Energetic use of solid products of pyrolysis technology

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Abstract: At the present time there is a lot of discussion amongst scientists, concerning substitution of non-renewable energy resources by renewable. Our paper addresses this issue with reference to the current dramatic increase in the consumption of fuels, which are produced from renewable energy resources. The aim of this paper is to describe the possibilities and the limitations of the use of solid biomass modified by pyrolysis technology into biochar for energetic purposes.

Analyzed samples have been processed into the form of pellets of 8 mm in diameter, made from rape straw. A laboratory pyrolysis unit was used, working on the principle of slow pyrolysis reaction, processing samples at a temperature of 400 ℃ during 220 minutes in a reactor filled by nitrogen medium. The experimental part of the work was based on elemental and stoichiometric analysis. The thermal-emission properties were determined on the combustion furnace device with lower fuel supply for the samples analyzed before and after the pyrolysis process. The resulting values were evaluated by statistical regression analysis.

Experiments have shown that concentration of sulphur, nitrogen and chlorine in the analysed biofuels samples, did not vary significantly after the pyrolysis processing. The advantage of pyrolysis processing is the fact that the calorific value of examined samples almost doubled (26.22 MJ/kg). The pyrolysis fuel processing of selected samples significantly reduces the proportion of volatile matter and oxygen. For energy use of these processed samples it is necessary to pay attention to these indicators. The results of biofuel stoichiometric properties and the resulting values of thermal-emission concentrations (Tflue-gas, CO, CO2 a NOx) depending on excess air coefficient show the final energy use of pyrolysis solid samples which differs in their properties significantly from the original samples. For energy use of assessed samples is limiting value was the excess air coefficient which significantly influenced the course of the experimental tests.

Keywords: Combustion, biofuels, elemental analysis, pyrolysis, stoichiometric analysis

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1 Introduction

Solid biomass fuel used in small combustion plants is a key issue in current scientific research. First of all the quality of biomass fuel is becoming increasingly important (Olsson et al., 2003; Johansson et al., 2004). Therefore, research of these problems is gradually becoming one of the crucial elements of national energy policies. The European Union aims to significantly increase of funding for energy research within the built ERA (European Research Area) and inclusion of energetic issues among the leading strategic areas. Research in energy production from solid biomass is developing direct methods, combustion processes or processing to more valuable and richer raw materials such as fuel gas, fuel oil or highly valuable products for the chemical industry (Young and Hauserman, 1995; McKendry, 2002).

With the imminent depletion of most of the non-renewable energy resources, questions arise

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concerning their possible substitution by renewables. Pyrolysis processing of biomass (Rensfelt and Östman, 2000) is one of the most interesting options. The subsequent pyrolysis by-products are mostly gases, liquids and semi-coke. The composition of these products depends on the pyrolysis processing method and the reaction parameters (Bridgwater and Grossi, 1991). Fast or ultra-fast pyrolysis is used to maximize the production of gas and liquid products, mainly providing thermal benefit. Slow pyrolysis is known as carbonization, and is used mainly for the production of charcoal (Bridgwater and Peacocke, 2000; Lu et al., 2009). Currently research focuses on the selection of process conditions for maximum production of the bio-oil, which can be used without transformation in stationary devices for production of heat and power applications, or may be further be modified in to the drop-in biofuels (Czernik and Bridgwater, 2004; Balat et al, 2009.; Bridgwater, 2012).

One of the neglected paths of research in biomass reprocessing methods for solid products is torrefaction. Torrefaction is a method of biomass thermal conversion in the low temperature range of 200°C -300°C without presence of oxygen (Stelt et al., 2011). Biomass is thus processes in to a high-quality solid biofuel that can be used for combustion and gasification (Ibrahim et al. 2013). The torrefaction technology consists in removing oxygen from biomass. The resultant fuel has higher energy density by reactive decomposition of hemicellulose fractions (Kim et al., 2012). Reaction conditions (temperature, inert gas, reaction time) and biomass sources leads to the production of a variety of solid, liquid and gaseous substances (Stelt et al., 2011).

Experiments were performed recently to evaluate and compare the quality of biochars (Zhang et al., 2013), with set out parameters of particle size distribution, elemental analysis, etc. Successful implementation of these technologies into practice implies, that the substances produced by the pyrolysis process must have a higher usability and must be significantly more valuable than the untreated raw biomass (Lin et al., 1999). Current research aims to identify the scope and the values of parameters that classified biochars according to their quality.

