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# The Influence of Modified Atmosphere on Natural Gas Combustion

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## 1. Introduction

Coal technology, dominant in Poland, ensures efficient production of electricity and heat. However, a large exploitation of existing reserves and the growing demand for electricity causes interest in other available fuels, primarily natural gas. Environmentally friendly modern technologies are constantly looking for the possibility of using energy sources other than coal. Research is carried out on various innovative technologies such as CO<sub>2</sub> capture and storage or unconventional combustion. Combustion in the oxygen-enriched gas mixtures at the first was conducted in the steel industry and metallurgy, which require very high temperatures to heat the metal and pig iron. Such a process can be classified as the future technology known as the clean combustion technologies.

The use of natural gas in the metallurgical processes requires the precise technological regime, therefore, it is difficult to make changes in the process (Amann, Kanniche & Bouallou, 2009; Choi & Katsuki, 2001; Flamme, 2001). However, appropriately organized gas combustion process can lead to a reduction in CO<sub>2</sub> emissions, which is required by applicable laws imposed by the European Union. The European Union environmental priorities promote the development of the new technologies of energy production from the conventional sources (coal and natural gas), e.g. oxy-combustion (Andersson & Johnsson, 2007; Czakiert et al., 2006; Davidson, 2007; H.K. Kim, Kim, Lee & Ahn, 2007; Kotowicz, 2007; Lampert & Ziębik, 2007; Li, Yan & Yan, 2009; Muskał et al., 2008; Seepana & Jayanti, 2009; Simpson & Simon, 2007; Szlęk et al., 2009; Tan et al., 2002). The oxy-fuel combustion process is conducted in an atmosphere enriched in oxygen which means that the reactor is supplied by the combustion gas mixture, in which the oxygen concentration is higher than the concentration of oxygen in the air. The study of the modified air combustion appears in scientific literature. There are various modified oxidizing atmospheres like O<sub>2</sub>/N<sub>2</sub>, O<sub>2</sub>/N<sub>2</sub>/CO<sub>2</sub>, O<sub>2</sub>/CO<sub>2</sub>. It should be emphasized that the majority of results of the oxy-combustion process refer to coal combustion, because of large amount of deposited coal in Poland (Buhre et al., 2005; Chen, Liu & Huang, 2007; Croiset, Thambimuthu & Palmer, 2000; Czakiert, Nowak & Bis, 2008; Kim et al., 2007; Normann et al., 2008). The research effort is focused on the conventional coal, fluidised bed combustion and co-generation solutions with the coal gasification (Lampert & Ziębik, 2007; Tan et al., 2002). Oxy-combustion is the subject of research in many international

research institutions (Chalmers University of Technology, Sweden, University of Leeds UK, CANMET Energy Technology Centre, Canada, Chicago USA Research Centre, Tokyo Institute of Technology, Japan, University of Newcastle Australia). In Poland, the research of oxy-combustion concerns mainly coal (Czestochowa University of Technology), but the natural gas research is also carried out in the field of high-temperature combustion gas HTAC technique (Normann, 2008; Seepana & Jayanti, 2009). Fuel combustion processes are the main source of environmental pollution. In many branches of industry, the main fuel is natural gas, because of the possibility of obtaining a high temperature process e.g. in the glass industry and in the manufacture of cement, the process of oxy-combustion can be applied, or air combustion enriched with oxygen. This of course raises the temperature of the combustion process, which is associated with increased concentrations of  $\text{NO}_x$ . Therefore, oxy-combustion is used simultaneously with the process of eliminating nitrogen from the air combustion (largely responsible for the formation of NO) and replacing it into exhaust gas (RFG - Recycled Flue Gas.). Despite the very high temperatures in the chamber (e.g. 1600 °C in the melting process), the concentration of nitrogen oxides may be lower, due to the elimination of nitrogen from combustion air. The primary obstacle to the propagation of oxy-combustion has so far been the high cost of obtaining pure oxygen. Since the pure oxygen production technologies have been improved and costs have been reduced, oxy-combustion can be applied in many industrial processes. Industry may be interested in this technology, where the conditions in the very high temperature contribute to the formation of large amounts of thermal NO. An additional advantage of the oxy-combustion is high combustion efficiency, lower volume of exhaust gases, less fuel consumption and therefore lower  $\text{CO}_2$  and  $\text{NO}_x$ . Combustion of natural gas in  $\text{O}_2/\text{CO}_2$  atmosphere allows optimising the combustion process. The results of experimental studies indicate that the combustion of natural gas in  $\text{O}_2/\text{CO}_2$  with recycling exhaust gases has positive effects on reducing  $\text{CO}_2$  emissions, a noticeable reduction or even elimination of  $\text{NO}_x$  and improve the efficiency of the furnace (Lampert & Ziębik, 2007; M.Wilk, Magdziarz & Kuźnia, 2010). It was noted that the major advantage of this technology is the ability to apply it in an existing energy plants (H.K. Kim, Kim, Lee & Ahn, 2007; Simpson & Simon, 2007).

Therefore, the problem seems to be both interesting and promising. The complex nature of the combustion process of natural gas causes the obtained experimental results which are not always repeatable so this issue requires further study.

## **2. Mechanisms of CO and $\text{NO}_x$ formation in natural gas combustion processes**

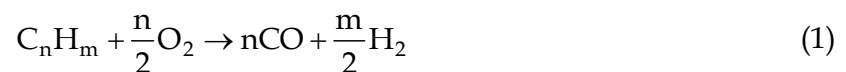
In many industries, because of the possibility of obtaining high temperatures, the primary fuel is natural gas, whose main component (ca. 98%) is methane. Combustion of natural gas is the source of the formation of many pollutants. During the combustion of natural gas in metallurgical furnaces the air pollutants are formed. These are nitrogen oxides, carbon oxides, and possibly trace amounts of hydrocarbons. The composition of natural gas varies slightly. The number and types of pollutants emitted from combustion are related to the composition of the fuel, the type of oxidation atmosphere used and the temperature prevailing in the combustion chamber.

## 2.1 The mechanism of CO formation

The relatively high CO emissions in combustion processes of natural gas occur in the following cases:

- Staging combustion for reduction of NO<sub>x</sub> emissions,
- Inadequate mixing of air and fuel,
- Very rapid cooling of combustion products in the cold boundary layer of the combustion chamber.

