of experimental oxy-hydrogen burner

Design data for achieving the 30 kW-experimental oxy-hydrogen burner are presented in Table 1.

Table 1. Design data for achieving the experimental burner

Design date	**Unit**	**Value**
Thermal power of the burner	kW	30
Nominal hydrogen flow rate	m^3N/h	10.1
Nominal hydrogen pressure	mbar	153
Nominal oxygen flow rate	m^3N/h	5.05
Nominal oxygen pressure	mbar	147
Hydrogen rate in the axial orifice	m/s	260
Hydrogen rate in the radial orifices	m/s	200
Oxygen rate in the peripheral axial orifices	m/s	215
Waste gases rate at the exit of the burner	m/s	135
Cooling water flow rate	m^3/h	1
Cooling water rate	m/s	1.4

The thermal power value of the experimental burner was arbitrarily adopted at 30 kW. Considering that the lower calorific value of hydrogen is 33.3 kWh/kg or 2.97 kWh/m^3N (Fuels, 2010), it results that the nominal hourly flow rate of the fuel corresponding to the 30 kW-thermal power is 10.1 m^3N/h. Since the oxygen required for the stoichiometric combustion of hydrogen according to reaction (4) is 0.5 m^3N/m^3N-hydrogen, it follows that the nominal hourly flow rate of oxygen is 5.05 m^3N/h. The nominal pressure values at the entering in the burner body were adopted at 153 mbar for hydrogen and 147 mbar for oxygen. Also, the hydrogen rate in the axial orifice was chosen at 260 m/s and in the radial orifices at 200 m/s, while the oxygen rate in the peripheral orifices was adopted at 215 m/s. The rate values of the two fluids in the minimum section of the orifices were adopted on the basis of the previous experience of the authors in the field of burners with stepped distribution of fuel and air or oxygen to obtain an optimal mixture between fluids and self-cooling the flame in order to significant reducing the GHG emissions.

Testing the experimental burner was performed in a first stage in free space on the territory of Cermax 2000 Patents SRL in the Institute of Metallurgical Research SA Bucharest in order to viewing the flame propagation and the degree of stability in operation (Figure 2 A). The determination of the functional parameters and identification of technical characteristics of the burner were made on the testing stand at the Technical University of Civil Engineering Bucharest (Figure 3). The light intensity and the length of the flame were observed through the visors of the testing stand (Figure 2 B).

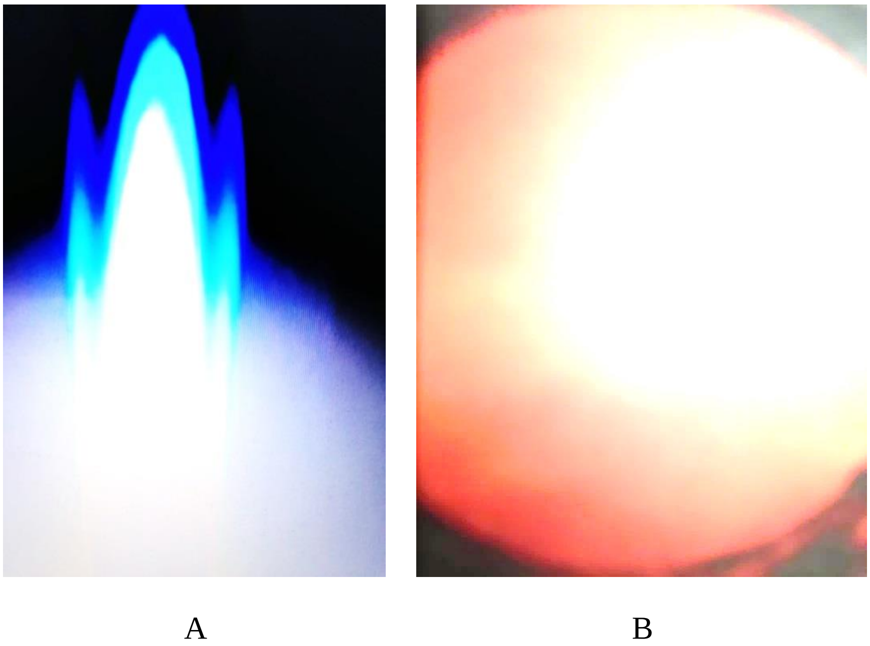

Figure 2.Oxy-hydrogen burner flames observed in free space (A) and through the visors of the testing stand (B)

The maximum thermal capacity of the testing stand (Figure 3) at the Technical University of Civil Engineering allows checking the operation of burners with thermal powers below 200 kW. The cylindrical working enclosure has an inner diameter of 400 mm and an outer diameter of 500 mm, being cooled with water. The length of the enclosure is a maximum of 1200 mm, and can be adjusted according to the power of the burner subjected to testing. Several visors for flame propagation observation are provided on the outer surface of the metal enclosure. Prior to removal to the atmosphere, waste gas is directed through a gas-water heat exchanger placed at the outlet end of the enclosure. Cooling water circulating in a closed circuit allows determining the amount of developed heat. A gas analyzer is attached to the stand, the gas samples being taken over with a capture pipe through any visor of the stand.

