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CARBON MONOXIDE AND NITRIC OXIDE CONCENTRATIONS IN THE FLUE GAS DURING THE COMBUSTION OF AGRICULTURAL BIOMASS BRIQUETTES IN A 25 KW LOG GASIFICATION BOILER

A study was made of the two-stage combustion of agricultural biomass briquettes (rye straw, miscanthus, hay, corn stover) in a 25 kW wood log gasification boiler. The following correlations for selected fuel loads and biomass types were shown: fluctuation of temperature over time both in the gasification chamber and in the combustion zone, variation over time of carbon monoxide and nitric oxide concentrations, correlation between nitric oxide and carbon monoxide concentrations in the flue gas, and variation of carbon monoxide and nitric oxide concentrations versus oxygen concentration and temperature in the combustion zone. Two-stage combustion of agricultural biomass proved to be efficient, as slag was not generated. Relatively high carbon monoxide concentrations in the flue gas resulted from the lack of automated regulation of air supply to the gasification chamber and the combustion zone.

Keywords: agricultural biomass; combustion; emission; boiler; slagging

Introduction

Firing agricultural biomass in grate furnaces with low carbon monoxide emission is much more challenging [Saez et al. 2011; Verma et al. 2012; Chandrasekaran et al. 2013; Guoquan 2013; Krugly et al. 2014; Fournel et al. 2015] than burning wood [Boman et al. 2011; Chandrasekaran et al. 2011; Fernandes and Costa 2011; Pettersson et al. 2011; Verma et al. 2011; Calvo et al. 2014; Venturi et al. 2018], since even at temperatures slightly below 800°C slag is generated in many types of agricultural residues [Wopienka et al. 2011; Cioaba et al. 2015; Nunes et al. 2016; Vassilev et al. 2016]. Many studies confirm that slagging is a serious issue when firing agricultural biomass on a

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grate [Musialik-Piotrowska et al. 2010; Dell'Antonia et al. 2011; Mediavilla et al. 2011; Ohman et al. 2011; Juszczak and Lossy 2012; Chandasekaran et al. 2016; Schonnenbeck et al. 2016; Yanoqin et al. 2016]. The slag obstructs access of air to the fuel, which results in significant levels of carbon monoxide production. Carbon monoxide oxidizes well to carbon dioxide only above 650°C [Marutzky 1997]. In order to avoid slag generation and reduce carbon monoxide emission, the combustion process requires temperatures between 650 and 800°C. In case of domestic use, however, such a narrow temperature range is extremely difficult to maintain.

The susceptibility of agricultural biomass to slagging is due to the chemical composition of the ash (high silica and potassium oxide content) [Ohman et al. 2011; Wopienka et al. 2011; Vassilev et al. 2013; Cioaba et al. 2015; Nunes et al. 2016; Vassilev et al. 2016; Yanquin et al. 2016], which sinters at relatively low temperatures. Slagging may be reduced by mixing agricultural biomass with wood or cork [Garcia-Cuevas et al. 2011; Mediavilla et al. 2011; Lajili et al. 2015]. The use of certain additives that elevate the ash softening temperature, such as halloysite, can prevent slagging. This approach was reported to bring positive effects in the case of power plant boilers: slagging was reduced, combustion conditions improved and the emission of incomplete combustion products decreased [Mroczek et al. 2011]. However, in case of low heat output boilers, using these agents may lead to the creation of deposits in the fire tubes, which compromises boiler heat efficiency. This also creates problems for end-users, as they are forced to clean the boiler on a daily basis [Juszczak et al. 2017].

The best kind of grate for agricultural biomass firing seems to be the moving step grate, because the reciprocating movement of the pushing-bars allows for slag disintegration. However, the most beneficial technique for agricultural biomass firing appears to be two-stage combustion, consisting of biomass gasification followed by burning of the generated gas [Garcia et al. 2014]. In this combustion mode, slag is not generated because the temperature in the gasification chamber is below the ash sintering temperature. The main aim of the study was to confirm whether the slagging process is indeed non-existent during two-stage combustion of agricultural biomass briquettes in a particular boiler type. High nitric oxide concentrations can appear during the firing of agricultural biomass, as even in the case of wood combustion the conversion factor of fuel-bound nitrogen to oxides in the flue gas is high [Dzurenda et al. 2015, 2017; Hroncova et al. 2016].