The formation of CO in the flame is one of the main paths of reaction in the mechanism of combustion of hydrocarbons. Fuel hydrocarbons during the chemical degradation can be partially converted to CO. The formation of CO is done quickly, right at the beginning of the flame. The proposed overall reaction of formation of CO is as follows (for gaseous and liquid hydrocarbons) and volatiles of solid fuels. (Wilk, 2002; Bartok & Sarofim, 1991)



Oxidation of CO is strongly catalysed by even small amounts of hydrogen or its compounds with oxygen. There are two paths of CO oxidation. Main path of oxidation takes place at  $T > 1500$  K (at  $p \approx 0,1$  MPa) and is as follows



The second path CO oxidation takes place at  $T = 1000 - 1500$  K and at  $p > 1$  MPa is as follows



HO<sub>2</sub> radical is produced in the recombination reaction:



HO<sub>2</sub> radical concentration is comparable to the concentration of OH radical reactions and rapid reactions (Bartok & Sarofim, 1991)



Competitive reactions to the above and CO oxidation reactions are the following recombination reaction





H and OH radicals can meet together on the walls where it comes to their exhaustion, and it causes stopping of the CO oxidation reaction. In practice it is the result of too rapid cooling of exhaust gases below 1000 K. This is the main reason of no oxidation of CO to CO<sub>2</sub>. The direct oxidation of CO by reaction:



is very unlikely, because this reaction is very slow (the activation energy of this reaction is very high).

## 2.2 The mechanisms of NO formation

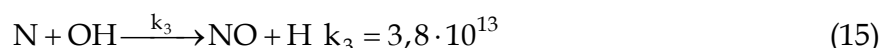
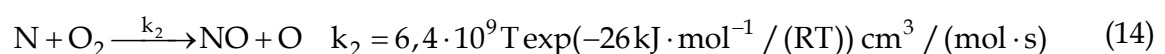
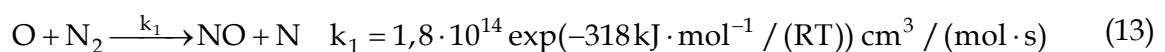
The primary adverse products of high temperature combustion are the nitrogen oxides NO<sub>x</sub>. Knowledge of the mechanism of the NO<sub>x</sub> formation can identify thermal and chemical conditions of furnaces and control of combustion processes, which affects prevention or reduction of the harmful substances emissions. The source of nitrogen oxides is nitrogen in the fuel and molecular nitrogen from the air. In combustion processes, there are two main types of nitrogen oxides: nitrogen monoxide, NO, and dioxide, NO<sub>2</sub>. The main component of NO<sub>x</sub> produced during natural gas combustion is NO, whose share in total NO<sub>x</sub> emissions is typically at least 95%, and the rest is NO<sub>2</sub>. The concentration of other oxides N<sub>2</sub>O, N<sub>2</sub>O<sub>3</sub> and N<sub>2</sub>O<sub>5</sub> is low. The amount of NO<sub>x</sub> in the exhaust gases depends mainly on the combustion temperature, excess air ratio and residence time in the reaction zone. There are four different mechanisms of the formation of NO:

- the thermal mechanism,
- the prompt mechanism,
- by means of N<sub>2</sub>O
- the fuel NO<sub>x</sub>.

During the combustion of natural gas, containing mostly methane, and not containing chemically bound nitrogen, the main way of NO<sub>x</sub> formation mechanism in natural gas combustion is the thermal mechanism.

### 2.2.1 The thermal mechanism of NO formation

The thermal mechanism is based on the oxidation reactions of nitrogen from the air supplied for combustion, the rate becomes significant above 1400 °C. These reactions were first described by Zeldovich (Bartok & Sarofim, 1991; Warnatz et al., 2006; Tomeczek & Gradoń, 1997; Muzio & Quartucy, 1997; Flamme, 1998):



The term "the thermal NO" is connected with a very high activation energy due to a strong, triple-atomic bond in the molecule N<sub>2</sub>. It is a highly endothermic reaction that runs with

considerable speed only at temperatures higher than 1400 °C. The reaction of a hydrocarbon radical OH plays an important role in the combustion of humidified hydrocarbon fuels.

The rate of formation of NO is expressed by the equation (Warnatz et al., 2006; Gardiner, 2000)

$$\frac{d[\text{NO}]}{dt} = k_1 [\text{O}][\text{N}_2] + k_2 [\text{N}][\text{O}_2] + k_3 [\text{N}][\text{OH}] \quad (16)$$

Atomic nitrogen is formed by the reaction (13) and is consumed in the reaction (14) and (15), hence the rate of formation is:

$$\frac{d[\text{N}]}{dt} = k_1 [\text{O}][\text{N}_2] - k_2 [\text{N}][\text{O}_2] - k_3 [\text{N}][\text{OH}] \quad (17)$$

Taking into account the fact that reactions (14) and (15) are so fast that their products reach the equilibrium state, the preliminary assumption can be given:

$$\frac{d[\text{N}]}{dt} = 0 \quad (18)$$

and then equation (16) takes the form

$$\frac{d[\text{NO}]}{dt} = 2k_1 [\text{O}][\text{N}_2] \quad (19)$$

$k_1$ ,  $k_2$ , and  $k_3$  are the rate constants of the reaction.

The rate of formation of NO is controlled by the first, slow reaction of Zeldovich (13). If one molecule of one atom of NO and N is produced in this reaction, it immediately becomes the second particle produced by rapid reaction of NO (14). The formation of thermal NO takes place just behind the flame front in a zone of high temperatures ( $t > 1400$  °C). The basic ways of reducing emissions of thermal NO in combustion processes are the reduction of the temperature, shortening the stay of the reagents in the zone of high temperatures and reducing the local concentration of  $\text{N}_2$  and  $\text{O}_2$ .

Malte and Pratt proposed the mechanism of taking into account the role of  $\text{N}_2\text{O}$  in NO formation by the following reaction at temperatures lower than 1800 K (Kordylewski, 2008 (in Polish), Steele et al, 1995)



These reactions, together with the reactions (13) and (14) in the literature are called "the extended thermal mechanism". The formation of NO by a mechanism of  $\text{N}_2\text{O}$  formation is particularly important at lower temperatures ( $T < 1200$  °C) in the flames rather poor ( $\lambda > 1$ ). Important role in the formation of  $\text{N}_2\text{O}$  plays the kind and characteristics of the third body M. It can be assumed that  $\text{H}_2\text{O}$  or its dissociation products (O, H, OH) affect the course of the reaction (20) (Wilk, 2002).

