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CARBON MONOXIDE AND NITRIC OXIDE CONCENTRATIONS IN FLUE GAS DURING COMBUSTION OF AGRICULTURAL BIOMASS IN THE RESIDENTAL BOILER

Concentrations of carbon monoxide and nitric oxide (in mg/m^3 and presented for 10% O_2) in the flue gas from firing hay and palm kernel shell pellets, as well as a mixture of coffee husk pellets and wood pellets at the weight ratios of 70:30 and 30:70 in a rotary furnace with a 20 kW nominal heat output amounted to: 695.102; 3492.271; 2921.165; 18344.244; 5297.315, respectively, for the boiler operating with minimum heat output, and 568.192; 2400.341; 4065.275; 20696.380 and 2272.123, respectively, for the boiler operating at a heat output close to maximum. The value of carbon monoxide concentration was proportional to the amount of slag generated during the combustion process and the value of nitric oxide concentration was proportional to the nitrogen content in the fuel. Variation of parameters in time: temperature in the combustion chamber, carbon monoxide and nitric oxide concentrations, oxygen concentration and air excess ratio while firing wood and agricultural biomass pellets were indicated. Correlation between nitric oxide and carbon monoxide concentrations as well as carbon monoxide and nitric oxide concentration versus oxygen concentration and temperature in the combustion chamber were presented. Agricultural biomass pellets should be mixed with wood pellets to improve combustion process and reduce carbon monoxide and nitric oxide concentrations. Pollutant concentration measurements were performed in real-life conditions resembling the ones experienced by boiler end-users.

Keywords: agricultural biomass; combustion; emission; boiler; rotary furnace

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Introduction

Firing agricultural biomass in grate furnaces with low carbon monoxide emissions is challenging [Hartge et al. 2000; Musialik-Piotrowska et al. 2010; Houshfar et al. 2011; Mediavilla et al. 2011a; Ohman et al. 2011; Saez et al. 2011; Verma et al. 2012; Liu et al. 2013; Qui 2013; Garcia et al. 2014; Krugly et al. 2014; Yanquin et al. 2016; Schonnenbeck et al. 2016; Pałaszyńska and Juszczak 2018] in comparison with firing wood [Johansson et al. 2004; Kjallstrand and Olsson 2004; Verma et al. 2009; Francisco Josephinum Wieselburg BLT 2010; Juszczak 2010; 2011; 2014b; 2016a, b; Boman et al. 2011; Dzurenda et al. 2015; 2017; Verma et al. 2011a, b], because as soon as the temperature gets a little below 800°C the slagging process originates in case of many agricultural biomass types [Verma et al. 2013]. On the other hand, carbon monoxide oxidizes well to carbon dioxide at the temperature of above 650°C [Nussbaumer 2003]. It is extremely difficult to ensure that end-users maintain such a narrow temperature range in real-life combustion conditions. Many examples of firing agricultural biomass on grates prove slag generation, which hampers the combustion process causing considerable carbon monoxide emission [Musialik-Piotrowska et al. 2010; Ohman et al. 2011; Saez et al. 2011; Yanquin et al. 2016; Pałaszyńska and Juszczak 2018]. The intensity of slagging depends on ash chemical composition [Colannino 2006; Vassilev et al. 2010; 2013a, b; Wopienka et al. 2011; Nunes et al. 2016]. The combustion process can be improved by mixing agricultural biomass with wood or other organic materials, e.g cork [Garcia-Cuevas et al. 2011; Mediavilla et al. 2011b; Mignon et al. 2011; Juszczak and Lossy 2012; Cioabla et al. 2015]. Ash softening and melting temperatures can be raised using certain additives, such as halloysite. Experiments using this approach in power plant boilers showed considerable slagging reduction, improvement of combustion conditions and decreased emission of incomplete combustion products [Juszczak 2014b]. In low heat output boilers, however, using these agents may lead to the building-up of deposits in the fire tubes, which compromises boiler heat efficiency, as well as results troublesome for the end-users, who are forced to clean the boiler on a daily basis [Garcia-Cuevas et al. 2011; Juszczak and Lossy 2012].

Out of all the existing grates, one that is seemingly the best for agricultural biomass firing is the moving step grate, as the reciprocating movement of the pushing-bars allows for slag disintegration. In this case, however, we must not exceed the ash softening temperature or otherwise the slag will get sticky, adhere to the pushing-bars and cannot be eliminated.

For the reasons presented above, the most adequate technique of agricultural biomass burning seems to be two stage combustion, consisting in biomass gasification followed by burning of the generated gas. In this case, the temperature in the gasification chamber usually ranges between 300 and 500°C. This is well below ash melting temperature and therefore slagging can be avoided. What can obviously still be problematic is proper gas combustion in order to maintain the concentrations of carbon monoxide, hydrocarbons and nitric oxide is as low as possible. Two stage combustion in domestic boilers with a heat output of roughly 20-50 kW is uncomfortable due to the fact that the fuel is supplied manually. Also, on occasions when too much air reaches to the gasification chamber micro-explosions can occur.