Materials and methods

The study used briquettes from rye straw, hay, miscanthus and corn stover, made in a Biomasser briquetting press, manufactured by Asket. The briquettes can be manufactured directly on agricultural holdings. Prior to briquetting, the straw, hay, miscanthus and corn stover, instead of being milled, were cut into fibers of considerable length, ranging from 60 to 70 mm, which prevents their fast disintegration during the firing process (fig. 4). The briquettes are cylindrical in shape, with a diameter of 80 mm and variable height (mostly 80 mm). The hole in the center of the briquette (of 10 mm diameter) provides a larger surface area, creating a higher combustion rate. The raw material used to make the briquettes was not dried in dryers, and therefore its moisture content was high, ranging between 18% and 30% (the ratio of water mass to wet briquette mass).

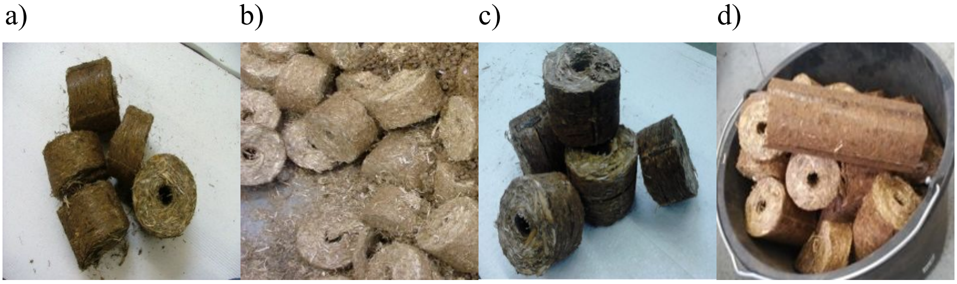

Fig. 1. Briquettes from hay (a), rye straw (b), miscanthus (c) and corn straw (d)

The ultimate analysis of the biomass used in the study comes from [Hartage et al. 2000; Cichy et al. 2009; Komorowicz et al. 2009; Hartmann et al. 2013; EN 14961-2: 2011] and the lower heating value was determined according to [EN 14918: 2009] (table 1). The results obtained in our laboratory were very similar to those presented in table 1.

Table 1. Ultimate analysis, lower heating value and moisture of the biomass used in the study, according to [Cichy et al. 2009; Komorowicz et al. 2009; Hartmann et al. 2013; EN 14961-2: 2011]. Results are presented for dry weight of fuel.

Biomass type/ Parameter	Unit	Rye straw	Hay	Miscanthus	Corn stover
C	%	42.54	45.69	48.15	45.70
H	%	6.12	6.44	6.09	5.30
N	%	0.69	1.00	0.23	0.7
S	%	0.10	0.39	0.05	0.18
Lower heating value	kJ/kg	13950	14100	15570	15100
Moisture	%	24	22	21	19

The boiler (fig. 2) used for experimental purposes was a Vitolig 150 log gasification boiler, manufactured by Viessmann. This boiler uses two-stage combustion. Initially, the fuel is de-gassed in an upper gasification chamber to generate volatile gasses, which then pass through a slotted orifice (nozzle), operating as a low-quality burner, and are fired in the lower combustion chamber.

This particular boiler is of simple and outdated design and low heat efficiency. The boiler heat efficiency measured during the experiments for all of the studied fuels is given in table 2. Air is supplied to the gasification (fuel) chamber and the combustion zone (nozzle) by a fan installed between the fuel chamber and combustion zone doors. The same fan supplies air to the gasification chamber and to the combustion zone. The boiler has no automated regulation of primary or secondary combustion air supply. The airflow is set manually by means of the fan inlet damper at the beginning of each fuel load.

The nozzle, apart from its main function of burning combustible gases, also serves to evacuate ash from the gasification chamber. The ash is then removed from the ash pan manually.

