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The dependence of COx and NOx emission concentrations on the excess air coefficient during combustion of selected agricultural briquetted by-products

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Abstract. The issue of CO, CO_2 and NOx emissions is very extensive and important. The aim of the paper is the experimental determination of the CO, CO_2 and NOx emission dependencies on the amount of excess air. Materials used for the experiments were several types of briquetted biomass. Crops used were Czech knotweed (*Reynoutria* × *bohemica*), Rumex hybrid OK 2 (*Rumex patientia* × *Rumex tianschanicus*), meadow hay and timothy grass (*Phleum pratense*). For all samples proximate and elemental analyses were performed (semiautomatic calorimeter LECO AC-600 elemental analyser CHN628 + S and analyser LECO TGA-701) and stoichiometric calculations of combustion were made. Combustion device used in combustion tests was a hot air stove with a grate fireplace and with manual fuel supply. The combustion process was monitored by analyser Madur GA-60. Parameters monitored during the combustion tests were primarily the flue gas temperature and the emission levels of carbon monoxide, carbon dioxide and nitrogen oxides.

Analyses and calculations of plant biomass samples indicate their good properties for energy use. The gross calorific value was as high as 19.55 $MJ.kg^{-1}$ in the sample of Rumex OK 2. Limiting factor is the high quantity of ash in plant material. The briquettes from timothy grass achieved 5.77% wt. ash in the dry matter. The excess combustion air had positive influence during combustion test. On the other hand, this caused heat loss by departing flue gases, wherein the flue gas temperature reached high values. The excess air coefficient also significantly affected the emission levels of carbon dioxide and monoxide and nitrogen oxides in the flue gases. Results were statistically analysed and complemented by regression equations, which in practice can be used to optimize the combustion process in boilers with manual fuel supply.

Key words: plant biomass, combustion device, calorific value, combustion gases, heat loss.

INTRODUCTION

Impacts of combustion processes on the environment are evident. Results of Esteban et al. (2014) clearly demonstrate the environmental benefits of using small scale produced biomass instead of fossil fuels. As for the combustion process quality, the first indicators are the levels of carbon oxides (Bradna & Malaťák, 2016). If at low excess air the highest possible concentration of CO_2 is achieved, losses caused by the flue gases at that temperature are minimal (Johansson et al., 2004).

Higher emissions of sulphur and nitrogen oxides observed in plant biomass compared to wood biofuel were reported by Brassard et al. (2014). Also the influence of discontinuous fuel supply on carbon monoxide and nitrogen oxides does not necessarily lead to a significant increase in the concentration of carbon monoxide emissions as compared with combustion devices with continuous fuel supply (Juszczak, 2016). Diaz-Ramirez et al. (2014) confirmed the influence of fuel nitrogen and higher proportion of air on increasing nitrogen oxides (NOx) emission concentrations. Gaseous emissions are significantly influenced by the type of fuel. High CO emission may be caused by high solid loading and high length to diameter ratios in solid fuels, while high NOx emissions can be caused by a very high content of nitrogen in the fuel (Garcia-Maraver et al., 2014).

This article aims to provide experimental determination of carbon monoxide and nitrogen oxides emission concentrations, and also flue gas temperature in dependence on the excess air coefficient during combustion of briquettes from plant biomass in a grate combustion device. For these materials elemental analyses and stoichiometric calculations were determined.

MATERIALS AND METHODS

To fulfill the objectives of article several plant biomasses were sampled. They were Czech knotweed (*Reynoutria × bohemica*), Rumex OK 2 (*Rumex patientia × Rumex tianschanicus*), meadow hay and timothy grass (*Phleum pratense*). Czech knotweed at harvest reaches height of 3 m and thanks to its rapid growth it produces a large amount of biomass. This plant due to its characteristics is being considered as a renewable energy source (Strašil & Kára, 2010). Rumex OK 2 for energy purposes was harvested just before reaching full maturity when the seeds were not yet released. Harvested material has good characteristics as a biofuel and the net calorific value approaches that of wood chips (Hedĕnec et al., 2015). Meadow hay as an agricultural source consisting of various kinds of grasses, legumes and herbs (Riedener et al., 2015), was obtained from the first harvest (having coarser texture) in the temperate climate of the Czech Republic. Timothy grass is often grown as fodder in the dry areas of the Czech Republic. The significance of this plant for its energetic value is generally described in (Naik et al., 2010).