In any case, low heat output boilers (20-50 kW) designed specifically for two stage combustion of agricultural biomass (in the form of briquettes and pellets) are not available on the market. Therefore, for the purposes of firing agricultural biomass pellets different types of furnaces are often used in low heat output boilers (20-50 kW), namely retort or horizontal-feed furnace, which are however designed for wood pellets. These furnaces are placed in small combustion chambers of boilers designed for wood pellet firing. Temperature in these furnaces usually exceeds agricultural biomass ash sintering temperature (but not wood ash), which results in slagging hampering furnace operation and elevating emissions of incomplete combustion products, i.e. carbon monoxide and hydrocarbons.

In order to fire biomass properly, special furnaces and bigger combustion chambers are needed, so that the temperature does not exceed the ash sintering temperature for agricultural biomass (often 750-800°C). This is why it is usually beneficial to mix agricultural biomass with wood, because ash sintering temperature for wood pellets is higher (above 1000°C) [Verma et al. 2011a].

The aim of this study is to examine the operation of a new kind of furnace with rotary combustion chamber with regard to firing agricultural biomass pellets and determine carbon monoxide and nitric oxide concentrations in the flue gas while burning the selected kinds of biomass. There are reasons to believe that the rotary movement of the combustion chamber would cause the generated slag to ultimately fall down and disintegrate, and thus combustion air access and the furnace operation would not be compromised and carbon monoxide concentration in the flue gas would not be as elevated.

Materials and methods

The study used agricultural biomass pellets made from wood, hay, palm kernel shells, coffee husks, as well as a mixture of coffee husk pellets and wood pellets at two different weight ratios, i.e. 70:30 and 30:70 [%] (Fig. 1). The pellets were cylindrical in shape, with a diameter of 8 mm and variable length of 10-20 mm. The ultimate analysis of the biomass used in the study and its lower heating value was presented in Table 1. The ultimate analysis was established in our accredited laboratory and the lower heating value was determined according to [Mroczek et al. 2011].

The moisture war established at 7% for all types of pellets.

Table 1. Ultimate analysis and lower heating value of the biomass used in the study

Biomass type /parameters	Unit	Wood	Hay	Palm kernel shell	Coffee husks	Coffee husks/wood 70:30 (weight proportion)	Coffee husks/wood 70:30 (weight proportion)
C	%	56.80	45.69	46.62	48.8	51.2	54.4
H	%	6.30	6.44	4.80	4.90	5.32	5.88
N	%	0.22	1.00	0.61	0.60	0.49	0.33
S	%	0.08	0.39	0.56	0.60	0.44	0.24
Lower heating value	KJ/kg	19080	15120	18120	18406	18610	18880

a) wood pellets

b) hay pellets

c) palm kernel shell pellets

d) coffee husk pellets

e) mixture of coffee husk pellets and wood pellets weight ratios 70:30

f) mixture of coffee husk pellets and wood pellets weight ratios 30:70

Fig. 1. Agricultural biomass types used in the study

The furnace used in the study (Fig. 2) – manufactured by Kippy Suchy Las – has a rotary combustion chamber with rotational speed regulation. As the combustion chamber rotates, the generated slag falls onto the bottom of the furnace gravitationally after reaching the highest position in the chamber. Each subsequent fuel load introduced to the combustion zone pushes the slag out of the furnace, which makes it a self-cleaning burner. Air stream for combustion can be regulated by adjusting fan resolutions.

The aforementioned rotary furnace was installed in a 20 kW boiler type Biomax manufactured by Lumo, which is located in a full scale experimental heat station belonging to the Poznan University of Technology, Institute of Environmental Engineering. Division of Heating, Air Conditioning and Air Protection. The heat station is connected to the district heating network, heat transfer unit, heat receivers: radiators and water heat storages. Therefore, it can be assumed that the pollutant emission measurements were performed in real-life conditions resembling the ones experienced by boiler end-users.

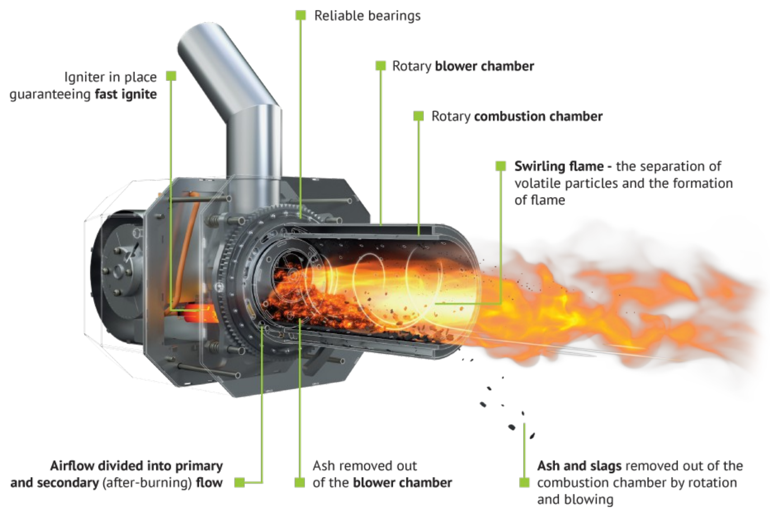

Fig. 2. Furnace with rotary combustion and blower chamber (Source: manufacturer's website, www.kipi.pl)