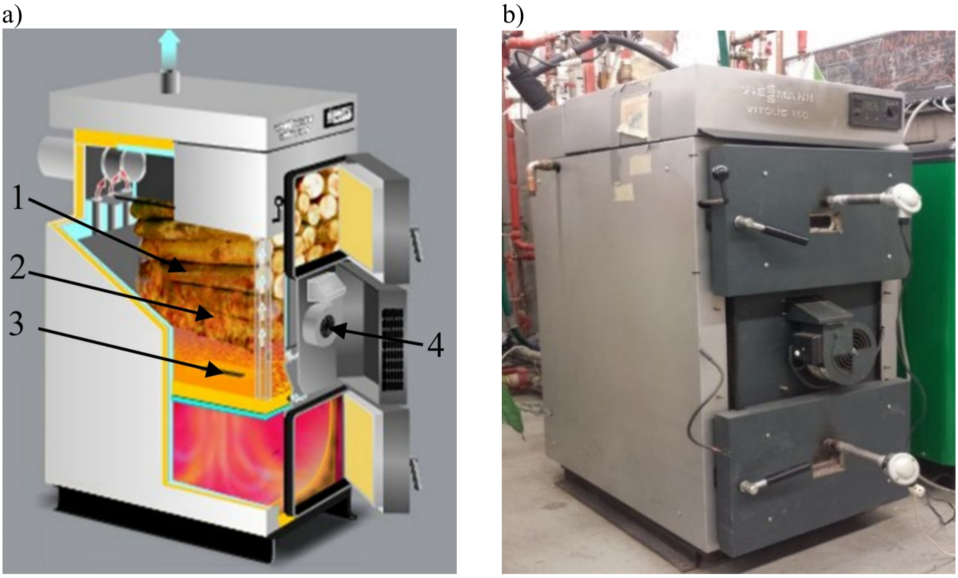

Fig. 2. A 25 kW log gasification boiler with two-stage combustion – gasification and gas combustion in a nozzle located at the bottom of the gasification chamber: a) operating scheme (1 – fuel, 2 – gasification chamber, 3 – nozzle, 4 – fan); b) view of combustion air fan and thermocouples

Concentrations of oxygen, carbon monoxide, nitric oxide, nitrogen dioxide and hydrocarbons (transformed into methane) in the flue gas downstream of the boiler were measured with an MRU Vario Plus flue gas analyzer. Oxygen, nitric oxide (NO) and nitrogen dioxide (NO_2) concentrations were measured using an electrochemical sensor, while carbon monoxide and hydrocarbon concentrations were measured using an IR sensor. NO_x concentration was calculated with the use of the gas analyzer by summing the concentrations of NO (transformed into

NO_2) and NO_2. The flue gas analyzer also measured the flue gas temperature downstream of the boiler, and calculated chimney heat loss and air excess ratio. The heat received by the boiler water and the boiler heat output were measured with a Kamstrup ultrasonic heat meter. The temperature in the combustion chamber was measured with a PtRhPt radiation shielded thermocouple. The boiler heat efficiency was calculated as heat transferred to the boiler water divided by fuel mass multiplied by fuel lower heating value. A Sartorius lab balance was used to determine fuel mass. Fuel moisture content was determined by weighing the fuel before and after drying, and the higher and lower heating values were determined using a bomb calorimeter [EN 14918: 2009].

Two-stage combustion of agricultural biomass briquettes in a 25 kW wood log gasification boiler was examined. For each type of fuel (rye straw, hay, miscanthus, corn stover), four fuel loads were burnt. The average load mass and burning time per load are indicated in table 2. Briquettes were loaded manually into the gasification (fuel) chamber. Air flow for combustion was set manually at the beginning of each test run (and then remained constant), with the purpose of obtaining the lowest possible carbon monoxide concentration in the flue gas downstream of the boiler. All measured parameters were recorded continuously and transferred in real time to computer memory, where they were registered every 5 seconds to calculate the average value.

It should be noted that the experiments were performed in real-life conditions, resembling those existing in end-users' domestic boilers, due to the fact that the boiler is installed in a full-scale heat station (belonging to the Division of Heating, Air Conditioning and Air Protection, Poznan University of Technology, Institute of Environmental Engineering) connected to a district heating network, a heat transfer unit and heat receivers (radiators and water heat storage devices in the building). This ensures more realistic results.

Fig. 3. View of the gasification chamber with open loading door

Fig. 4. Ash with visible fragments of incompletely combusted fuel

Results and discussion

The results of two-stage agricultural biomass combustion performed during the study are given in table 2. Hydrocarbon concentrations were rarely detected by the gas analyzer or were negligible; this parameter is therefore omitted from table 2.

Table 2. Mean parameter values from the combustion of rye straw, hay, miscanthus and corn stover briquettes (four loads per fuel type) in a 25 kW two-stage log gasification boiler

Parameters	Unit	Rye straw	Hay	Miscanthus	Corn stover
Heat output	kW	10.8	9.8	11.2	11.7
Boiler heat efficiency	%	70	63	61	67
Temperature in the combustion chamber	°C	610	600	620	650
Temperature in the gasification chamber	°C	250	240	270	260
Flue gas temperature downstream of the boiler	°C	140	160	150	165
O_2 concentration	%	12.3	9.2	11.0	10.3
CO concentration	mg/m^3 (10% O_2)	5800	3550	4600	4350
NO concentration	mg/m^3 (10% O_2)	470	350	310	290
NO_x concentration	mg/m^3 (10% O_2)	720	540	470	440
Air excess ratio	–	2.4	1.8	2.1	2.0
Burning time per fuel load	h	3.9	3.8	3.6	3.8
Fuel load mass	kg	15.520	15.060	15.220	15.780