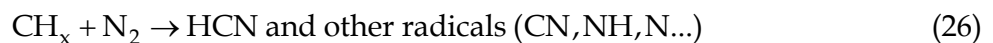


### 2.2.2 The prompt mechanism of NO formation

Fenimore conducting the experimental research on the combustion of rich mixtures ( $\lambda < 1$ ) with various hydrocarbons (methane, ethane, propane) found that relatively high concentrations of NO occur in combustion zone just before the flame where the temperature does not exceed 750 °C. Fenimore was confident that there should be another mechanism for the generation of thermal NO and called him a "prompt" which is immediate. In the hydrocarbon flames there are not only O, H, OH radicals but also hydrocarbon radicals, where its highest concentration was found in the reaction zone of the flame. The hydrocarbon radicals are capable of activating N<sub>2</sub> reaction with the formation of nitrogen oxides in the flame. Fenimore assumed that CH<sub>i</sub> hydrocarbon radicals react with nitrogen air according to the following reaction:



Generally, these reactions can be written:



The forming of amino and cyano compounds, among which the most important are HCN, NH, and CN, is oxidized to NO in the flame with the participation of radicals H, O, OH (Glarborg, Alzueta & Dam-Johansen, 1998). The prompt NO is formed very quickly during combustion. Velocity of its formation is like combustion velocity. The amount of formed NO depends weakly on temperature, but strongly depends on the local concentration of N<sub>2</sub>. The NO prompt participates in further reactions running along the flame and lose their individuality.

### 2.2.3 The fuel NO mechanism

The amount of nitrogen in the fuel composition is very diverse. Nitrogen, in the gaseous fuel, is not chemically bonded with the combustible gas. However, it can occur as free molecular nitrogen N<sub>2</sub>, which is the source of thermal or prompt nitrogen oxides. It is assumed therefore, that during the gas combustion the fuel nitrogen oxides do not occur.

### 2.2.4 The formation of NO<sub>2</sub>

Miller and Bowman gave the most plausible explanation of the mechanism of formation of NO<sub>2</sub>. They assumed that as a result of diffusion of H radicals from the flame in the area of low temperature ( $T < 750$  °C) and high concentration of O<sub>2</sub> the reaction occurs (Miller & Bowman, 1989; Hori, 1986).



At the same time from the flame to low temperature zone NO diffuses, which comes in a rapid reaction with a peroxide radical reaction of HO<sub>2</sub> by:



In parallel, at higher temperatures, the  $\text{NO}_2$  decomposition reactions may occur:



Therefore, under normal conditions of combustion ( $T = 1000 - 1700 \text{ }^\circ\text{C}$ ,  $\lambda \leq 1,3$ ) the final of  $\text{NO}_2$  emission is low and does not exceed 5% of the total  $\text{NO}_x$  emissions. Significant impact on the formation of  $\text{NO}_2$  next to a low temperature is connected with a high combustion pressure and the presence of hydrocarbons. The increase in the pressure favours the growth of the concentration of  $\text{NO}_2$ .

### 3. Experimental apparatus

The investigation of oxy-combustion process was conducted on a laboratory reactor containing a specially designed combustion burner, oxidizer preparation system, and temperature system, flow rate of combustion substrates control system, exhaust gas analysis system and exhaust gas system (Fig. 1).

The study included the characterization of the basic parameters of combustion, and above all took into account the effect of oxygen and carbon dioxide concentrations in the oxidizer on the exhaust gas composition and temperature profile along the combustion chamber.

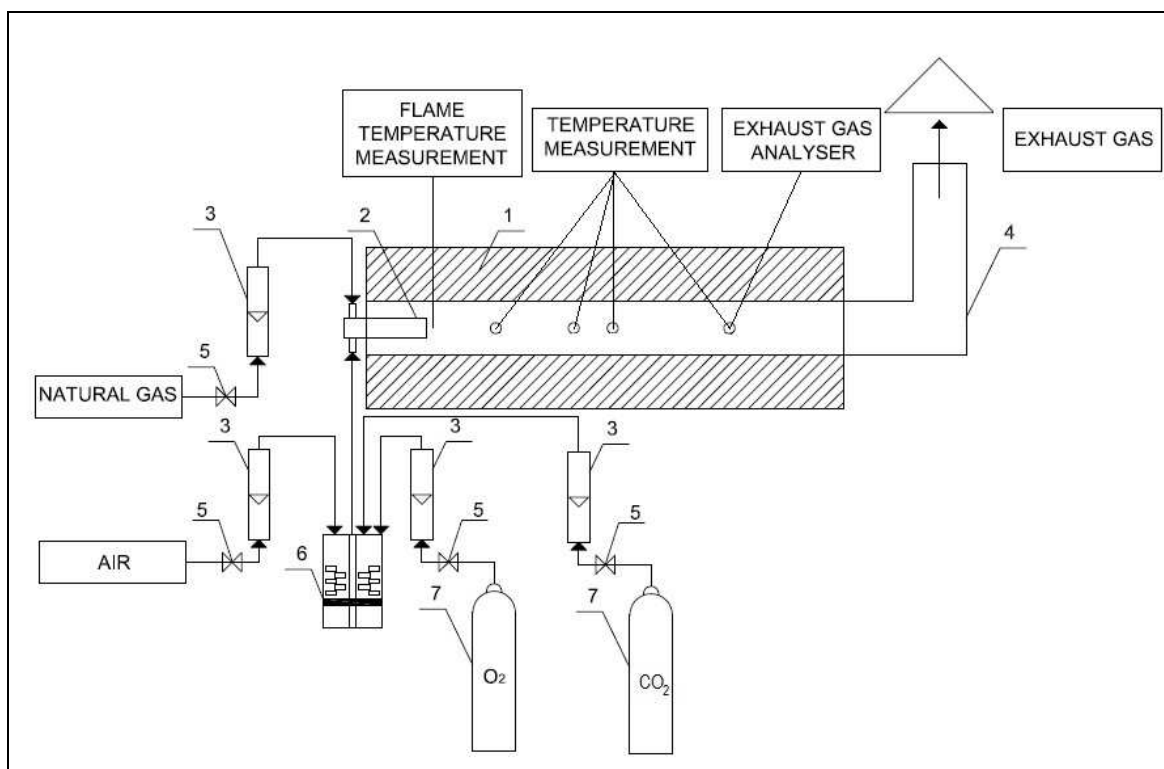


Fig. 1. Scheme of the experimental apparatus: 1 - combustion chamber, 2 - burner, 3 - rotameter, 4 - exhaust gas system, 5 - control valves, 6 - mixer, 7 - cylinder with oxygen, 8 - cylinder with carbon dioxide.



The specially designed kinetic burner, so called "pipe in pipe", was used. The inside diameter of burner was 34 mm and the outer diameter was 47 mm (Fig. 2). The combustion chamber was made from the heat-resistant steel with a length of 1310 mm and a diameter of 160 mm. Thermal isolation chamber was made of ceramic fibre with a thickness of 150 mm. Along the combustion chamber were holes, which allowed the measurement or analysis of the exhaust gas temperature inside the furnace. Oxidizer preparation system consisted of oxidizer pressure cylinders containing oxygen, air supply system with a fan and a mixer filled with the ceramic fittings, which enable more efficient mixing of the streams brought air and oxygen. The flow rates control system of combustion substrates included the rotameters and control valves. Mixing of fuel with an oxidizer took place in space between two pipes inside the burner, and was enforced by the system of holes in the outer pipe of the burner. The homogeneous mixture was introduced into the combustion chamber. The homogeneous mixture was introduced into the combustion chamber.