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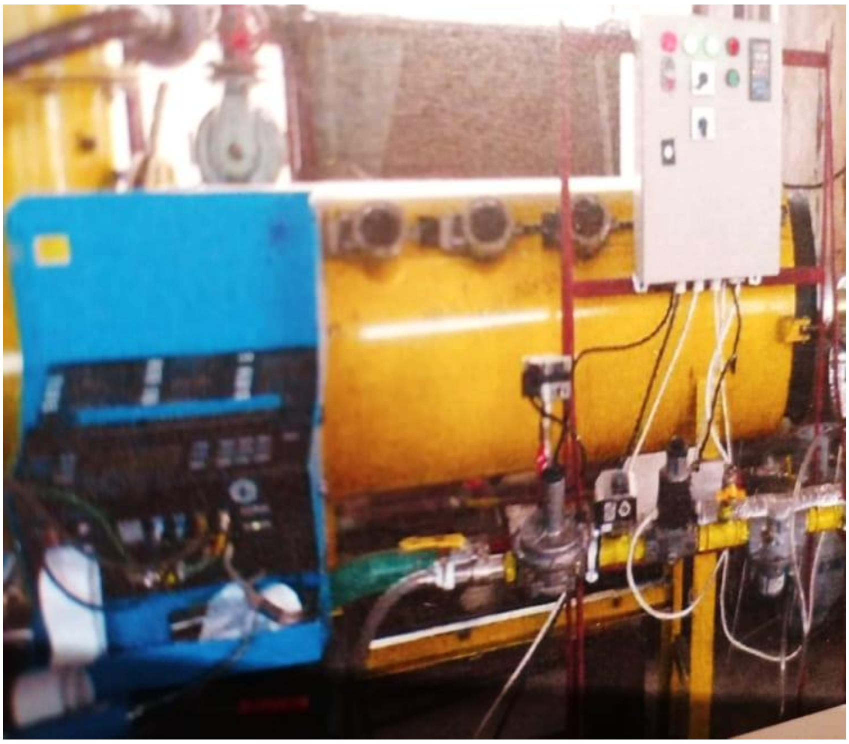

Figure 3.Overall image of the testing stand (Reproduced by permission of Technical University of Civil Engineering Bucharest, Department of Thermotechnics and Thermal Equipment)

The accuracy level of the measurements of flow rate, pressure, temperature and oxide composition of hydrogen, oxygen, waste gas and cooling water offered by the equipment related to the testing stand is very high, at European level.

The experimental method adopted by the authors provides the operation testing of the burner in the range of hourly hydrogen flow rates between 5.00-10.10 m^3N/h. Eight thermal regimes of stable operation were chosen, the hydrogen flow rate having successive values: 10.10; 8.50; 6.20 and 5.00 m^3N/h and the hourly flow rate of oxygen was the one that allows a stoichiometric combustion (without oxygen excess), i.e. 5.05; 4.25; 3.10 and 2.50 m^3N/h. To these four thermal regimes, four other regimes were added, in which the oxygen flow rate was supplied with a small excess (1.04) compared to the stoichiometric requirement, i.e. 5.25; 4.42; 3.22 and 2.60 m^3N/h.

The hourly flow rates of hydrogen and oxygen and their pressures at the entrance in the burner body were read on the control panel of the equipment. Also, the values of cooling water parameters were noted for determining the heat energy amount taken over by it and implicitly, in order to calculate the value of the waste gas temperature at the exit of the testing stand. The flame length corresponding to each thermal regime was measured through visors provided on the enclosure wall. The waste gas oxide composition was controlled with the analyser existing as component part of equipment, the values of oxygen (vol. %), NO and NO_2 (in ppm converted into mg/m^3N), CO_2 and CO (vol. %) being continuously indicated.

The main rules regarding the process of ignition and combustion in conditions of maximum safety of the hydrogen-oxygen mixture, according to (Safety standard, 1997), were applied in this experiment.

Results and Discussion

The two energy fluids (hydrogen and oxygen) were used in gaseous form bottled under pressure in metal containers. Hydrogen was purchased from Linde Gas Romania in 8.9 m^3

bottles at 200 bar and the technical oxygen was purchased from Artego SA Tg.Jiu, Romania in 6.35 m^3 steel containers at 150 bar.

The experimental results are shown in Table 2 containing the eight thermal regime adopted for testing.