The NO concentrations presented in table 2 are very high despite the low temperatures in the combustion chamber and high CO concentrations. This is probably due to the fact that the boiler combustion chamber is small. For this reason, the time of passage of the flue gases through the combustion chamber is too short for the reduction of nitric oxide (NO) to nitrogen (N_2) to occur satisfactorily (according to the following formula: $NO + CO = ½ N_2 + CO_2$). A similar phenomenon was observed in our laboratory in the case of firing of wood pellets in 20 kW boilers. The diagrams in figures 5 and 7 illustrate fluctuations of temperature over time, both in the combustion chamber (nozzle) and in the gasification chamber, for a selected fuel load of each type of agricultural biomass used in the study. In turn, figures 6 and 8 show the variation of CO, NO and NO_x concentrations over time, corresponding to the fuel loads and temperature fluctuations presented in figures 5 and 7.

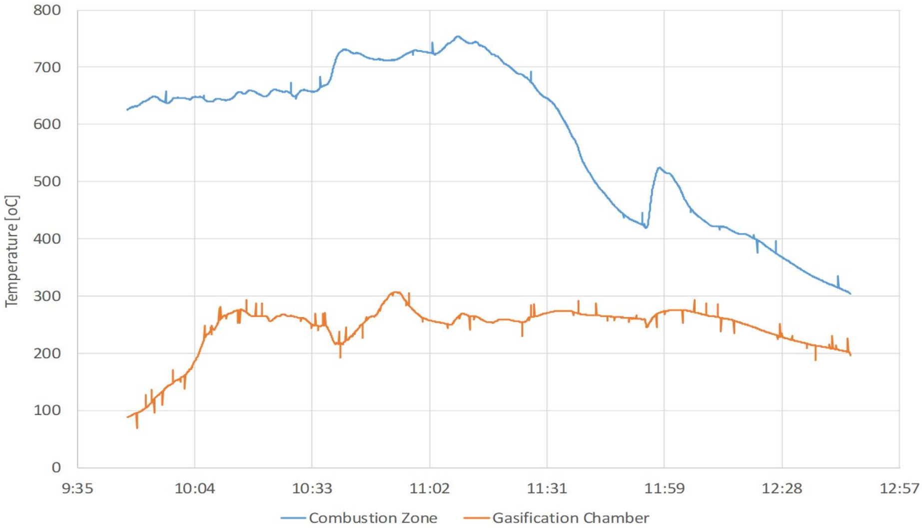

Fig. 5. Temperature fluctuations over time: in the combustion zone (nozzle) and in the gasification chamber for rye straw briquettes

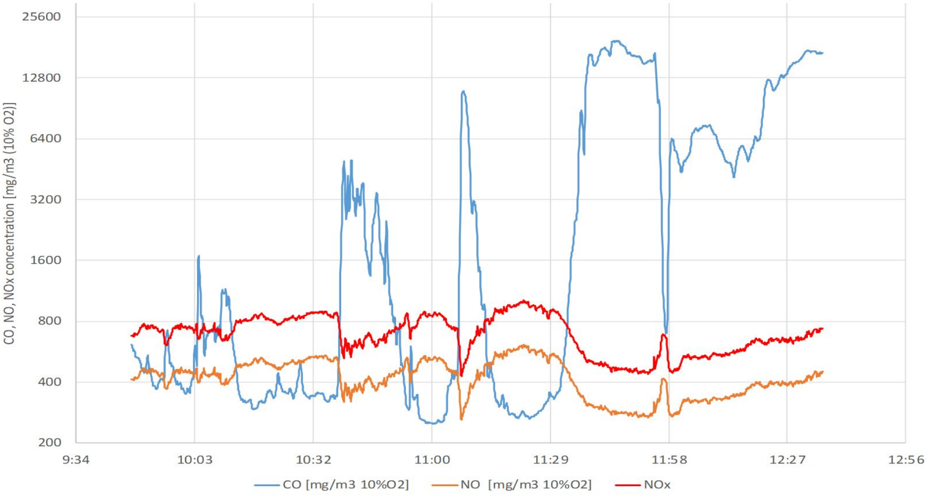

Fig. 6. Variation over time of carbon monoxide (CO), nitric oxide (NO) and nitrogen oxides (NO_X) concentrations for rye straw briquettes