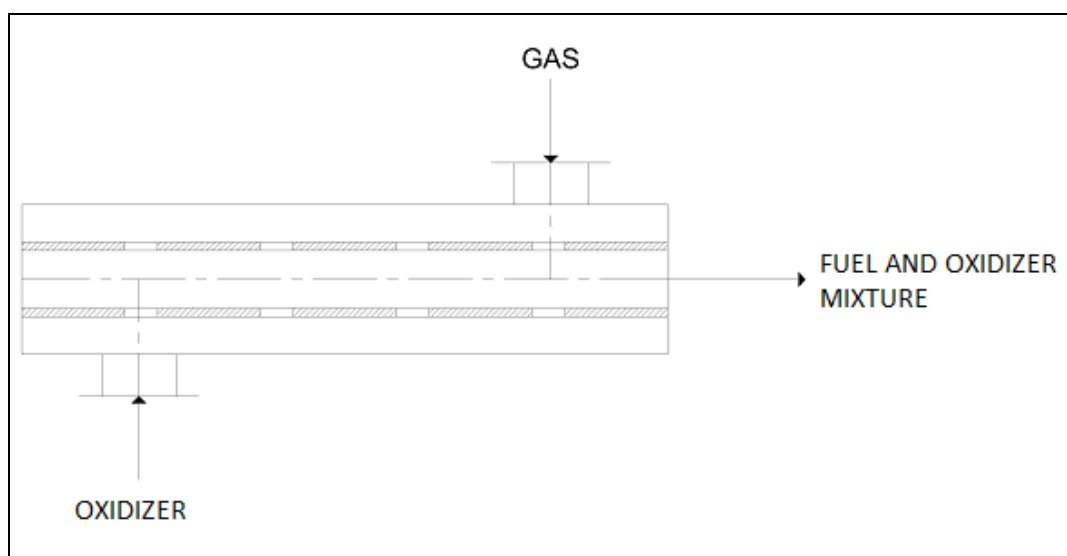


Fig. 2. Scheme of a specially designed burner.

To measure the flame temperature the thermocouple (PtRh-Pt) is installed in the wall of the chamber combined with a digital millimetre. The temperature profile along the length of the furnace was measured by NiCr-NiAl thermocouples at four points connected to a multichannel temperature recorder Czaki WRT-9 consisting of a microprocessor thermometer EMT 200, the switch places the PMP test. Concentrations of the combustion products ( $O_2$ , CO,  $CO_2$ , NO) were measured by the means of a gas analyser Testo 350 XL.

#### 4. Results and discussion

The investigation of the oxy-combustion of natural gas is takes into account three types of oxidizing mixtures with an increased oxygen contents: 25%  $O_2$ , 27%  $O_2$ , 29%  $O_2$ . The parameters of the combustion process of the natural gas with the addition of oxygen to the combustion air are shown in Table 1. The study concerned the natural methane rich gas from the city with the following average composition:  $CH_4$  - 98%,  $C_2$ - $C_4$  - 0,9%,  $N_2$  - 1%,  $CO_2$  - 0,1%.

To study the combustion of natural gas in modified atmosphere three options were carried out: the first - for the selected excess air ratio, the second - assumed a steady stream of gas

$\dot{V}_g, \text{ m}^3/\text{h}$	$\dot{V}_{\text{oxidizer}}, \text{ m}^3/\text{h}$	$\text{O}_2, \%$	$\dot{V}_{\text{CO}_2}, \text{ m}^3/\text{h}$	$T_{\text{flame}}, \text{ }^\circ\text{C}$	$T_{\text{exhaust gas}}, \text{ }^\circ\text{C}$ (in measured point)
0,8	6,3 - 9,5	21 - 29	0 - 2	1231 - 1316	885 - 1277

Table 1. The parameters of the combustion process of natural gas.

and air mixture for each oxygen-enriched air, and the obtained values  $\lambda$  resulted from the mixture of air and oxygen, and third option concerned the study of  $\text{CO}_2$  addition to oxidizer whereas the air combustion was oxygen-enriched up to 27% and excess air ratio was:  $\lambda_1 = 1,25$ . The excess air ratio was calculated by taking into account the increased oxygen content in the mixture.

The first investigations were conducted for three values of the air excess ratio:  $\lambda_1 = 1,15$ ,  $\lambda_2 = 1,20$ ,  $\lambda_3 = 1,25$ . The flow of gas was constant  $\dot{V}_g = 0,8 \text{ m}^3/\text{h}$ . For each value of  $\lambda$ , by choosing an appropriate air and oxygen flows ratio, the air was oxygen-enriched in the range 21 - 29%. Table 2 and 3 shows the average values of measured experimental data.

The effect of oxygen addition to the combustion air on CO concentration for three different excess air ratios  $\lambda_1 - \lambda_3$  was presented in Figure 3. For all the cases an increase in CO concentration with increasing oxygen concentration in the oxidising mixture was observed. The higher the excess air ratio, the lower was the concentration of carbon monoxide. It was observed that a small addition of oxygen around 4% slightly increases the concentration of CO, and the addition of 6% - 8% strongly increases the CO concentration. There was no

$\lambda_1 = 1,15$				
$\dot{V}_{\text{air}}, \text{ m}^3/\text{h}$	$\dot{V}_{\text{O}_2}, \text{ m}^3/\text{h}$	$\text{O}_2, \%$	CO, ppm	NO, ppm
8,67	0	21	175	125
7,014	0,38	25	192	231
6,317	0,534	27	230	347
5,715	0,663	29	301	533
$\lambda_2 = 1,20$				
$\dot{V}_{\text{air}}, \text{ m}^3/\text{h}$	$\dot{V}_{\text{O}_2}, \text{ m}^3/\text{h}$	$\text{O}_2, \%$	CO, ppm	NO, ppm
9,04	0	21	161	114
7,32	0,40	25	170	228
6,59	0,56	27	224	352
5,96	0,69	29	293	521
$\lambda_3 = 1,25$				
$\dot{V}_{\text{air}}, \text{ m}^3/\text{h}$	$\dot{V}_{\text{O}_2}, \text{ m}^3/\text{h}$	$\text{O}_2, \%$	CO, ppm	NO, ppm
9,421	0	21	112	78
7,620	0,418	25	118	206
6,866	0,580	27	179	305
6,212	0,720	29	215	458

Table 2. The results of experimental studies of natural gas combustion in oxygen enriched atmosphere for  $\dot{V}_g = 0,8 \text{ m}^3/\text{h}$ ,  $\dot{V}_{\text{CO}_2} = 0$ .