Table 2.Experimental results of the 30 kW-oxy-hydrogen burner

Functional parameter	**Thermal regime**							
	1	2	3	4	5	6	7	8
Hydrogen flow rate (m^3N/h)	10.10	10.10	8.50	8.50	6.20	6.20	5.00	5.00
Hydrogen pressure (mbar)	153	153	103	103	59	59	42	42
Oxygen flow rate (m^3N/h)	5.05	5.25	4.25	4.42	3.10	3.22	2.50	2.60
Oxygen pressure (mbar)	147	155	96	101	54	58	38	40
Waste gas oxide composition:								
-O_2 (vol. %)	-	0.19	-	0.16	-	0.12	-	0.10
-NO (mg/m^3N/h)	238	226	216	200	185	171	159	151
-NO_2 (mg/m^3N/h)	248	235	224	210	206	189	180	166
-CO_2 (vol. %)	-	-	-	-	-	-	-	-
-CO (vol. %)	-	-	-	-	-	-	-	-
Flame temperature (°C)	1854	1837	1806	1798	1795	1789	1749	1728
Flame length (mm)	410	430	380	390	350	370	300	340
Heat taken over by cooling water from waste gas (MJ/h)	114.9	113.7	96.7	95.8	70.6	70.1	56.9	56.2

The experimental oxy-hydrogen burner with the nominal thermal power of 30 kW, designed for using in industrial heating processes, was tested in the hourly hydrogen flow rate range 5-10.1 m^3N/h. According to the data in Table 2, the technical oxygen used for combustion had the hourly flow rate value between 2.50-5.26 m^3N/h. Eight thermal regimes in the field of stable flame operation have been achieved, regimes no. 1, 3, 5 and 7 being characterized by ensuring the stoichiometric ratio (2:1) between the volumes of hydrogen and oxygen participating in the combustion process, while the regimes no. 2, 4, 6 and 8 had a small excess of oxygen (excess coefficient of 1.04) compared to the stoichiometric ratio. The supply pressure of the two energy fluids (at the entrance into the burner body) was between 42-153 mbar for hydrogen and between 33-155 mbar for oxygen.

The method applied by design to distribute hydrogen in stages in the combustion zone so that there is a first mixture between fluids in the inferior part of the combustion chamber through radial hydrogen jets and part of the peripheral oxygen flow and then in the front combustion zone by the central axial jet of hydrogen and most of the peripheral flow of oxygen. Due to the higher rate (maximum 260 m/s) of the axial jet of hydrogen, it tends to aspirate the peripheral annular flow of oxygen so that the reaction between hydrogen and oxygen also takes place in the frontal area of flame propagation, the cold oxygen having also the role to partially cool the very hot oxy-hydrogen flame. Theoretically, by burning the two fluids without self-cooling methods, the flame temperature reaches 2660 ºC (Typical flame, 2019), while the measured flame temperature in this experiment was between 1728-1854 ºC.

According to the data in Table 2, the oxide composition of waste gas (in the front area of the flame) does not contain GHG emissions (CO_2 and CO) and contains O_2 traces (0.10-0.19 vol. %) only in regimes 2, 4, 6 and 8. The NO_x emissions (NO_2 and NO) inherent in the high temperature conditions of waste gases had relatively low values, below the maximum allowed limits. The highest values of NO_2 and NO were reached at the nominal thermal regime, without and respectively, with the self-cooling of the flame (248/238 mg/m^3N and 235/226 mg/m^3N, respectively). These values decrease with the reduction of the flame temperature reaching the minimum values at the minimum thermal regimes 7 and 8 (180/159 mg/m^3N and 166/151 mg/m^3N, respectively).

The length of the oxy-hydrogen flame in the range of maximum operating stability between 14.85-30 kW had relatively normal values (300-430 mm) equivalent to other similar burners in the world. According to the literature, the oxy-hydrogen flame is characterized by a high impulse. The rate value of waste gases at the exit from the burner in the current design is 135 m/s (Table 1), that is typical for an impulse burner.

Therefore, the objective of the work to design, realize and test in industrial conditions a high-performance burner in terms of energy with low emissions of pollutants, in ecological terms, has been met. Hydrogen, which is a biofuel generally obtained as an industrial by-product, is a suitable substitute for fossil fuels and its exclusive use completely eliminates greenhouse gas (GHG) emissions. Furthermore, the total replacement of combustion air with technical oxygen can lead to a significant reduction in NO_x emissions, especially if the technique of distribution in stages of hydrogen is applied and, implicitly, the self-cooling of the flame.

Conclusion

Biofuels, as a bioenergy form, have a wide range of applications both in road transport and various forms of electricity and heat generation. These areas of activity generate significant amounts of greenhouse gases (GHG) by the current use of fossil fuels. Hydrogen is one of the biofuels that can contribute to the reduction or complete elimination of these pollutants (mainly CO_2). The objective of this work is to achieve and test an oxy-hydrogen burner for heat generating only with hydrogen and technical oxygen. The work is one of the few achievements in this field in the world due to the special features and conditions of the oxy-hydrogen combustion process, quite different from known conventional combustion processes. The burner applies by design all the modern methods on fluid mixing in stages and self-cooling the flame to reduce NO_x emissions as well as the exclusive use of hydrogen as fuel and technical oxygen as an oxygen supplier for combustion, without including other fossil fuels and without using air even enriched in oxygen in high proportions. As a result, the designed burner has a stable operation without risk of explosion in the thermal power range 14.85-30 kW, GHG emissions (CO_2 and CO) are zero and NO_2 and NO emissions are below the maximum limits allowed in the world. According to the literature, the application of the oxy-hydrogen burner in the version of the Japanese company Toyota Motor with the "in situ" production of hydrogen in the field of the internal combustion engine for cars and the forging heating furnace is in an advanced stage. For the future application of the Romanian project, additional tests and verifications are needed.

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