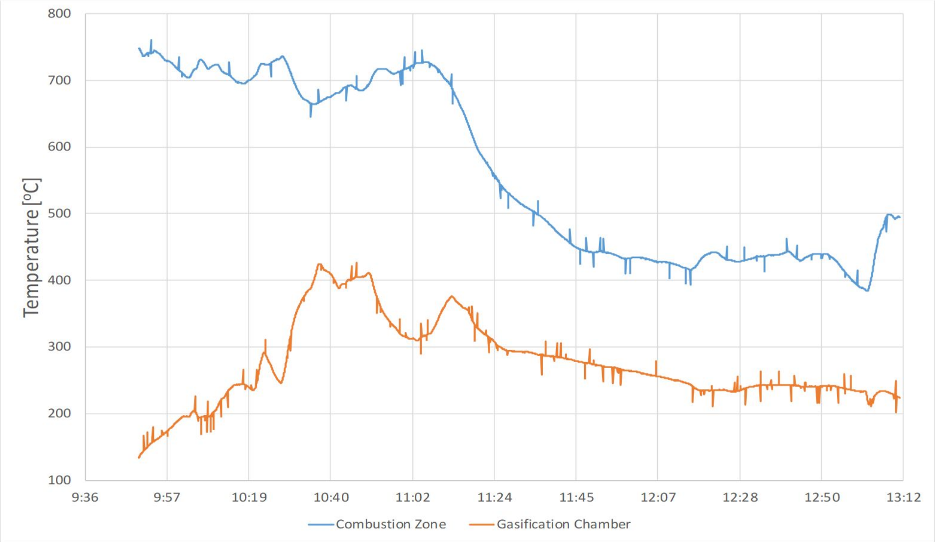

Fig. 7. Temperature fluctuations over time: in the combustion zone (nozzle) and in the gasification chamber for miscanthus briquettes

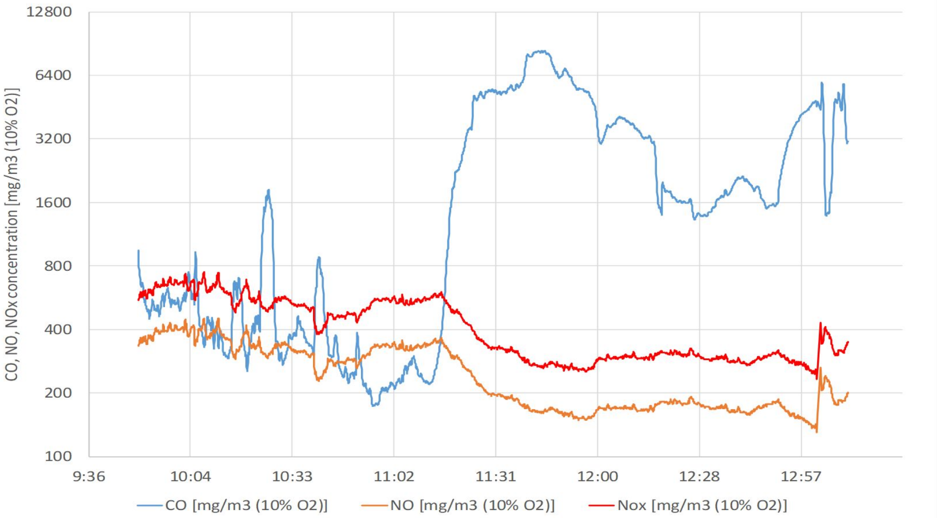

Fig. 8. Variation over time of carbon monoxide (CO), nitric oxide (NO) and nitrogen oxides (NO_X) concentrations for miscanthus briquettes

Figures 9 and 10 show the correlation between nitric oxide concentration and carbon monoxide concentration in the flue gas for selected biomass types and loads (hay and corn stover).