Concentrations of: oxygen, carbon monoxide, nitric oxide, nitrogen dioxide and hydrocarbons (transformed to methane) in the flue gas downstream the boiler were measured using Vario Plus flue gas analyzer (MRU brand). Oxygen, nitric oxide (NO) and nitrogen dioxide (NO_2) concentrations were measured with electrochemical cells, while carbon monoxide and hydrocarbon concentrations using the infrared procedure. NO_x concentration was calculated with the gas analyzer by summing up the concentration of NO (transformed to NO_2) and NO_2. The flue gas analyzer also measured the flue gas temperature with electrochemical cells, while carbon monoxide and hydrocarbon concentrations using the infrared procedure. NO_x concentration was calculated with the gas analyzer by summing up the concentration of NO (transformed to

Fig. 3. 20 kW boiler with a pellet furnace equipped with rotary combustion chamber, installed in a full scale experimental heat station; thermocouple above the furnace

NO_2) and NO_2. The flue gas analyzer also measured the flue gas temperature downstream the boiler and calculated chimney loss and air excess ratio. Heat received by the boiler water and boiler heat output were measured with Kamstrup ultrasonic heat meter. The temperature in the combustion chamber was measured with a thermocouple PtRhPt that was radiation shielded in order to reduce the negative effect of radiation. The boiler heat efficiency was calculated as heat transferred to the boiler water divided by fuel mass multiplied by fuel lower heating value. Fuel mass was determined with a Sartorius lab balance. Fuel moisture content was determined with the weighing method using a dryer, and the higher heating value, as well as the lower heating value, by the bomb calorimetric method [Mroczek et al. 2011].

Several-hour test runs were performed for four types of agro-pellets as well as for wood pellets (for comparison purposes). In order to describe the value of the amplitude of fluctuations of the different measured parameters, this continuous measurement was divided into half-hour periods, for which then averaged values were calculated. Treating these half-hour periods as separate measurements, mean value was determined based on these partial mean values and uncertainty intervals were calculated with a 95% confidence level (Table 2 and 3). Air stream for combustion was set at the beginning of each test run (and then remained constant), so that carbon monoxide concentration in the flue gas downstream the boiler was as low as possible and the temperature in the combustion chamber could be maintained in the range between 650°C and 750°C. All the measured parameters were recorded continuously and transferred in real time to computer memory, where they were registered every 5 seconds for averaged value calculation. The temperature measured with a thermocouple placed about 0.2 m above the combustion chamber (Fig. 3), indicated in Table 2 and 3 as „temperature in the combustion chamber”, is approx. 100°C lower than the temperature in the rotary combustion chamber. This was confirmed by the indications of the pyrometer.

Results and discussion

Measurement results for minimum boiler heat output and heat output close to its maximum value were presented in Table 2 and 3. Gas analyzer detected hydrocarbon concentration only on rare occasions, thus the detected values were not included in Tables 2 and 3 as negligible.

Table 2. Mean parameter values from several-hour test runs examining the combustion of agricultural biomass pellets in a rotary furnace operating with minimum heat output

Parameters	Unit	Wood	Hay	Palm kernel shell	Coffee husk/wood 70:30	Coffee husk/wood 30:70
Heat output	kW	9.4 ±1.9	10.6 ±1.2	10.7 ±0.9	10.7 ±0.8	12.0 ±1.1
Fuel mass	kg	16.200	24.400	21.600	22.000	11.300
Boiler heat efficiency	%	88 ±0.4	83 ±0.9	79 ±0.7	75 ±0.9	81 ±0.6
Temperature in the combustion chamber	C	412 ±22	490 ±30	451 ±35	446 ±26	511 ±27
Flue gas temperature downstream the boiler	C	147 ±7	148 ±8	161 ±8	159 ±4	182 ±9
O_2 concentration	%	15.0 ±0.9	11.3 ±0.6	13.6 ±0.7	13.3 ±0.9	10.9 ±0.8
CO_2 concentration	%	6.1 ±1.0	10.2 ±0.5	7.1 ±0.7	6.7 ±1.1	10.6 ±0.8
CO concentration	mg/m^3 (10% O_2)	695 ±46	3492 ±1072	2921 ±540	18344 ±1494	5297 ±1196
NO concentration	mg/m^3 (10% O_2)	102 ±9	271 ±49	165 ±15	244 ±33	315 ±20
NO_x concentration	mg/m^3 (10% O_2)	156 ±14	416 ±75	253 ±23	374 ±51	483 ±31
Time of experiment	h	8	8	8	8	8
Air excess ratio	–	3.4 ±0.5	2.1 ±0.3	2.9 ±0.4	3.1 ±0.5	1.9 ±0.2

Table 3. Mean parameter values from several-hour test runs examining the combustion of agricultural biomass pellets in a rotary furnace operating with a heat output close to maximum

