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EVALUATION OF CO AND NO_x EMISSIONS IN REAL-LIFE OPERATING CONDITIONS OF HERBACEOUS BIOMASS BRIQUETTES COMBUSTION

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The issue of carbon monoxide and nitrogen oxides emissions into the atmosphere is very current. This article thus focuses on the assessment of elemental composition of selected herbal biomass species and emission concentrations during combustion in a commonly available grate combustion device for briquetted fuel. In tests, emission concentrations were evaluated in contrast to the oxygen concentration in flue gas and flue gas temperatures. Samples of camelina (*Camelina sativa*), giant miscanthus (*Miscanthus giganteus*), reed canary grass (*Phalaris arundinacea* L.) and sorghum (*Sorghum bicolor*) were used. Elementary and stoichiometric combustion analyses were conducted for these samples (LECO AC-600 semi-automatic calorimeter, CHN628 + S elemental analyser and LECO TGA-701 analyser). Analyses of C, H, N and S concentrations and calorific values showed that samples of briquetted herb biomass had insignificant differences and could be used for energy purposes without limitation. The limiting factor was the high amount of ash, which amounted to 6.59% of dry weight in reed canary grass briquette sample. Furthermore, a high percentage (1.91% wt.) of nitrogen was observed in reed canary grass briquettes. Such a high amount of nitrogen during combustion tests resulted in an increase in NO_x emission levels in flue gas of the *Phalaris arundinacea* L. sample, in which it reached the maximum concentration of 375.20 mg·m⁻³. The combustion tests showed that even under steady-state conditions, high concentrations of carbon monoxide could not be avoided for tested briquetted biofuels without active regulation of the combustion process.

Keywords: calorific value; elementary analysis; stoichiometry; coefficient of excess air; flue gas

Reduction of impacts of fossil fuel combustion includes their replacement by renewable energy sources. Particularly, it is wood or herbaceous biomass in the field of solid fuels (Oberberger et al., 2006; Dhaundiyal and Hanon, 2018). Especially for wood, further modifications are made to obtain more valuable materials, not only for energy purposes (Kučerová et al., 2016; Kučerová and Výbohová, 2018). Considering the herbaceous biomass, which does not offer as wide a possibility of treatment as wood, it is primarily direct combustion (Brunerová et al., 2017; Malaták et al., 2017). Biomass of plant origin contains a significant energy potential that varies with plant physiology (Hnilička et al., 2015). Impacts of the renewable energy fuel utilization lead to environmental improvements (Esteban et al., 2014).

When assessing the quality of the combustion process, the first indicators are the levels of carbon oxides in the flue gas (Bradna and Malaták, 2016). The maximum CO₂ concentrations are achieved at low coefficients of excess air (Johansson et al., 2004) together with the highest combustion temperature (Malaták et al., 2017). At the same time, other components of flue gas, such as sulphur and nitrogen oxides, must be taken into consideration. Their concentrations are always higher in herbaceous biomass than in wood (Brassard et al., 2014). Production of nitrogen oxides is dependent on combustion temperatures and the

amount of incoming combustion air (Díaz-Ramírez et al., 2014). Garcia-Maraver et al. (2014) confirmed the effects of higher nitrogen content in the fuel on increased emission concentrations.

Emissions production is influenced not only by the type of fuel (Zosima and Ochsenkiihn-Petropoulou, 2015) but also by the type of combustion technology (Malaták and Passian, 2011; Win et al., 2012). Above all, discontinuous fuel supply leads to increased concentrations of carbon monoxide and nitrogen oxides in comparison to continuous fuel supply (Juszczak, 2016).

In this paper, four herbaceous biomass fuels have been tested with a hypothesis that increasing the excess of combustion air would lead to a significant reduction in nitrogen oxide emissions caused by simultaneous decrease in flue gas temperatures. However, at the same time, carbon monoxide emissions have been expected to increase in volume. To test this hypothesis, samples were combusted under typical operating conditions of a grate fired combustion device while measuring the emission concentrations of nitrogen oxides and carbon monoxide; the flue gas temperature; and the coefficient of excess air. These measurements were supplemented by analyses of proximate and elemental compositions, as well as calorific values.

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Material and methods

Biomass materials

Four herbaceous biomass samples were selected. These were camelina straw (*Camelina sativa*, CS); giant miscanthus (*Miscanthus giganteus*, MG); reed canary grass (*Phalaris arundinacea* L., PA); and sorghum (*Sorghum bicolor*, SB).

CS, once grown over large areas in Europe, is an undemanding crop with a short growing season and no need for high-tech agricultural machinery. It grows well in most soils (Eynck and Falk, 2013). At 8% wt. moisture content, the net calorific value of straw is approx. 15.1 MJ·kg⁻¹. The sample came from a 2018 harvest.