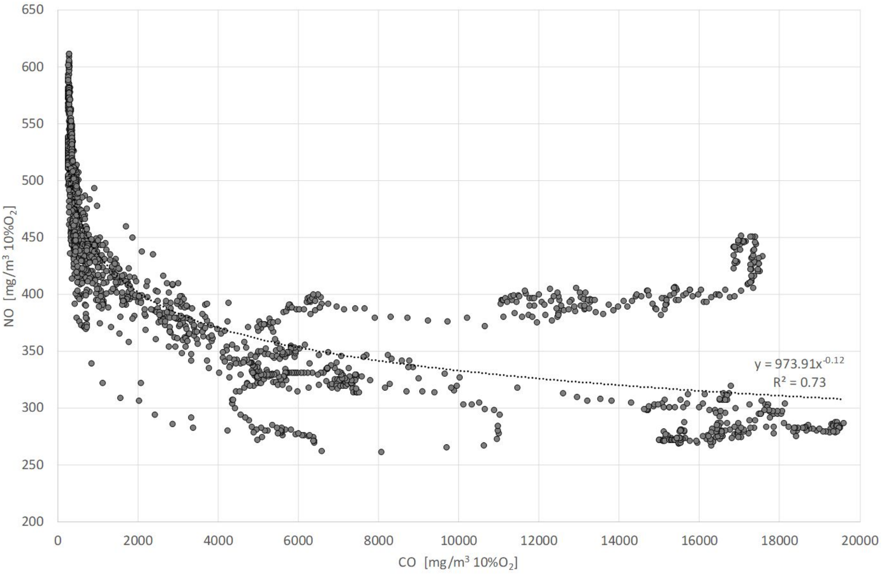

Fig. 9. Correlation between nitric oxide concentration and carbon monoxide concentration in the flue gas for straw briquettes

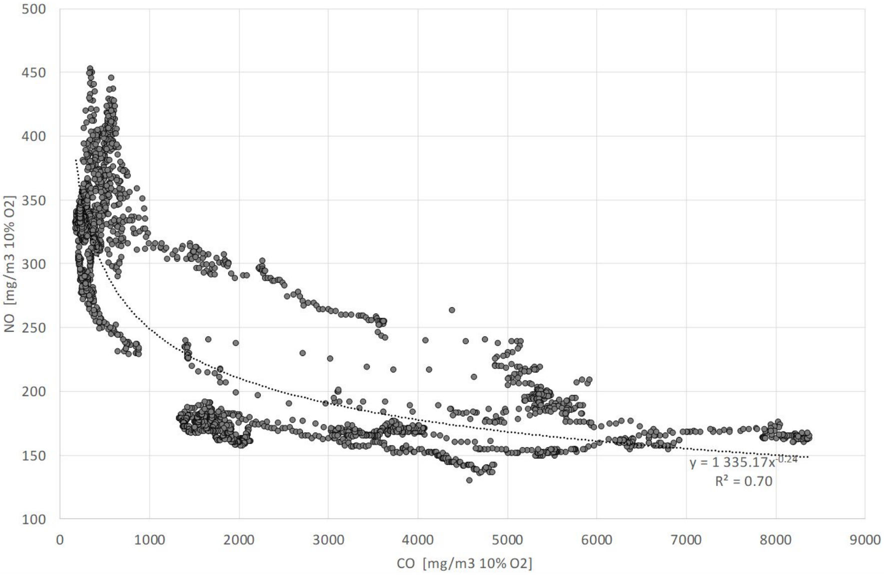

Fig. 10. Correlation between nitric oxide concentration and carbon monoxide concentration in the flue gas for miscanthus briquettes

Figures 11-14 show how carbon monoxide and nitric oxide concentration depend on oxygen concentration and temperature in the combustion zone for the analyzed fuel loads and selected briquette types (rye straw and miscanthus).

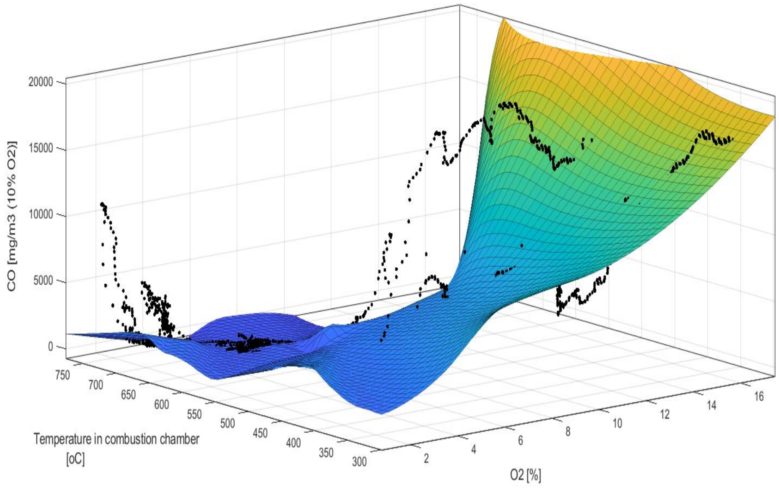

Fig. 11. Carbon monoxide concentration versus oxygen concentration and temperature in the combustion zone (nozzle) for rye straw briquettes