Parameters	Unit	Wood	Hay	Palm kernel shell	Coffee husk/wood weight ratio 70:30	Coffee husk/wood weight ratio 30:70
Heat output	kW	18.6 ±1.9	18.2 ±1.4	22.0 ±2.0	18.7 ±1.6	17.2 ±1.8
Fuel mass	kg	30.600	40.400	42.000	37.200	30.800
Boiler heat efficiency	%	92 ±0.4	86 ±3	82 ±4	78 ±5	85 ±5
Temperature in the combustion chamber	C	537 ±22	509 ±52	577 ±27	602 ±20	509 ±46
Flue gas temperature downstream the boiler	C	184 ±7	174 ±2	179 ±10	183 ±14	166 ±9
O_2 concentration	%	10.0 ±0.9	8.3 ±0.4	8.1 ±1	8.1 ±0.5	17.3 ±0.3
CO_2 concentration	%	11.2 ±1.0	14.0 ±0.4	12.6 ±1.7	12.0 ±0.5	3.8 ±0.4
CO concentration	mg/m^3 (10% O_2)	568 ±46	2400 ±334	4065 ±365	20694 ±2269	2272 ±983
NO concentration	mg/m^3 (10% O_2)	192 ±9	341 ±34	275 ±26	380 ±32	123 ±13
NO_x concentration	mg/m^3 (10% O_2)	294 ±14	523 ±52	422 ±40	583 ±49	189 ±20
Time of experiment	h	8	8	8	8	8
Air excess ratio	–	1.9 ±0.3	1.5 ±0.3	1.7 ±0.4	1.7 ±0.3	5.5 ±1.0

Fig. 4. Burner during firing a mixture of coffee husk pellets and wood pellets at a weight ratio of 70:30. First and second figure from the left: burner clogged with slag (partly and completely); third figure: burner after having been cleaned manually

Fig. 5. Deposits in the boiler after firing a mixture of coffee husk pellets and wood pellets at a weight ratio of 70:30

The diagrams shown in Figures 6, 10, 14 illustrate the fluctuations of different combustion parameters (temperature in the combustion chamber, carbon monoxide and nitric oxide concentrations, oxygen concentration and air excess ratio) in time for different kinds of pellets used in the study.

Figures 9, 13, 17 show the correlation between nitric oxide concentration and carbon monoxide concentration in the flue gas for different kinds of pellets used in the study.

The results also reflect how carbon monoxide concentration (Figs. 7, 11, 15), as well as nitric oxide concentration (Figs. 8, 12, 16) depend on oxygen concentration and temperature in the combustion chamber for the analyzed pellet types.

Carbon monoxide concentration is in this case an indicator of the combustion process quality: the lower it is, the higher the quality of combustion. The limit value for carbon monoxide in the flue gas for boilers with a heat output of up to 500 kW and automated fuel supply according to the country regulations currently in force is set at 3000 mg/m^3 (presented for 10% O_2 concentration in the flue gas) [EN 14918: 2009]. In case of wood pellets, carbon monoxide concentration was below this value. However, for agricultural biomass pellets, it was only a little below the legal limit or, more often, this limit was significantly exceeded, especially in case of the mixture of coffee husk pellets and wood pellets at a weight ratio of 70:30 (Table 1 and 2). This shows the differences in the quality of the combustion process for wood pellets and agricultural biomass pellets. Combustion quality can be also seen while observing the fluctuation of different parameters of the combustion process in time (Fig. 6, 10, 14). The higher the amplitude of parameter fluctuations, the worse and less stable the combustion.

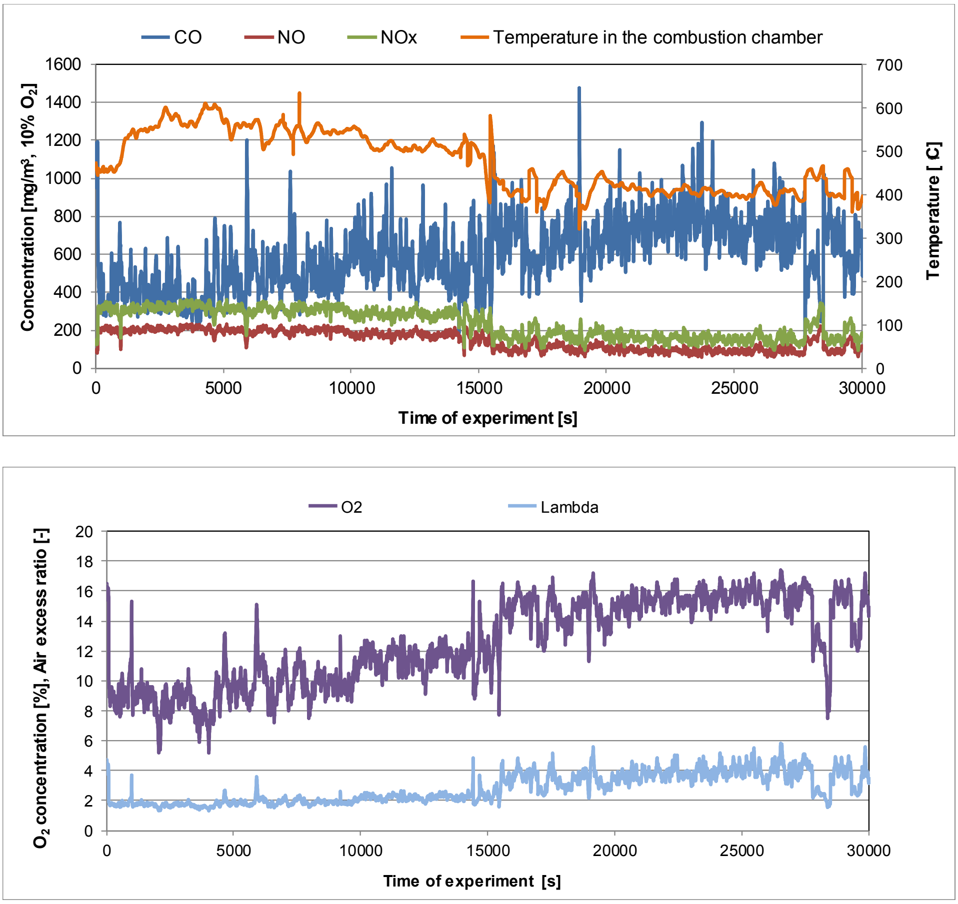

Fig. 6. Variation of parameters in time: temperature in the combustion chamber, carbon monoxide and nitric oxide concentrations, oxygen concentration and air excess ratio while firing wood pellets