MG is one of major energy crops. This plant, which requires favourable soil and climatic conditions, can show high yields of up to 28.19 t·ha⁻¹ of dry matter in the Czech Republic (Stražil, 1999). The harvest was carried out in 2018 in March and April.

PA is a plant native to the Czech Republic with an average yield of 10 t·ha⁻¹. Samples from the 2018 spring harvest were used. Spring harvest is recommended because the potassium, chlorine, nitrogen and sulphur contents tend to decrease during later harvests in contrast to early harvest dates (Platace et al., 2013).

SB is one of many energy crops grown in the Czech Republic. Usually, it is used as a substitute for maize (Amon et al., 2007). In the Czech Republic, the average yield is 16 t·ha⁻¹ of dry matter. Since SB is harvested only once a year, samples were collected in 2018.

Fuel analysis

Elemental analysis to determine the percentage of combustible matter components, such as carbon (C), hydrogen (H), nitrogen (N) and sulphur (S), were conducted nine times for each sample. A LECO CHN628 + S elemental analyser was used for this purpose. The detection method for C, H, S was infrared absorption; thermal conductivity cell was utilized to determine the N content.

LECO TGA-701 thermogravimetric analyser was used to determine the percentages of non-flammable substances, i.e. ash and total moisture content. Here, the measurements were repeated six times for each sample with an accuracy of ±0.02%. Another measured parameter was the gross calorific value. It was determined by combustion utilizing the isoperibol calorimeter LECO AC-600 in accordance with the DIN 14918 (2010). The net calorific value was specified via calculation utilizing the results of elemental and proximal analyses of individual samples. This relationship between gross calorific value Q_g (kJ·kg⁻¹) and net calorific value Q_i (kJ·kg⁻¹) is as follows:

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

where:

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

Stoichiometric combustion calculations add to the characterisation of the samples. This was mainly the determination of the theoretical amount of combustion air; theoretical amount of dry flue gas; and theoretical concentration of carbon dioxide in dry flue gases. The method of calculation has been described by Malaták et al. (2016). Stoichiometric calculations are necessary to compare the theoretical and actual combustion process. They exploit the results of the previous analyses. The results of the stoichiometric calculations were used to set the sample characteristics in the flue gas analyser, as well as to determine the excess air coefficient, which can be calculated as follows:

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

where:

- $CO_{2, \max}$ – theoretical volumetric concentration of carbon dioxide in dry flue gases (% vol.)
- CO_2 – volumetric concentration of carbon dioxide in dry flue gases (% vol.)
- $V_{sp, \min}$ – theoretical mass amount of dry flue gas (m³·kg⁻¹)
- $L_{\min}$ – theoretical amount of air for complete combustion (m³·kg⁻¹)

Combustion tests

Emission concentrations were measured using a combustion device with a fixed combustion grate with a manual fuel supply CALOR (WAMSLER, Ismaning, Germany). Similar devices are commonly used for household heating. This combustion plant was also selected for the ability to achieve a large range of operating conditions. The device nominal heat output is 12 kW and the standard fuel consumption is 3.5 kg·h⁻¹. During the combustion tests, mass flows of the samples into the combustion plant were recorded to maintain the rated thermal output. Throughout the combustion process, the nominal heat output was maintained for the combustion plant at 12 kW with an efficiency of 80%. The corresponding hourly input of briquetted samples was 3.45 kg·h⁻¹ for CS; 3.42 kg·h⁻¹ for MG; 3.54 kg·h⁻¹ for PA; and 3.62 kg·h⁻¹ for SB. For each fuel sample, combustion tests lasted for 6 hours and the fuel dosing interval was set to 30 minutes. The experimental data were acquired under stable combustion conditions at intervals of 15 minutes and each data point was averaged by the analyser values over one minute. Throughout the combustion process, the primary combustion air supply was controlled. This was monitored by the oxygen concentration in the flue gas.

The emission concentrations were measured using a Madur GA-60 flue gas analyser. During the measurements, the analyser monitored the ambient temperature; flue gas temperature; and concentrations of O₂, CO, NO and NO₂ in the flue gas. Before the measurement of each sample, the analyser was calibrated. The signals of the transducers were proportional to the volumetric concentration of the measured components in ppm. The concentrations of dry flue gas components were converted to normal conditions and concentrations in mg·m⁻³ at the reference oxygen

content of 13% in the flue gases. The results of emission measurements were processed utilizing the regression analysis in order to convey the dependence of carbon dioxide and nitrogen oxides on the coefficient of excess air and the flue gas temperature.