Each sample was disintegrated by a hammer mill and pressed into the form of briquettes. The briquetting press BrikStar 150 with a hydraulic unit was used for the samples processing. Density of the briquettes was up to 1,100 kg m^{-3} at the operating pressure up to 18 MPa and the operating temperature of 60 °C. Briquettes have a cylindrical shape with a diameter of 65 mm and length 50 mm.

Samples of biomass briquettes were analysed for fuel properties. The first task is to determine the chemical composition (by elemental analysis) on the combustible components such as carbon (C), hydrogen (H), nitrogen (N) and sulphur (S). The analyses were repeated six times and are carried out in an elemental analyzer LECO CHN628 + S. Detection method for carbon and hydrogen is dispersive infrared absorption, for nitrogen thermal conductivity and for sulphur infrared absorption. Accuracy range from 0.01 mg to 0.05 mg.

Non-combustible substances, i.e. ash and total moisture content, were determined in the thermogravimetric analyzer LECO TGA-701 with an accuracy of ± 0.02%. The gross calorific value of assessed biofuel samples was determined by burning in a isoperibolic calorimeter LECO AC-600 according to DIN 14918 (2010). The net calorific value was determined by calculation using the results of elemental and proximate analysis of individual samples (see Table 1).

Table 1. The average values from proximate and elemental analysis

Sample / Average values	Water Content (% wt.)	Ash (% wt.)	Gross Calorific Value (MJ.kg^{-1})	Net Calorific Value (MJ.kg^{-1})	Carbon C (% wt.)	Hydrogen H (% wt.)	Nitrogen N (% wt.)	Sulphur S (% wt.)	Oxygen O (% wt.)
	W	*A*	Q_s	Q_i	*C*	*H*	*N*	*S*	*O*
Czech knotweed original sample	**5.93**	**3.99**	17.62	**16.31**	45.87	5.33	0.30	0.03	**38.48**
Czech knotweed dry matter	**-**	**4.242**	18.73	**17.34**	48.76	5.66	0.32	0.03	**40.91**
Rumex OK 2 original sample	**7.75**	**2.75**	18.04	**16.86**	46.43	4.54	0.69	0.07	**37.72**
Rumex OK2 dry matter	**-**	**2.98**	19.55	**18.27**	50.33	4.92	0.75	0.07	**40.89**
Meadow hay original sample	**4.63**	**5.08**	17.25	**15.76**	42.86	6.34	0.44	0.08	**40.37**
Meadow hay dry matter	**-**	**5.33**	18.09	**16.52**	44.94	6.65	0.46	0.084	**42.33**
Timothy grass original sample	**10.65**	**5.16**	15.90	**14.38**	44.68	5.82	0.68	0.10	**32.54**
Timothy grass dry matter	**-**	**5.77**	17.79	**16.08**	50.01	6.51	0.76	0.11	**36.42**

The relationship between the gross calorific value Q_s (kJ kg^{-1}) and the net calorific value Q_i (kJ kg^{-1}) was expressed in the following equation:

$$Q_i = Q_s - (0.02442 \cdot 1{,}000) \cdot (W + 8.94 \cdot H) \quad (1)$$

where: W – the water content in test sample (*%*); *8.94* – coefficient for the conversion of hydrogen to water; H – hydrogen content in test sample (*%*); 0.02442 – value that corresponds to energy consumed in heating *1%* of water at 25 °C.

Stoichiometric calculations were made for calculation of other combustion characteristics, such as the excess air coefficient (*n*) expressed in the equation:

$$n = 1 + \left(\frac{CO_{2,max}}{CO_2} - 1\right) \cdot \frac{V_{sp.min}}{L_{min}} \quad (2)$$

where: $CO_{2,max}$ – theoretical volumetric concentration of carbon dioxide in dry flue gases (*%*); CO_2 – volumetric concentration of carbon dioxide in dry flue gases (*%*); $V_{sp.min}$ – theoretical mass amount of dry flue gas (m^3_N kg^{-1}); L_{min} – theoretical amount of air for complete combustion (m^3_N kg^{-1}).