In terms of the intensity of slag and deposit generation, firing coffee husk pellets alone, without mixing them with wood pellets, resulted in clogging of the rotary combustion chamber. While firing a mixture of coffee husk pellets and wood pellets deposits were created in the boiler (Fig. 5), which hampered heat exchange between the flue gas and boiler water, causing boiler heat efficiency to diminish as a consequence. High carbon monoxide concentrations were due to the generation of slag, which obstructed air access to the fuel, thus hampering the combustion process and reducing the temperature in the furnace. It can be assumed that the value of carbon monoxide concentration was proportional to the amount of generated slag.

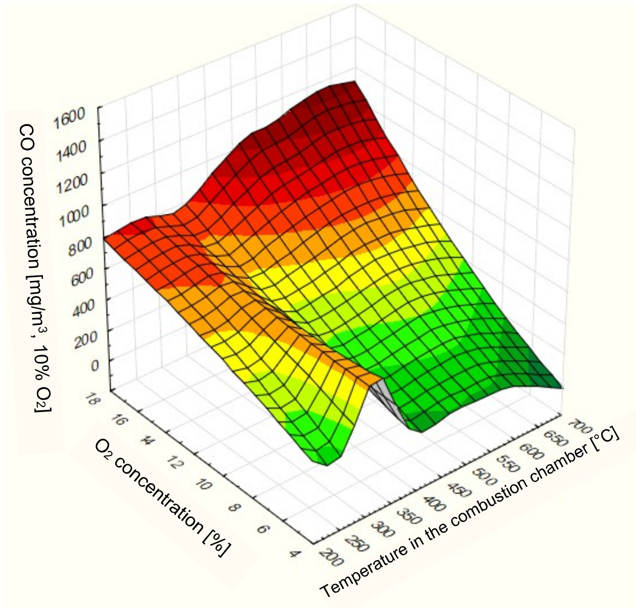

Fig. 7. Carbon monoxide concentration versus oxygen concentration and temperature in the combustion chamber while firing wood pellets

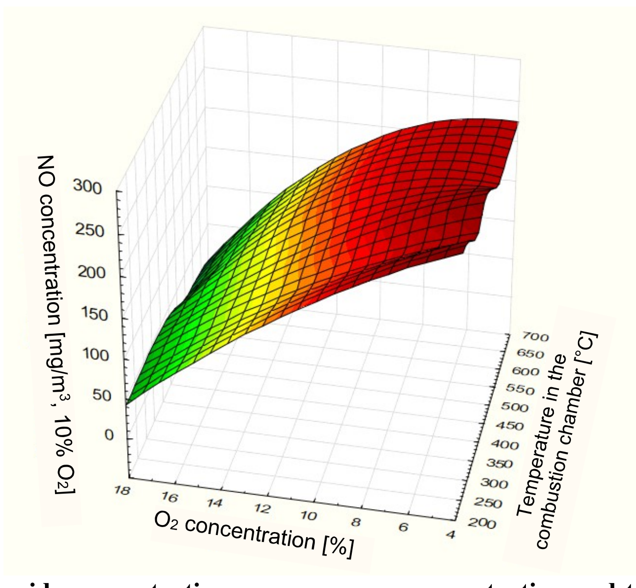

Fig. 8. Nitric oxide concentration versus oxygen concentration and temperature in the combustion chamber while firing wood pellets

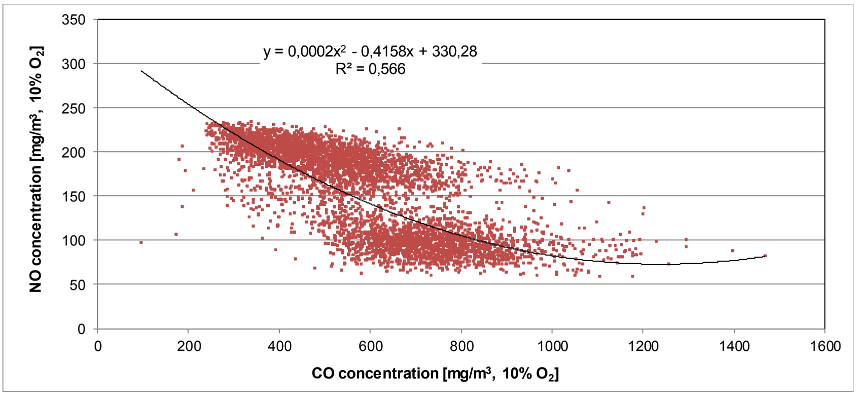

Fig. 9. Correlation between nitric oxide concentration and carbon monoxide concentration while firing wood pellets