Results and discussion

The elemental analysis of all samples is shown in Table 1. The results are given both at the original moisture content and in a dry state. There were no statistically significant differences in the contents of combustible matter elements at significance level of 95% with the exception of N content in PA. In PA, the N at 1.91% wt. was higher in contrast to all other samples. Such a high amount of nitrogen can increase NO_x emission levels from combustion (Malaták and Bradna, 2017).

Percentages of non-flammable substances, such as ash and total moisture content shown in Table 1, indicate high contents of ash in the samples, especially in PA with 6.59% wt. of ash in dry matter. However, the ash contents are consistent with data on other herbaceous biomasses presented by other authors (Vassilev et al., 2010; Tao et al., 2012). Compared to wood biomass (Johansson et al., 2004), the ash content is generally higher in herbaceous biomass, especially in straws (Gürdil et al., 2009).

Very similar average gross calorific values were observed for CS and MG. The lowest average gross calorific value was

achieved for SB. This corresponds with the results of Vassilev et al. (2015).

The stoichiometric combustion calculations presented in Table 2 complement the fuel properties of the samples for evaluation of their combustion characteristics. Stoichiometric calculations use the average values for the original samples from Table 1. The results did not reveal any significant differences neither in the theoretical air consumption nor in the theoretical concentrations of the dry flue gas components between the materials. Larger differences were determined for CS in comparison to other samples. This material had the lowest combustion air consumption and the lowest dry flue gas production with the highest carbon dioxide content in the flue gas, the values of which reached 21.76% wt. – 21.37% vol.

The average emission concentrations are listed in Table 3. Statistically significant differences in average values in combustion tests were obtained between SB and MG briquettes. Combustion of SB achieved the highest average carbon monoxide emissions. When combusting the PA, the average emission concentrations of nitrogen oxides were at least twice as large as other briquette samples.

In all briquette samples, the emission concentration dependencies were similar as shown in Figs. 1–4. Emission concentrations of carbon monoxide increased with an increase in excess air ratio. The reason was lowering temperatures and premature cooling of combustion gases in the combustion chamber, as confirmed by Liu et al. (2010).

Table 1 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
<i>Camelina sativa</i> original sample	7.81	4.89	16.82	15.63	43.28	4.58	0.81	0.12	38.51
<i>Camelina sativa</i> dry matter	–	5.30	18.24	17.16	46.95	4.96	0.88	0.13	41.78
<i>Miscanthus giganteus</i> original sample	7.15	4.48	17.11	15.78	43.56	5.30	0.86	0.09	38.56
<i>Miscanthus giganteus</i> dry matter	–	4.82	18.43	17.18	46.91	5.71	0.93	0.10	41.53
<i>Phalaris arundinacea</i> L. original sample	6.29	6.18	16.59	15.25	42.11	5.45	1.79	0.10	38.08
<i>Phalaris arundinacea</i> L. dry matter	–	6.59	17.71	16.44	44.94	5.81	1.91	0.11	40.64
<i>Sorghum bicolor</i> original sample	6.52	5.72	16.21	14.90	42.58	5.28	0.58	0.11	39.21
<i>Sorghum bicolor</i> dry matter	–	6.12	17.34	16.11	45.55	5.65	0.62	0.12	41.94

Table 2 Results of stoichiometric combustion 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.
<i>Camelina sativa</i> original sample	4.90	3.77	7.29	3.76	21.76	21.37
<i>Miscanthus giganteus</i> original sample	5.18	3.99	7.51	3.93	21.26	20.57
<i>Phalaris arundinacea</i> L. original sample	5.08	3.91	7.40	3.85	20.87	20.28
<i>Sorghum bicolor</i> original sample	5.03	3.87	7.36	3.82	21.20	20.68

Table 3 Average values of flue gas temperature and the concentrations of components in dry flue gas in combustion tests

	Flue gas temp.	O ₂	CO ₂	CO (O _{2r} = 13%)	NO _x (O _{2r} = 13%)
	°C	% vol.	% vol.	mg·m ⁻³	mg·m ⁻³
<i>Camelina sativa</i> – original sample					
Average	558.45	10.50	7.49	2,508.91	101.21
Standard deviation	52.85	2.31	2.19	773.31	34.42
Max.	683.00	13.81	14.10	4,107.00	187.70
Min.	472.80	5.50	5.20	1,133.00	55.80
<i>Miscanthus giganteus</i> – original sample					
Average	587.90	10.59	6.99	1,848.99	75.55
Standard deviation	57.14	1.70	1.99	1,193.45	14.65
Max.	705.00	13.08	12.00	4,572.00	111.32
Min.	489.80	6.29	4.10	343.00	55.33
<i>Phalaris arundinacea</i> L. – original sample					
Average	489.83	10.24	7.92	1,773.26	227.40
Standard deviation	0.10	0.17	0.18	0.33	0.20
Max.	655.00	13.33	15.20	2,757.00	375.20
Min.	423.90	5.48	6.20	442.00	167.20
<i>Sorghum bicolor</i> – original sample					
Average	499.69	12.41	4.85	2,680.05	91.25
Standard deviation	34.81	1.38	1.62	820.08	19.95
Max.	608.00	13.84	9.80	4,132.00	143.50
Min.	460.80	8.17	3.20	978.00	57.29