The test device calculates the CO_2 content on the basis of measured values and characteristics of the fuel according to the formula:

$$CO_2 = CO_{2,max}\left(1 - \frac{O_{2,m}}{20.95}\right) \quad (3)$$

where: $CO_{2,max}$ – theoretical volumetric concentration of carbon dioxide in dry flue gases (%); $O_{2,m}$– real (measured) volumetric concentration of oxygen in dry flue gases (%); 20.95 – concentration of oxygen in atmosphere (%).

In stoichiometric calculations real molar volumes of gases. are used. Stoichiometric calculations are described in the paper of Malaťák et al. (2016). Results of stoichiometric calculations further serve to adjust the characteristics of the samples for flue gas analyzer and other essential combustion characteristics. Stoichiometric calculations are converted to standard conditions (to the temperature T = 0 °C and pressure p = 101.325 kPa).

Experimental measurements were carried out using a hot air grate combustion device with manual fuel supply from the company CALOR CZ. The nominal power of the combustion device is 12 kW and standard fuel consumption is 3.6 kg h^{-1}. The samples of biomass briquettes were monitored during sustained combustion. The emission concentrations were not monitored during ignition, start-up and extinguishing of fire.

Each sample was combusted for six hours. During the whole combustion process a nominal heat output was maintained which is rated for the combustion device at 12 kW with an efficiency of 80%. The corresponding hour supply of samples was for Czech knotweed $m_p = 3.31$ kg h^{-1}, Rumex OK 2 $m_p = 3.20$ kg h^{-1}, samples of meadow hay $m_p = 3.43$ kg h^{-1} and timothy grass $m_p = 3.76$ kg h^{-1}. For each fuel sample the measurement was carried out for 6 hours. The interval of fuel loading was set to 30 minutes. The measurements took place under stable combustion conditions during intervals of 15 minutes and the data was averaged for each measurement after one minute. During the whole combustion process the supply of primary combustion air is regulated, which is monitored by the amount of oxygen in the flue gas by the flue gas analyzer.

The emission concentration measurement was performed by flue gas analyser Madur GA-60. During all measurements the analyser monitored the ambient temperature, flue gas temperature and concentration of gases O_2, CO, NO, NO_2 in flue gas. Signal of converters is proportional to the volume concentration of the measured component in *ppm*. Recording interval of average individual components is set to one minute. Before each sample measurement was performed calibration of the measuring apparatus. The emission concentrations of dry flue gas are converted from *ppm* levels to normal conditions and transferred to the concentration in mg m^{-3} and reference oxygen content in the flue gas of 13%. Results of air emission measurements are processed by regression statistical analysis to express dependencies of carbon monoxide and carbon dioxide, flue gas temperature and nitrogen oxides on the excess air coefficient.

RESULTS AND DISCUSSION

The values from the elemental analysis indicated in Table 1 confirmed the fact from other authors Müller et al. (2015) and Ružbarský et al. (2014) that most decisive for the energetic utilization of selected biomass samples is the net calorific value, which depends on the water content and ash in the fuel. The best results in the net calorific value of dry matter on average reached samples of Rumex OK 2, which had compared to the other samples the lowest amount of ash.

A higher percentage of ash content is determined generally in samples of herbaceous biomass such as straw (Gurdil et al., 2009), in contrast to wood biomass (Johansson et al., 2004). Timothy grass had the highest concentration of ash up to 5.77% in the dry matter, which resulted in low gross calorific value. Low water content in the samples is a positive factor because moisture affects their behavior in combustion process and exhaust gas volume produced per unit of energy (Malaťák & Bradna, 2014). Another essential element in samples of plant biomass is the amount of oxygen which reduces the amount of oxidizable elements in the fuel. The average oxygen content of all samples was determined at 40.14% in the dry matter, which corresponds to the results presented in this work Tao et al. (2012).

The values from stoichiometric calculations (see Table. 2) did not reveal fundamental differences in the theoretical air consumption, the theoretical concentration of dry flue gases of individual samples. Larger differences were determined in the theoretical maximum concentration of carbon dioxide in flue gas in the samples of Rumex OK 2, where the values reached 21.30% vol. This fact should be reflected in the emission measurements.