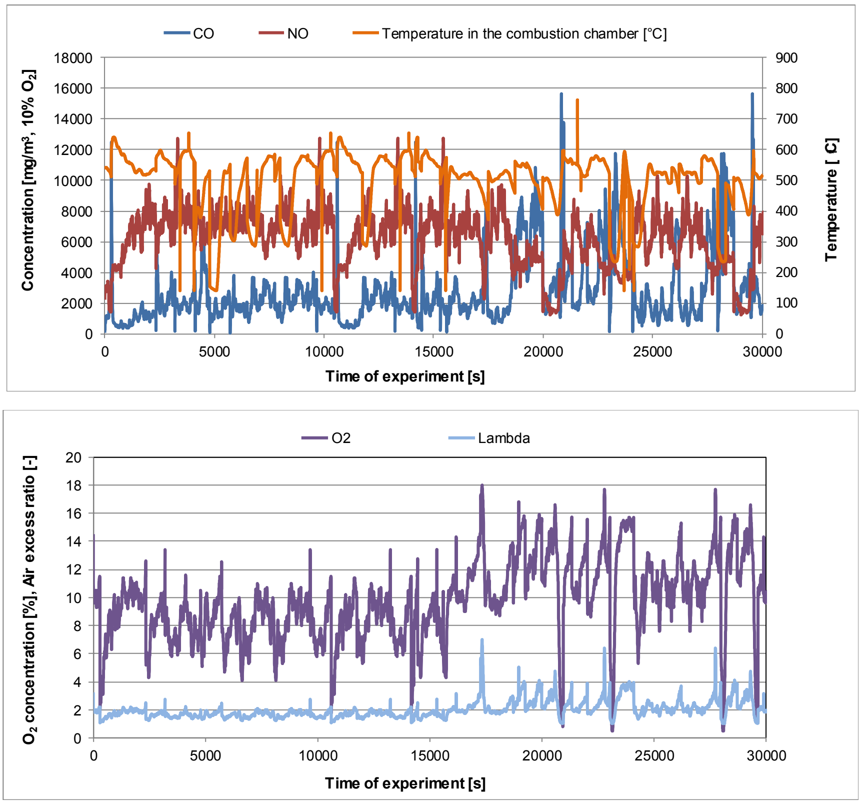

Fig. 10. Variation of parameters in time: temperature in the combustion chamber, carbon monoxide and nitric oxide concentrations, oxygen concentration and air excess ratio while firing hay pellets

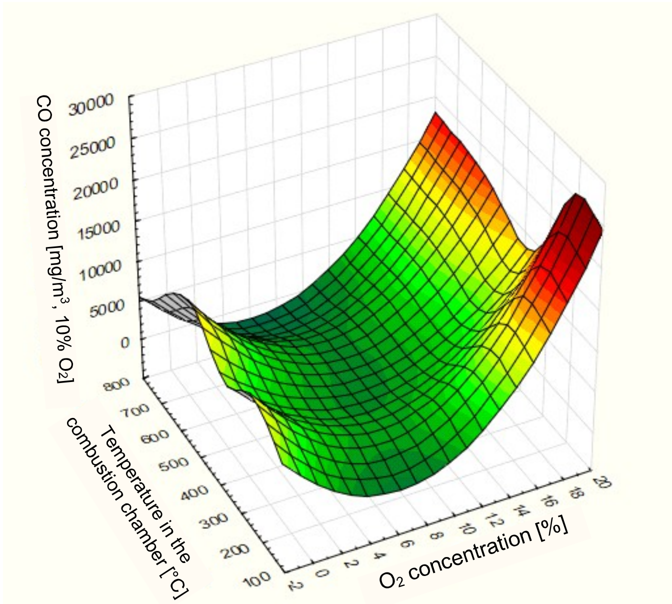

Fig. 11. Carbon monoxide concentration versus oxygen concentration and temperature in the combustion chamber while firing hay pellets

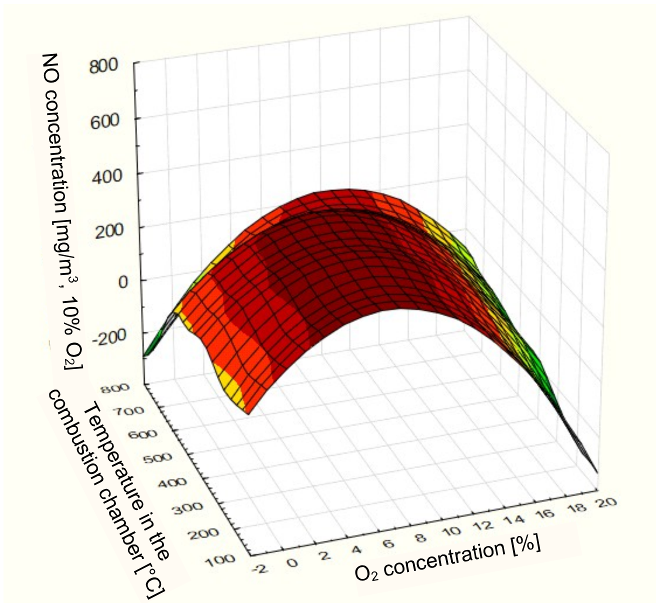

Fig. 12. Nitric oxide concentration versus oxygen concentration and temperature in the combustion chamber while firing hay pellets