Fig. 1 Emission concentration dependencies for *Camelina sativa*, values were converted to normal gas conditions and oxygen content 13% vol. in the flue gas

Emission concentrations of nitrogen oxides decreased with an increment in the excess air ratio. Similar trends for the observed emission concentrations were shown by Eskilsson et al. (2004) during combustion of pellets made of herbaceous biomass. Similar results were achieved by Strehler (2000); Houshfar et al. (2012); Černý et al. (2016) for fire wood pellets. In their case, the decreasing amount of combustion air reduced the amount of nitrogen oxides in the flue gas, but on the other hand, it also increased the unburnt carbon emissions. Since the emission concentrations are influenced by both the excess air coefficient and the flue gas temperature, these two parameters are plotted against each other in Fig. 5. It is clear from the graph that decreasing the value of the excess air coefficient increases the flue gas temperatures. Therefore, at the highest combustion temperature reached, the lowest value of the excess air coefficient was determined and vice versa. The high exhaust temperatures

Fig. 2 Emission concentration dependencies for *Miscanthus giganteus*, values were converted to normal gas conditions and oxygen content 13% vol. in the flue gas

Fig. 3 Emission concentration dependencies for *Phalaris arundinacea* L., values were converted to normal gas conditions and oxygen content 13% vol. in the flue gas

Fig. 4 Emission concentration dependencies for *Sorghum bicolor*, values were converted to normal gas conditions and oxygen content 13% vol. in the flue gas

during combustion tests were also achieved thanks to the high calorific values of the materials.

High concentrations of nitrogen oxides (375.20 mg·m⁻³) were found in combustion of PA at low excess air $n = 1.39$ and flue gas temperature 655 °C. This concentration exceeds the other materials several times over. The reasons for these high emissions are both high nitrogen content in the fuel, 1.79% wt., and high combustion temperature (Houshfar et al., 2012; Díaz-Ramírez et al., 2014; Kažimírová and Opáth, 2016). On the other hand, PA achieved the lowest carbon monoxide emission concentrations at low exhaust gas temperatures CO = (2,757.00 mg·m⁻³), whereas the other samples showed high emission concentrations above 4,000 mg·m⁻³. The emission measurements were processed by a regression statistical analysis to express the dependencies of carbon dioxide and nitrogen oxides on the coefficient of excess air and flue gas temperature. Figs. 1–4 show the graphs together with the regression equations. The correlation coefficient was always above 0.92 and mostly above 0.95.

Conclusion

This article is a response to stricter emission limits for combustion of briquette biomass in household combustion devices. The compositional, calorimetric and stoichiometric analyses of samples of herbaceous biomass briquettes characterise this type of fuel in many areas of its final use. Most important is their energy value, which is expressed in terms of the net calorific value and determines the thermal output of the combustion plant, as well as the production of gaseous emissions. Increasing the value of the excess air coefficient for combustion of briquettes resulted in a significant reduction in the emission levels of nitrogen oxides, as the flue gas temperature was reduced; however, carbon monoxide emissions were increased at the same time. It can be said that this general rule applies fully to both woody and herbaceous biomass samples despite the fact that elemental composition of pure wood differs mainly in terms of calorific value and ash content. The work also showed

Fig. 5 Flue gas temperature dependency on the excess air coefficient for all materials

the influence of increased fuel nitrogen on the total emission concentration of nitrogen oxides in samples of *Phalaris arundinacea* L.

The results of the measurement of the emission concentrations confirm that the excess air coefficient is an important parameter for achieving the optimal combustion. The main factors include carbon monoxide and nitrogen oxides emissions, as well as the flue gas temperature, which is dependent on the coefficient of excess air. Particularly for *Miscanthus giganteus* briquettes, the highest concentrations of carbon monoxide were determined at the maximum excess air coefficient reached. Therefore, it makes sense even during combustion of these briquettes to regulate the combustion process using a lambda probe, which would guarantee the combustion under optimal conditions. Considering the combustion in the areas of optimal excess air coefficient, the CO and NO_x emission levels are produced with the lowest impact on the environment. By regulating the optimal amount of combustion air for this type of briquette biomass, heat losses can be avoided, thereby increasing the efficiency of briquette-fired combustion units.

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