Table 2. The results of stoichiometric calculations

Sample / Average values	Theoretical amount of air		Theoretical amount of dry flue gases		Theoretical concentration of carbon dioxide in dry flue gases	
	kg kg^{-1}	m^3 kg^{-1}	kg kg^{-1}	m^3 kg^{-1}	% wt.	% vol.
Czech knotweed original sample	5.45	4.20	7.80	4.13	21.56	20.59
Rumex OK 2 original sample	5.28	4.07	7.69	4.04	22.12	21.30
Meadow hay original sample	5.38	4.14	7.63	4.03	20.59	19.72
Timothy grass original sample	5.74	4.42	7.98	4.29	20.53	19.32

The graph of the emission measurements for Czech knotweed depending on the excess air coefficient is shown in Fig. 1.

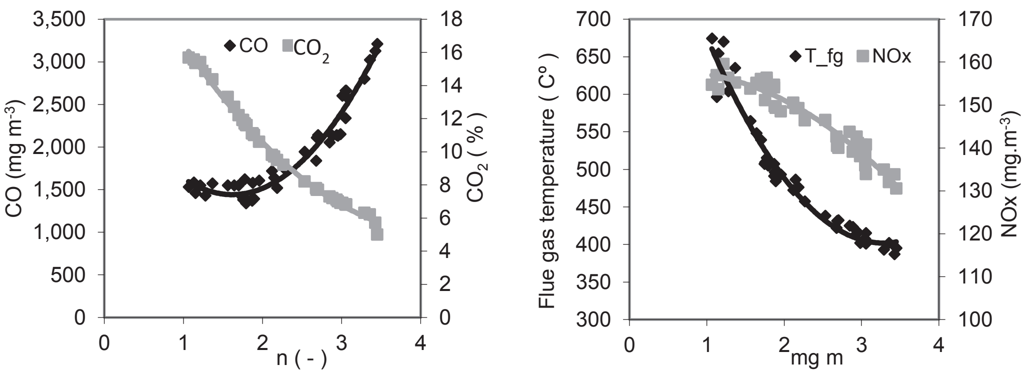

Figure 1. Graph of the emission measurements for Czech knotweed.

The combustion process quality during stable combustion is described by regression equations for Czech knotweed depending on the excess air coefficient n (-):

$$CO = 504.43n^2 - 1{,}631.3n + 2{,}760.8;\ R^2 = 0.9515 \quad (4)$$

$$CO_2 = 2.4138n^2 - 15.965n + 32.782;\ R^2 = 0.9869 \quad (5)$$

$$NOx = -3.1361n^2 + 3.3643n + 156.93;\ R^2 = 0.9368 \quad (6)$$

$$T_{\_fg} = 52.64n^2 - 345.54n + 968.92;\ R^2 = 0.9624 \quad (7)$$

where: CO – carbon monoxide emission concentration (mg m^{-3}); CO_2 – carbon dioxide emission concentration (%); NOx – nitrogen oxides emission concentration (mg m^{-3}); $T_{\_fg}$ – flue gas temperature (°C).

The graph of the emission measurements for Rumex OK 2 depending on the excess air coefficient is shown in Fig. 2.

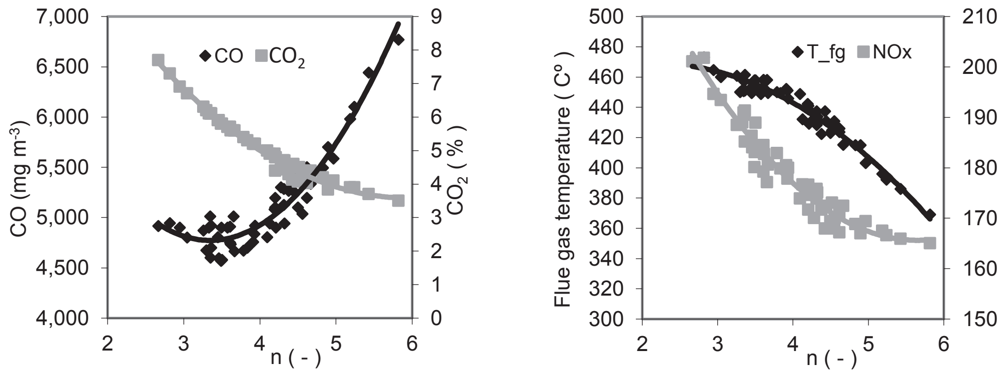

Figure 2. Graph of the emission measurements for Rumex OK 2.