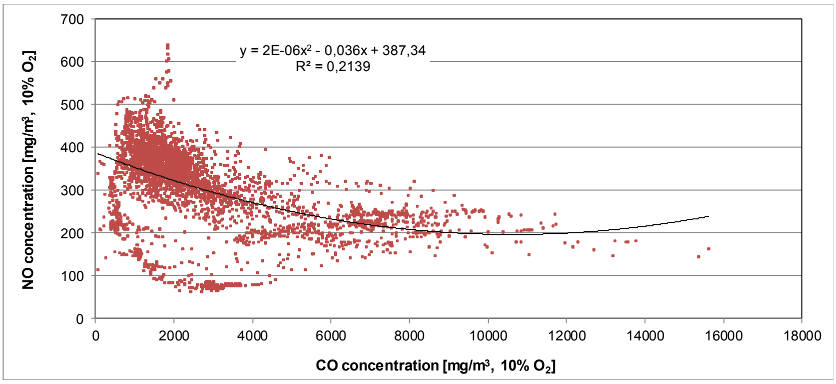

Fig. 13. Correlation between nitric oxide concentration and carbon monoxide concentration while firing hay pellets

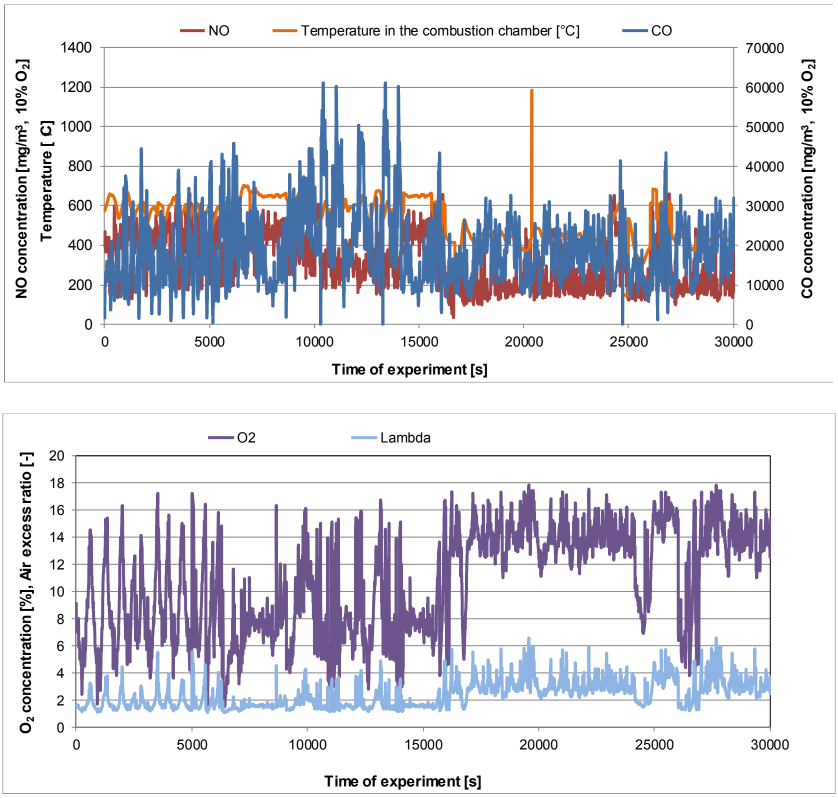

Fig. 14. Variation of parameters in time: temperature in the combustion chamber, carbon monoxide and nitric oxide concentrations, oxygen concentration and air excess ratio while firing a mixture of coffee husk pellets and wood pellets at a weight ratio of 30:70

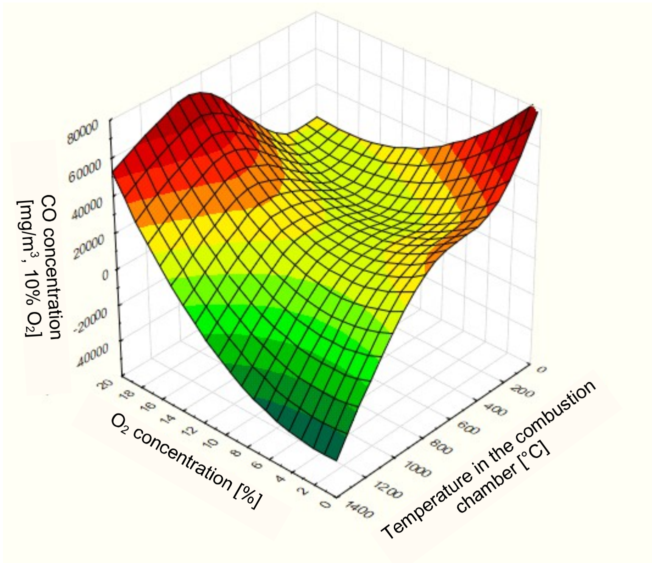

Fig. 15. Carbon monoxide concentration versus oxygen concentration and temperature in the combustion chamber while firing a mixture of coffee husk pellets and wood pellets at a weight ratio of 30:70