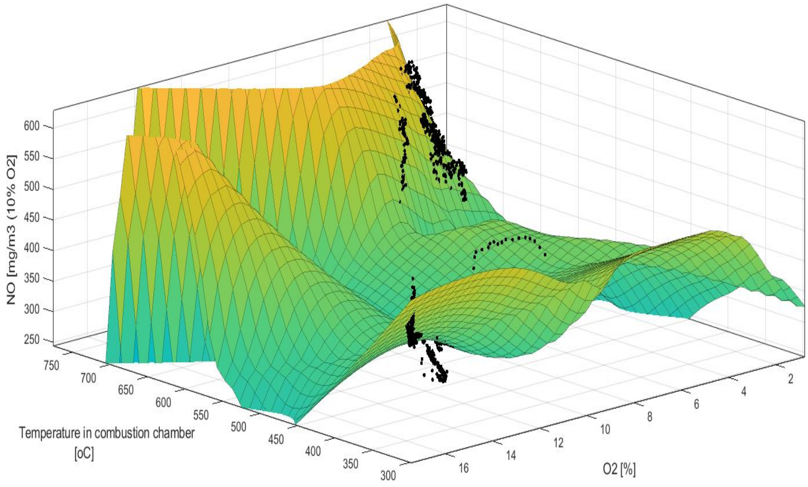

Fig. 12. Nitric oxide concentration versus oxygen concentration and temperature in the combustion zone (nozzle) for rye straw briquettes

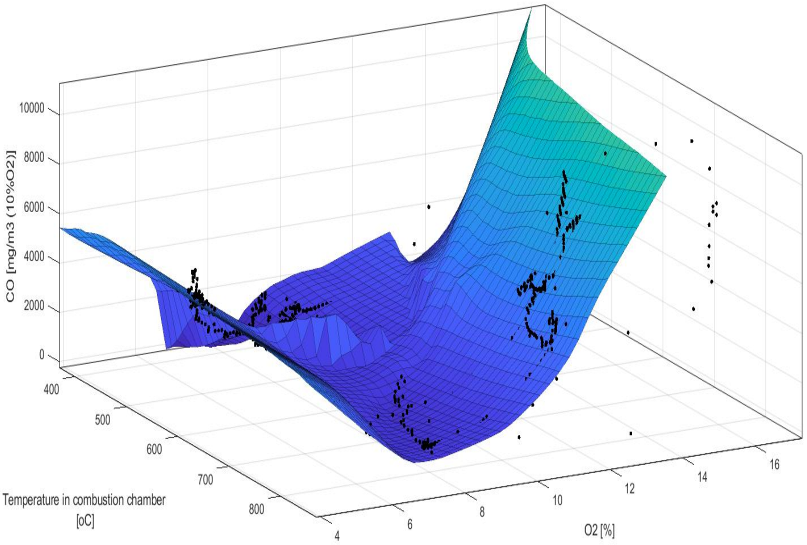

Fig. 13. Carbon monoxide concentration versus oxygen concentration and temperature in the combustion zone (nozzle) for miscanthus briquettes

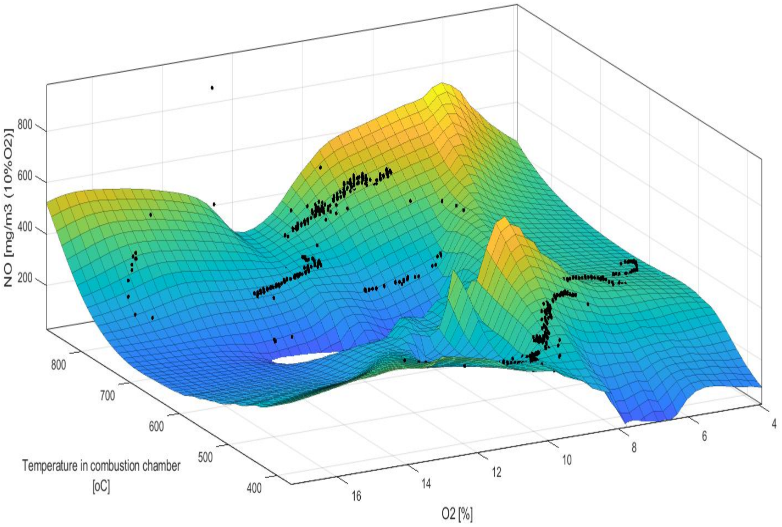

Fig. 14. Nitric oxide concentration versus oxygen concentration and temperature in the combustion zone (nozzle) for miscanthus briquettes

The correlations presented in the diagrams were similar for the other fuel loads and biomass types shown in table 2. Due to the large volume of the results, only the most significant correlations are presented.