The combustion process quality is described by regression equations for Rumex OK 2 depending on the excess air coefficient n (-):

$$CO = 352.36n^2 - 2{,}357.6n + 8{,}716.4;\ R^2 = 0.9124 \quad (8)$$

$$CO_2 = 0.3773n^2 - 4.482n + 16.897;\ R^2 = 0.9812 \quad (9)$$

$$NOx = 4.1513n^2 - 46.934n + 298.27;\ R^2 = 0.942 \quad (10)$$

$$T_{\_fg} = -7.6581n^2 + 32.954n + 433.61;\ R^2 = 0.9515 \quad (11)$$

where: CO – carbon monoxide emission concentration (mg m^{-3}); CO_2 – carbon dioxide emission concentration (%); NOx – nitrogen oxides emission concentration (mg m^{-3}); $T_{\_fg}$ – flue gas temperature (°C).

A graph of the emission measurements for Meadow hay depending on the excess air coefficient is shown in Fig. 3.

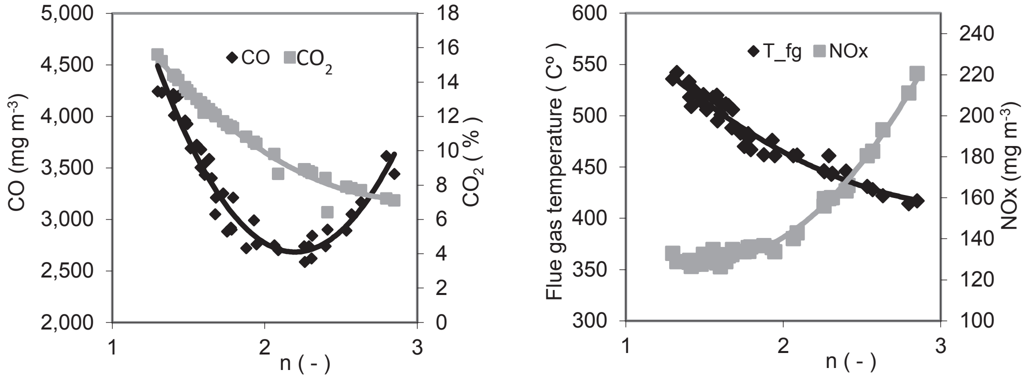

Figure 3. Graph of the emission measurements for Meadow hay.

The combustion process quality is described by regression equations for the Meadow hay depending on the excess air coefficient n (-):

$$CO = 2{,}231.2n^2 - 9{,}805.3n + 13{,}456;\ R^2 = 0.9376 \quad (12)$$

$$CO_2 = 2.9569n^2 - 17.529n + 33.14;\ R^2 = 0.9817 \quad (13)$$

$$NOn = 50.639n^2 - 154.81n + 247.79;\ R^2 = 0.9888 \quad (14)$$

$$T_{\_fg} = 31.81n^2 - 209.63n + 756.73;\ R^2 = 0.9456 \quad (15)$$

where: CO – carbon monoxide emission concentration (mg m^{-3}); CO_2 – carbon dioxide emission concentration (%); NOx – nitrogen oxides emission concentration (mg m^{-3}); $T_{\_fg}$ – flue gas temperature (°C).

A graph of the emission measurements for Timothy grass depending on the excess air coefficient is shown in Fig. 4.