$\lambda_3 = 1,25; O_2 = 27 \%$		
$\dot{V}_{CO_2}, m^3/h$	CO, ppm	NO, ppm
0	179	305
0,5	182	144
1	210	73
1,5	242	43
2	355	24

Table 3. The results of experimental studies of natural gas combustion in CO<sub>2</sub> and oxygen enriched atmosphere for  $\dot{V}_g = 0,8 m^3/h$ ,  $\lambda_3 = 1,25$ ;  $O_2 = 27 \%$ .

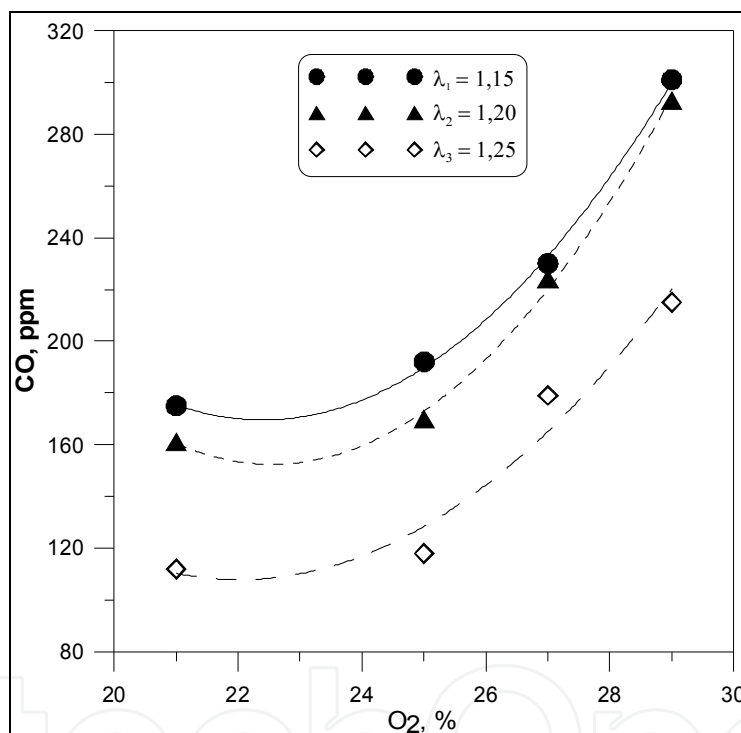


Fig. 3. The effect of oxygen addition to the combustion air on CO concentration in the natural gas combustion process with the oxygen enriched air depending on the excess air ratio.

difference in the concentration of CO depending on the excess air ratio used if air has been enriched to 29% O<sub>2</sub>. It should be noted that the increase in O<sub>2</sub> concentration decreased the flow of oxidising mixture, so the observed increases of the concentrations of CO and NO are quite large. Figure 4 shows the effect of the oxygen addition to the combustion air for three different excess air ratios  $\lambda_1 - \lambda_3$  of on the concentration of nitrogen oxide NO. Addition of oxygen to the oxidizer also causes an increase in NO concentration in the exhaust gas. NO concentration for all cases is at the same level.

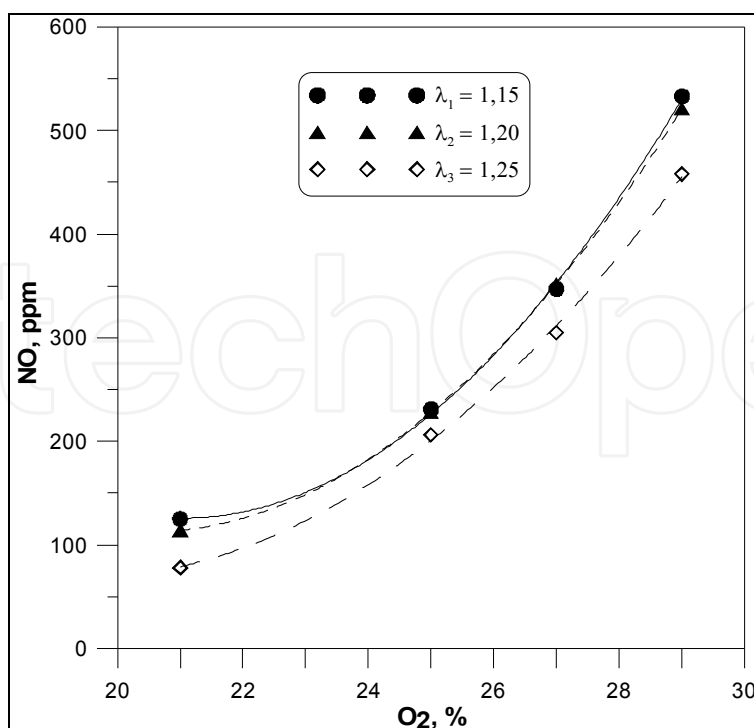


Fig. 4. The effect of oxygen addition to the combustion air on NO concentration in the natural gas combustion process with the oxygen enriched air depending on excess air ratio.

In the second series, the investigations were carried out for a fixed flow of gas and air in natural gas combustion in oxygen-enriched atmosphere. The combustion air was enriched from 21 to 29% oxygen, thereby generating the excess air ratios  $\lambda$  from 1,18 to 1,63. The results are presented in Table 3 and the graphs (Fig. 5 - 7).

$\lambda$	O <sub>2</sub> , %	CO, ppm	NO, ppm	T <sub>exhaust</sub> , °C
1,18	21	23	85	991
1,40	25	42	167	987
1,52	27	50	203	985
1,63	29	71	290	983

Table 4. The results of natural gas combustion in oxygen-enriched atmosphere for  $\dot{V}_g = 0,8 \text{ m}^3/\text{h}$  and  $\dot{V}_{\text{air}} = 9 \text{ m}^3/\text{h}$ ,  $T_{\text{flame}} = 1140 - 1160 \text{ }^\circ\text{C}$ .

Figure 5 shows the effect of oxygen addition to the CO concentration in the combustion of natural gas in oxygen enriched air for constant flows of gas and air ( $\dot{V}_g = 0,8 \text{ m}^3/\text{h}$  and  $\dot{V}_{\text{air}} = 9 \text{ m}^3/\text{h}$ ).