As the temperature in the gasification chamber was maintained below 300°C, as expected, no slagging was observed during the process. In the range of temperatures obtained in the combustion zone (below 1000°C), only nitric oxide was generated, and no nitrogen dioxide. However, for evaluation of compliance with legal regulations (NO_x emission standards), the concentration of nitric oxide was expressed as NO_2, according to the formula $NO + ½ O_2 = NO_2$.

The boiler used is of old design and is not adapted for agricultural biomass firing, because no boiler with a heat output of approx. 20 kW designed for two-stage agricultural biomass combustion is available on the Polish market. The nozzle located at the bottom of the gasification chamber acts as a low-quality burner (see table 2), thus generating high carbon monoxide emissions. The nozzle also allows some biomass briquettes that have not been fully burnt (fig. 1) to pass through to the combustion zone, which significantly reduces the boiler's heat efficiency (table 2).

Another drawback is the lack of automated combustion air flow regulation, which by definition prevents the obtaining of optimum combustion conditions that would ensure low carbon monoxide concentrations (table 2). The limit value for carbon monoxide in the flue gas, which in this case is the criterion of combustion process quality, for boilers with a heat output of up to 50 kW and manual fuel supply, is currently set at 5000 mg/m^3 (presented for 10% O_2 content in the flue gas) [PN-EN-305-5: 2012]; however, in 2020 the European Union is to implement a regulation reducing this limit value to 700 mg/m^3 (presented for 10% O_2 content in the flue gas) [Commission Regulation (EU) 2015/1189: 2015].

The same EU regulation will establish a limit value for nitrogen oxides (NO_x) at 200 mg/m^3 (presented for 10% O_2 content in the flue gas). This is not addressed at all in the Polish standard currently in force for boilers with a heat output of up to 500 kW [PN-EN-305-5: 2012]. During the study, NO_x concentrations significantly exceeded this value, due to the considerable nitrogen content in agricultural biomass in comparison with wood (approximately 0.2%) [EN 14961-2: 2011; EN 14918: 2009]. This means that two-stage combustion of biomass will become difficult or even impossible in low heat output boilers, because in order to reduce the level of nitrogen oxides emissions it is necessary to use chemical methods. In practical terms, this means that two-stage combustion of agricultural biomass will only be cost-effective in boilers with a higher heat output, above 30 MW.

On the graphs of temperature over time (figs. 5, 7) and on the corresponding graphs presenting the variation of carbon monoxide concentration over time (figs. 6, 8), it can be clearly seen that carbon monoxide concentration increases as the temperature in the combustion chamber decreases, and vice versa.

As far as nitric oxide concentration is concerned, it generally increases with an increase in oxygen concentration and temperature in the combustion zone

[Rybak 2006]; however, there is a reciprocal relationship between carbon monoxide and nitric oxide (figs. 9 and 10), according to the formula $CO + NO \rightarrow \frac{1}{2} N_2 + CO_2$ [Marutzky 1997]. This formula shows that carbon monoxide lowers nitric oxide concentration by reducing nitric oxide (NO) to molecular nitrogen (N_2). At the same time, carbon monoxide oxidizes to carbon dioxide, and thus the CO concentration in the flue gas decreases. In figures 9 and 10, the correlation between nitric oxide concentration and carbon monoxide concentration can be clearly seen: when carbon monoxide concentration increases, nitric oxide concentration significantly decreases. This effect of carbon monoxide on nitric oxide is also reflected in diagrams illustrating how the concentrations of carbon monoxide and nitric oxide depend on the temperature and oxygen concentration in the combustion zone (figs. 11-14). These diagrams have distinctive and sometimes surprising shapes, precisely because of this particular mutual correlation.

Conclusions

The study has confirmed that two-stage combustion of agricultural biomass in boilers with a heat output below 50 kW is the most effective method for its firing, because of the fact that no slag is generated due to the low temperatures in the gasification chamber.

However, in order to ensure good combustion conditions and obtain acceptable values of carbon monoxide concentration, modern boilers must be equipped with high-quality gas burners and automated combustion air flow regulation systems. Unexpectedly high NO concentrations (calculated to NO_2 and presented as NO_x) were observed despite the low temperatures in the combustion chamber and high CO concentrations.

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EN 14961-2:2011 Solid biofuels. Fuel specification and classes. Part 2: wood pellets for non-industrial use

EN 14918:2009 Solid biofuels. Determination of calorific value

PN-EN 303-5:2012 Heating boilers. Part 5. Heating boilers for solid fuels, hand and automatically stocked, nominal heat output of up to 500 kW. Terminology, requirements, testing and marking

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