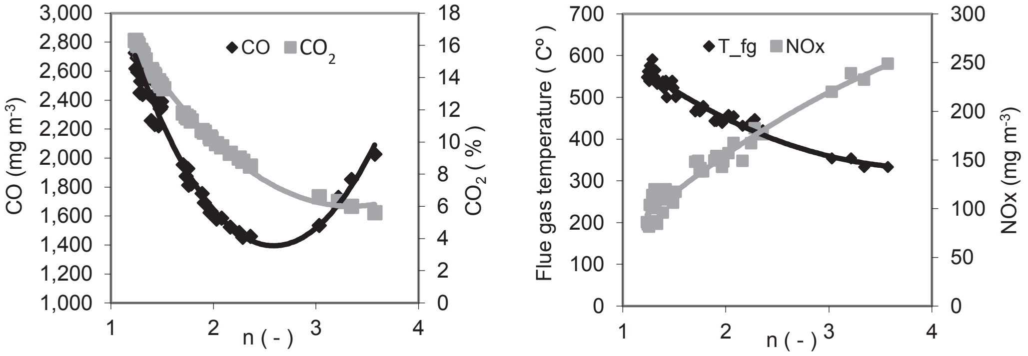

Figure 4. Graph of the emission measurements for Timothy grass.

The combustion process quality is described by regression equations for Timothy grass depending on the excess air coefficient *n* (-):

$$CO = 718.7n^2 - 3720n + 6{,}208.2;\ R^2 = 0.9595 \quad (16)$$

$$CO_2 = 2.2576n^2 - 15.093n + 31.233;\ R^2 = 0.9953 \quad (17)$$

$$NOn = -9.5214n^2 + 111.63n - 29.756;\ R^2 = 0.9528 \quad (18)$$

$$T_{\_fg} = 30.44n^2 - 242.2n + 812.06;\ R^2 = 0.9509 \quad (19)$$

where: CO – carbon monoxide emission concentration (mg m^{-3}); CO_2 – carbon dioxide emission concentration (%); NOx – nitrogen oxides emission concentration (mg m^{-3}); $T_{\_fg}$ – flue gas temperature (°C).

Under conditions of low excess air coefficient ($n < 2$) incomplete combustion occurs and high concentrations of carbon monoxide are found. Conversely with higher excess air coefficient ($n > 3$) rising concentration of carbon monoxide emissions occurs, mainly due to low temperature and premature cooling of the flue gas. These trends are observed in all examined samples. For samples of Rumex OK 2 particularly high excess air coefficients were tested as well and with *n* approaching 6 carbon monoxide in the flue gas got up to 6,500 mg m^{-3}. The increasing trend of carbon monoxide concentration in the flue gas is a result of the lowering temperature and high excess air coefficient in combustion zone, where the high CO concentration in flue gas is firstly resulted by cooling of the burning furnace. This fact is also confirmed in the combustion test in Malaťák & Bradna (2017) and also in Liu et al. (2010).

Similar trends for emission concentrations were achieved by Eskilson et al. (2004) during the combustion of pellets from plant biomass. Strehler (2000); Houshfar et al. (2012) & Černý, et al. (2016) achieved similar results during combustion of wood pellets. In their case decreasing amount of air in the combustion chamber reduced amount of nitrogen oxides in flue gas, but on the other hand increased emissions of unburned carbon.

All samples reach an optimal concentration of nitrogen oxides below 250 mg m^{-3}. For samples of meadow hay and timothy grass with rising excess air coefficient an increase of nitrogen oxides emission occurred. These trends were also observed by Diaz-Ramirez et al. (2014). The highest flue gas temperatures 680 °C were observed for Czech knotweed.

CONCLUSIONS

The results of experimental measurements confirm the fact, that the proximate and elemental composition affect not only the net calorific value, but also the behaviour during the combustion process. Especially high amount of oxygen and ash in plant biomass samples will affect the final emission concentrations. This difference is primarily visible in the samples of Czech knotweed and Timothy grass compared to other analysed samples. The higher consumption of combustion air and high amount of flue gas affect setting of the combustion device.

The results of emission measurements confirm also the fact, that the excess air coefficient is an important parameter for achieving optimum combustion of plant biomass of the selected samples. The main factors are the concentration of carbon monoxide and nitrogen oxides, but also the flue gas temperature. Especially with samples of Rumex OK 2 higher concentrations of carbon monoxide at higher excess air coefficient were achieved. Under optimal conditions, the achieved reduction of CO and NOx, with the greatest reduction represents case with excess air coefficient around 2. The results of measurements confirm the fact that the emissions of CO and NOx were very sensitive to the excess air ratio. By regulating an optimal combustion air amount for each type of biomass fuel heat losses can be prevented thereby increasing the efficiency of the combustion device.

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