The increase of oxygen concentration in the oxidizer from 21 to 29% O<sub>2</sub>, an increase in CO concentrations up to 300% was observed. Addition of oxygen in the oxidizer does not improve the complete combustion of the gas. CO molecule is more stable than CO<sub>2</sub> and its oxidation by oxygen is very slow, it is unknown whether pure carbon monoxide CO could be burnt (Kotowicz & Janusz, 2007). However, the oxidation of CO is possible by means of even a small concentration of hydrogen and its compounds, e.g. addition of a small concentration of water vapour would cause the CO combustion.

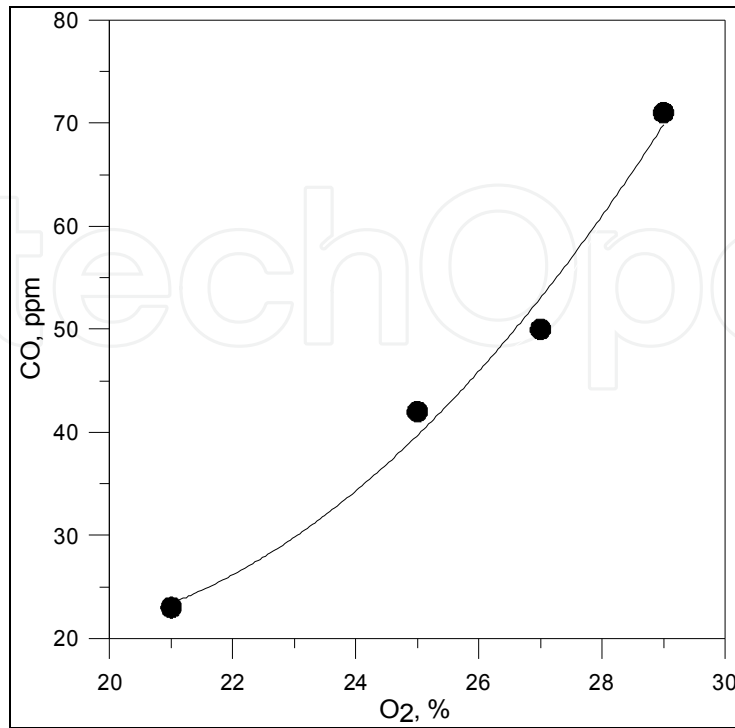


Fig. 5. The effect of oxygen addition on the CO concentration in the natural gas combustion in oxygen-enriched oxidizer for  $\dot{V}_g = 0,8 \text{ m}^3/\text{h}$  and  $\dot{V}_{\text{air}} = 9 \text{ m}^3/\text{h}$ .

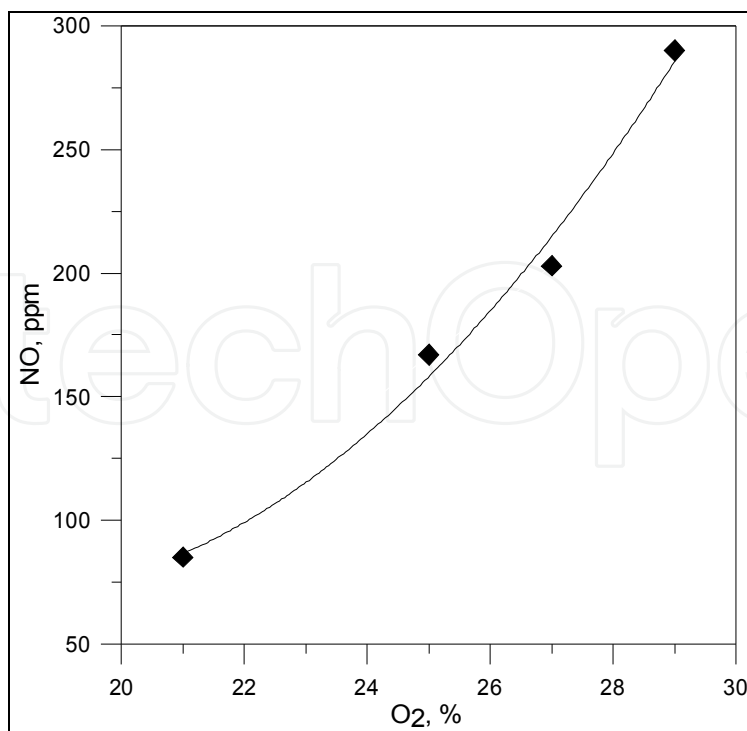


Fig. 6. The effect of oxygen addition on the NO concentration in the natural gas combustion in oxygen-enriched oxidizer for  $\dot{V}_g = 0,8 \text{ m}^3/\text{h}$  and  $\dot{V}_{\text{air}} = 9 \text{ m}^3/\text{h}$ .

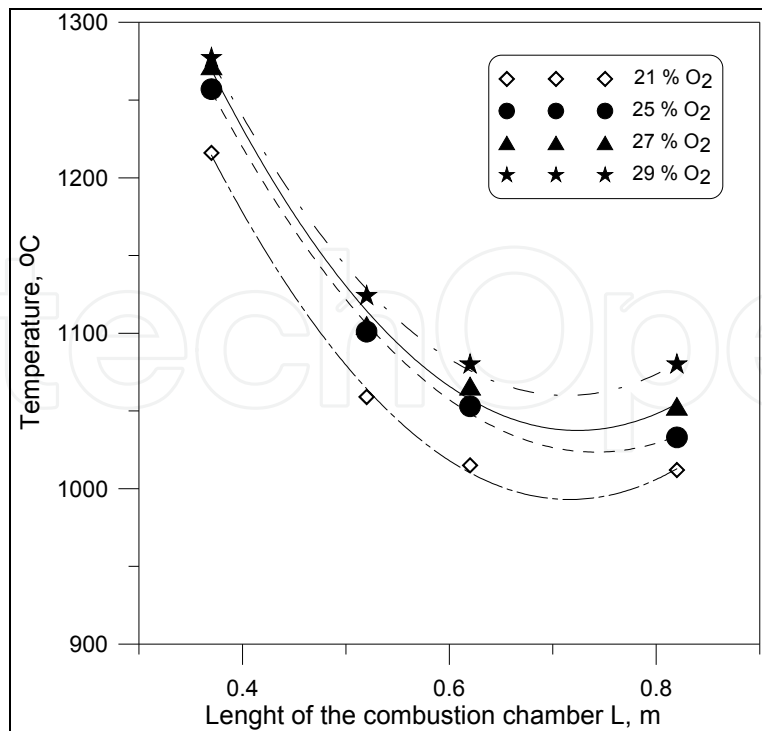


Fig. 7. The exhaust gas temperature profile along the length of the combustion chamber during combustion of natural gas in oxygen-enriched air.