The authors performed many scientific experiments in order to identify the essential fuel and thermal-emission properties of pellet samples from rape straw and pellet samples from rape straw processed by slow pyrolysis. The aim is to find other possibilities and limit values for energy use of solid biomass processed by pyrolysis technology to semi-coke. For further assessment the suitability of solid biofuels combustion in determined type of combustion device, or in assessing the quality of biofuels, it is necessary to identify the properties and specific characteristics of burned samples, on the basis of elemental, stoichiometric and thermal-emission analysis of selected solid biofuel samples.

2 Materials and methods

In order to meet our objectives, i.e. demonstrate optimal energy use of solid products from pyrolysis technology, we have collected in 2013 straw samples of waste rape from the experimental fields at the Czech University of Life Sciences Prague. Rape straw was then crushed in a laboratory mill and granulated into pellets using a granulating press with flat matrix, with a cross cut of eight millimetres .

A laboratory pyrolysis unit was used for rape straw pellets samples processing, operating on the principle of slow pyrolysis reaction. A prerequisite for the use of the pyrolysis process is maximum solid component production – semi-coke. For carbonization of the samples we used container test line TKKA ZLK (PolyComp). This pyrolysis unit consists of a reactor, where, after the insertion of fuel samples, atmospheric air is replaced with nitrogen gas. After heating to a temperature of 400°C and the residence time of 220 minutes, is solid semi-coke is removed from the pyrolysis unit.

An elemental analysis of fuel properties and evaluation of examined samples is the carried out. This is essential for every calculation of any combustion device thermal work. The individual weight percentages of carbon, hydrogen, oxygen, sulphur, nitrogen and all water in samples were determined. Non-combustible substances of fuels, i.e. ash content and all water are determined by incineration, respectively dewatering the sample.

Elemental analyses were performed at The Institute of Chemical Technology Prague, Faculty of Environmental Technology within the internal grants of authors. The elements carbon, hydrogen and nitrogen were determined on the analyzer CHN Elementar vario EL III (Elementar) according to ČSN ISO 29541 (2012). Accuracy of the method is determined by the manufacturer for the simultaneous determination 5 mg of standard 4-amino-benzen sulphanilic acid in the module CHNS < 0.1% abs. for each component. For chlorine and sulphur determination, samples were burned in an oxygen-hydrogen flame on the Wickbold Combustion Apparatus according to ČSN EN 15289 (2011). A certified moisture analyser Ohaus MB 25 was used to determine the total water content according to ČSN EN 14774-1 (2010). Higher heating value of examined samples was determined by measuring in the calorimeter IKA 2000 according to ČSN EN 14918 (2010). Lower heating value was determined by calculation according to ČSN EN 14918 (2010) and the results of the samples elemental analyses were used.

The following stoichiometric analysis of combustion processes complements the sample characteristics, and it is the basis for any thermal calculation. It is particularly important for many problems in design practice, as well as for overseeing the existing combustion apparatus. The amount of oxygen (air) required for complete combustion of fuel, the quantity and composition of fuel and the specific density of exhaust gas are determined in these calculations (Friberg and Blasiak, 2002; Malatak and Passian, 2011). For our stoichiometric analysis we consider the complete combustion and the reference quantity of oxygen in the combustion gases. Calculation of the air consumption and the amount of flue gas were determined in this work by analytical manner, i.e. according to elemental analysis using stoichiometric equations. All volumes and weights of combustion air and flue gases are given for the normal conditions, i.e., at t = 0 ℃ and pressure p = 101.325 kPa, and the reference oxygen content in the flue gases O_r = 11 %.

As a part of the research project, pellet samples made from rape straw were experimentally burned on combustion device with automatic fuel supply to lower retort intake (Lycka-AM 24 Licotherm) with a rated output of 24 kW heat application. The chosen samples before and after pyrolysis processing to biochar, were then