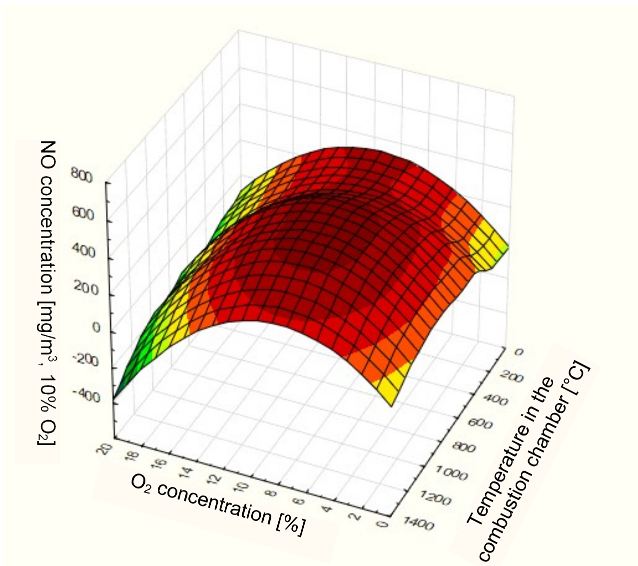

Fig. 16. Nitric oxide concentration versus oxygen concentration and temperature in the combustion chamber while firing a mixture of coffee husk pellets and wood pellets at a weight ratio of 30:70

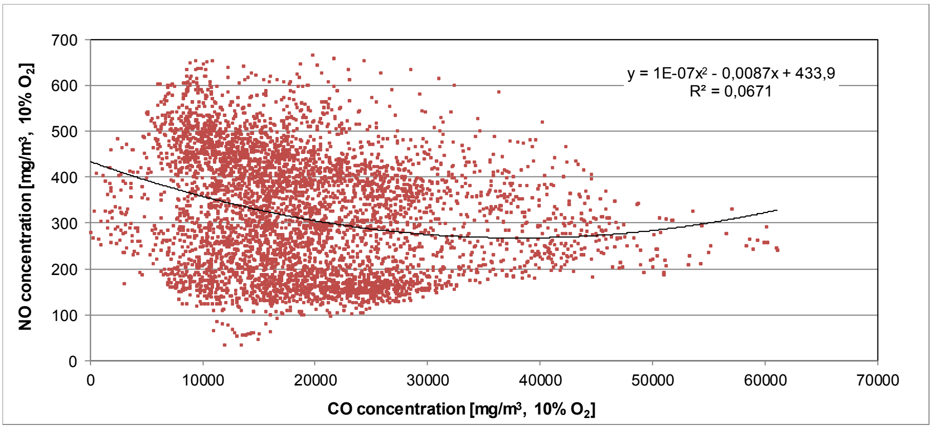

Fig. 17. Correlation between nitric oxide concentration and carbon monoxide concentration while firing a mixture of coffee husk pellets and wood pellets at a weight ratio of 30:70

In the range of temperatures obtained in the combustion zone (below 1000°C), only nitric oxide was generated. However, for comparison with legal regulations, the concentration of nitric oxide is calculated to nitrogen dioxide (NO_2) and expressed as NO_x (Table 2 and 3).

The concentration of carbon monoxide and nitric oxide in the flue gas depend to a large extent on oxygen concentration and temperature in the combustion chamber (Figs. 7, 8, 11, 12, 15, 16). Additionally, the concentration of nitric oxide depends on the nitrogen content in the fuel.

Generally speaking, carbon monoxide concentration decreases with an increase in oxygen concentration at first and then increases. The values of oxygen concentration at which the minimum carbon monoxide concentration is achieved, should serve to set an adequate combustion air flow value. Also, in general terms, carbon monoxide concentration decreases with an increase in temperature in the combustion chamber. As far as nitric oxide concentration is concerned, it increases with an increase in oxygen concentration and temperature in the combustion chamber .

It is important to note that nitric oxide concentration and carbon monoxide concentration also mutually affect each other. The surfaces representing carbon monoxide concentrations (Figs.7, 11, 15) and nitric oxide concentrations (Figs. 8, 12, 16) have ambiguous shapes among other reasons because carbon monoxide reduces nitric oxide to molecular nitrogen, at the same time oxidizing itself to carbon dioxide, according to the following formula:

$$CO + NO = \frac{1}{2} N_2 + CO_2 \quad \text{[Nussbaumer 2003]}$$

The above formula is reflected in Figures 7, 13,17 as a correlation between the concentrations of nitric oxide and carbon monoxide. For agricultural biomass pellets this correlation is low (Fig. 13,17), significantly lower than for wood pellets (Fig. 9). Probably, it is a consequence of instable combustion.

Conclusions

The furnace with rotary combustion chamber is well suited for firing wood pellets. In case of burning agricultural biomass pellets, however, some combustion-related problems occur, resulting from slagging. For this reason, this particular type of furnace can only be recommended for agricultural biomass combustion to users who generate agricultural residues anyway (agricultural holdings). Obtaining lower boiler heat efficiency does not seem to be problematic for the farmers, because they have large quantities of free-of-charge biomass at their disposal.

In case of experiencing combustion-related difficulties and furnace clogging while firing agricultural biomass pellets, they should be mixed with wood pellets in a proportion that would considerably reduce slag generation and ensure maintaining carbon monoxide concentration below the legally permitted value [PN-EN 303-5: 2012], so that the furnace can operate with no disturbances and doesn't get clogged.

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List of standards

EN 14918:2009 Solid biofuels. Determination of calorific value

PN-EN 303-5:2012 Heating boilers, part5. Heating boilers for solid fuels, hand and automatically stocked nominal heat output of up to 300 kW. Terminology, requirements and marking

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