The consequence of fuel combustion in oxygen-enriched atmospheres in  $O_2 + CO_2$  system (replacing nitrogen by  $CO_2$ ), is the reduction of pollutants emissions, mainly  $NO_x$  (Amann et. al, 2009; Li, Yan & Yan, 2009; Muskał et al., 2008). In the studied system (modified atmosphere: oxygen-enriched air combustion) lower concentrations of nitrogen oxide NO are not obtained, but on the contrary, more than threefold increase in NO concentration was observed (Fig. 6). Addition of oxygen increases the flame temperature, and therefore it also increases the NO concentration. Increased concentration of NO may also result from the larger concentration of oxygen in the reacting system, making easier the connection between the air nitrogen and oxygen at high temperature.

The temperature profile along the length of the furnace was also performed (Fig. 7). It was found that the exhaust gas temperature decreased along the furnace chamber in the measured points of the furnace, as well as an increased concentration of oxygen in the oxidizer. Oxygen enrichment of combustion air is done in order to raise the temperature of combustion in the furnace and to raise the growth rate of fuel combustion, which causes shortening of the flame. The reduction of the flame length explains the decrease of the exhaust gas temperature in the measured fixed points along the length of the furnace. The larger was the addition of oxygen, the flame was shorter and the temperature was lower in the test point. The flame temperature with the addition of oxygen increased from 1140 to 1160 °C.

In the course of the experiment it was observed that addition of oxygen resulted in visible changes in shape and colour of the flame, the bright crown of the burner nozzle and a very bright flame colour. Opportunity to observe these changes undoubtedly comes from reduction of the flame length. Adding oxygen to the combustion air also caused a change in



the sound of the furnace operation to the louder, more intense one, associated with changes in the fuel and oxidizer mixture flow within the combustion chamber.

The study of natural gas combustion was also conducted in another modified atmosphere:  $O_2/CO_2/N_2$ . The process was studied under excess air ratio  $\lambda_3 = 1,25$  and the oxidizer was oxygen-enriched up to 27%.  $CO_2$  was added to the oxidizer in the range of 0 to 2  $m^3/h$ .

The effect of carbon dioxide addition on the CO and NO concentrations of the studied process were investigated (Figure 8 and Figure 9). The CO concentration increased with  $CO_2$  addition. The addition of 2  $m^3/h$  of  $CO_2$  has generated two times larger CO concentration comparing to the process operated in conventional atmosphere  $O_2/N_2$ . NO concentration, in contrary, decreased with increasing addition of  $CO_2$ . The maximum of  $CO_2$  addition (2  $m^3/h$ ) decreased NO concentration ca. 12 times. The decrease of NO was connected with lower temperature obtained in the combustion chamber, because of large  $CO_2$  thermal capacity. That fact confirms the flame temperature measured close to the burner nozzle in the axes of the flame presented in Figure 11. The decrease of flame temperature was observed with increasing  $CO_2$  addition to the oxidizer. The thermal NO formation, main way of NO formation during natural gas combustion, takes place just behind the flame front in a zone of high temperature ( $t > 1400$  °C). Therefore, the efficient method of the NO reduction is the lower range of temperature used, shortening the stay of the reagents in the zone of high temperatures and reducing the local concentration of  $N_2$  and  $O_2$ .

The exhaust gas temperature profile was conducted along the combustion chamber including the  $CO_2$  addition (Figure 10). The exhaust gas temperature decreases with  $CO_2$  addition along the combustion chamber. The more  $CO_2$  is added the lower temperature is obtained.

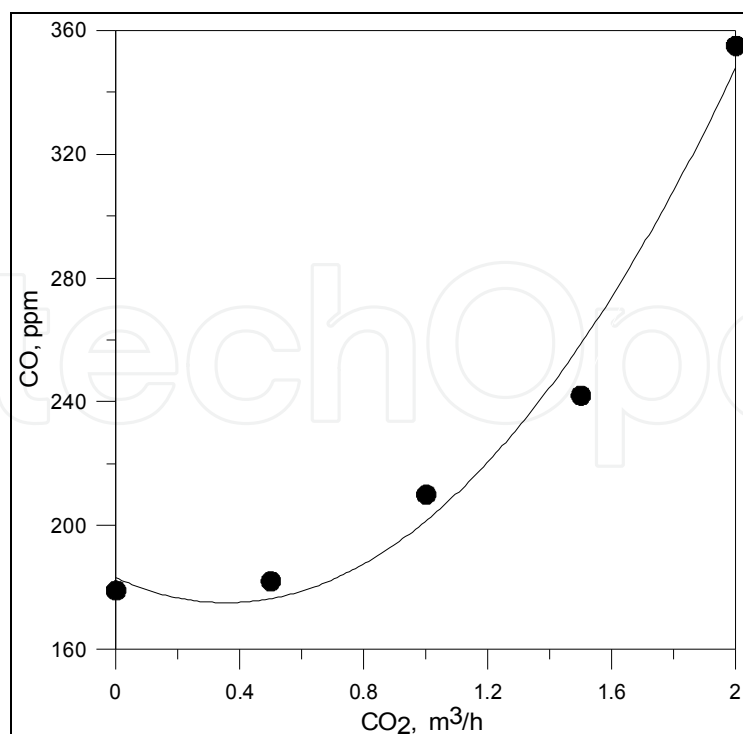


Fig. 8. The effect of carbon dioxide on the concentration of CO in the combustion of natural gas with oxygen-enriched oxidizer up to 27%  $O_2$  and  $\lambda_3 = 1,25$ .

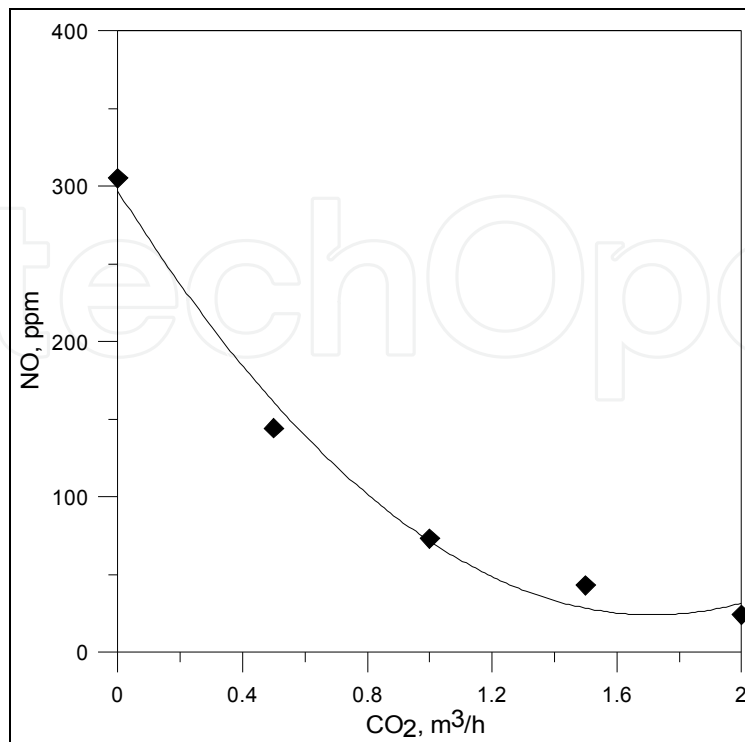


Fig. 9. The effect of carbon dioxide on the concentration of NO in the combustion of natural gas with oxygen-enriched oxidizer up to 27% O<sub>2</sub> and  $\lambda_3 = 1,25$ .

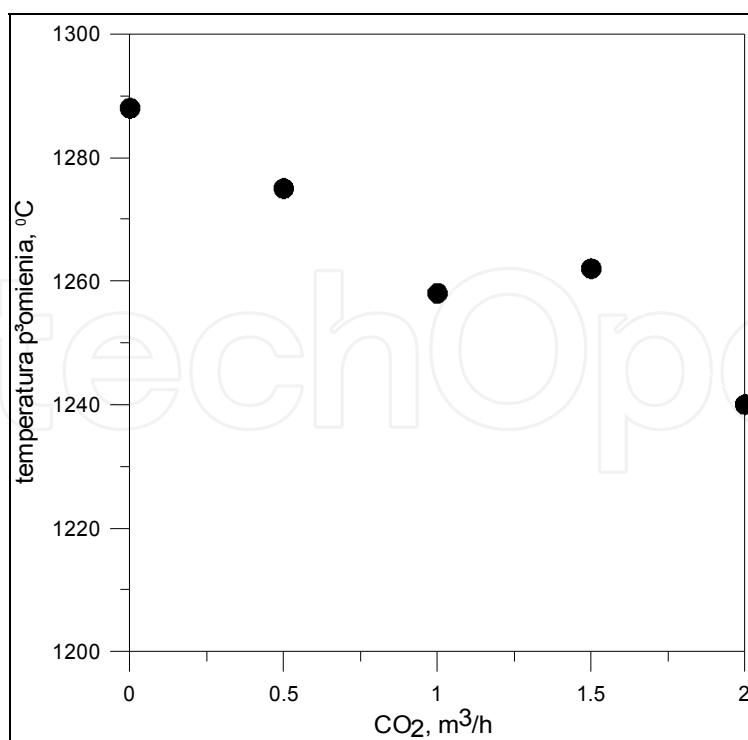


Fig. 10. The effect of carbon dioxide on the flame temperature in the combustion of natural gas with oxygen-enriched oxidizer up to 27% O<sub>2</sub> and  $\lambda_3 = 1,25$ .

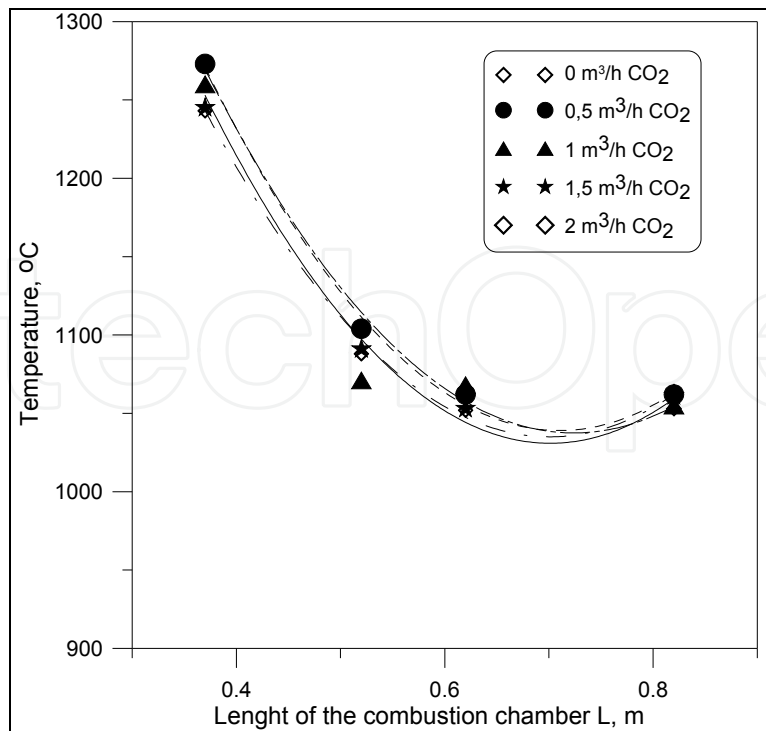


Fig. 11. The effect of carbon dioxide on the exhaust gas temperature measured along the combustion chamber in the combustion of natural gas with oxygen-enriched oxidizer up to 27% O<sub>2</sub> and  $\lambda_3 = 1,25$ .

## 5. Conclusion

In industrial processes, which are required to maintain very high temperatures such as in glass or in the production of cement, the oxy-combustion process can be used, or oxygen-enriched combustion air. This of course raises the temperature of the combustion process, which is associated with an increase in the concentration of NO<sub>x</sub>. Therefore, the oxy-combustion should be used simultaneously with the process of elimination of nitrogen from combustion air (largely responsible for the formation of NO). Otherwise, the addition of oxygen increases the NO concentration, and therefore undesirable effect is achieved. A mixture of oxygen and carbon dioxide by replacing the air can lead to lower concentrations of nitrogen oxides by eliminating nitrogen from combustion air.

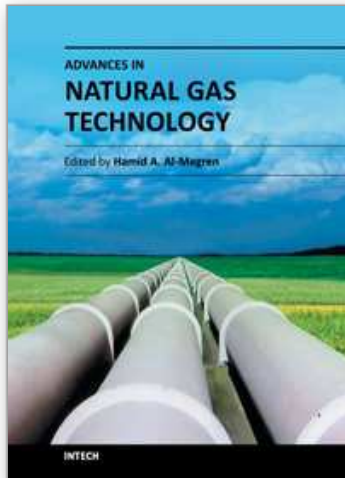
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Natural gas is a vital component of the world's supply of energy and an important source of many bulk chemicals and speciality chemicals. It is one of the cleanest, safest, and most useful of all energy sources, and helps to meet the world's rising demand for cleaner energy into the future. However, exploring, producing and bringing gas to the user or converting gas into desired chemicals is a systematical engineering project, and every step requires thorough understanding of gas and the surrounding environment. Any advances in the process link could make a step change in gas industry. There have been increasing efforts in gas industry in recent years. With state-of-the-art contributions by leading experts in the field, this book addressed the technology advances in natural gas industry.

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