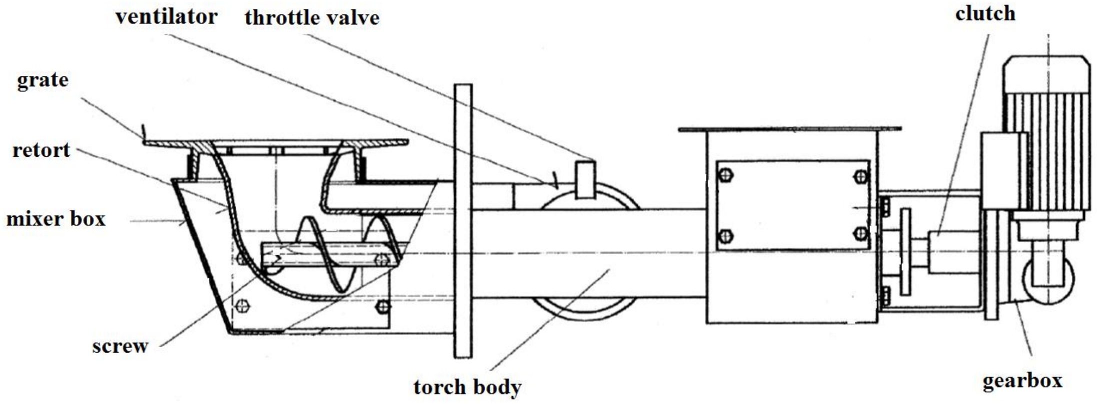

Figure 1 Combustion retort device with lower fuel supply

burned. The main task of this part was to set out the thermal emission concentrations of CO, CO_2, NO_x and flue gas temperature, depending on the excess air coefficient.

To determine the mass flow rates, emission factors and characteristics of solid particles during biomass pellets, we used the measuring unit GA – 60. This is a multi-purpose flue gas analyzer. This analyzer is equipped by transmitters for analysis of flue gas components: Oxygen (O_2), Carbon Monoxide (CO), Nitrogen Monoxide (NO), Nitrogen Dioxide (NO_2), Sulphur Dioxide (SO_2) and Chloride (Cl). The device GA - 60 allows measuring both ambient temperature (flow) and flue gas temperature ($T_{flue\text{-}gas}$). It is assessed fuel and combustion device through these measurements with regard to thermal parameters and emission rates.

Operational tests were carried out according to ČSN EN 070245 "*Warm water and low-pressure steam boilers. Warm water boilers with output up to 50 kW. Technical requirements*" (1993). The chimney draft values depending on the nominal thermal output fluctuated within a prescribed period of 12 ± 2 Pa (the static pressure in the measuring section of the flue gas) because the heating system was tested with lockable fireplace. The average concentrations of carbon monoxide and other gaseous emissions were converted to 11% oxygen content in dry flue gases during all measurements.

All measured values were statistically evaluated by statistical regression analysis with the expression of the regression equation and the value of reliability and are converted to normal conditions.

3 Results and discussion

The resulting values of the rape straw samples elemental analysis are given in Table 1. Stoichiometric analysis of volume combustion under normal conditions and reference oxygen content in the flue gas O_r = 11% are shown in Table 2.

Based on the results of elemental and stoichiometric analysis of modified biofuel samples by pyrolysis technology, we can say, that their changed properties affect their overall use (Malatak and Kucera, 2013).

Table 1 Resulting average elemental analysis of biofuel samples

Sample	Water content , w %	Ashes, w %	Combustible matter volatile , w %	Combustible matter non-volatile , w %	Gross calorific value , MJ/kg	Net calorific value, MJ/kg	Carbon C , w %	Hydrogen H, w %	Nitrogen N, w %	Sulphur S, w %	Oxygen O, w %	Chlorine Cl, w %
Rape straw pellets (Ø 8 mm)	5.45	4.83	72.28	17.44	17.36	16.15	43.70	4.90	0.72	0.23	40.17	0.23
Biochar: rape straw pellets (Ø 8 mm)	4.88	13.3	22.72	59.13	26.88	26.22	68.38	2.48	1.36	0.44	9.19	0.37

Table 2 Stoichiometric analysis of biofuel samples volume combustion under normal conditions and reference oxygen content in the flue gas $O_r = 11\%$

Sample	Theoretical amount of oxygen for complete combustion, m^3/kg	Theoretical amount of air for complete combustion, m^3/kg	Real amount of air content for perfect combustion , m^3/kg	Real mass amount of dry flue gas, m^3/kg	Theoretical mass amount of dry flue gas, m^3/kg	Volume amount of CO_2 , m^3/kg	Volume amount of SO_2, m^3/kg	Volume amount of H_2O, m^3/kg	Volume amount of N_2, m^3/kg	Volume amount of O_2, m^3/kg	Theoretical concentration of carbon dioxide in dry flue gases, vol %
Rape straw pellets (Ø 8 mm)	0.81	3.84	8.07	8.08	3.82	0.81	0.0	0.94	6.31	0.89	21.22
Biochar: rape straw pellets (Ø 8 mm)	1.35	6.43	13.51	13.44	6.30	1.27	0.0	0.88	10.6	1.49	20.11

One of the criteria for the use of these fuels is the content of nitrogen, because high content of this element restricts the use of these biofuels within combustion devices. We can say that the pyrolysis process did not affect the overall change of nitrogen concentration in the fuel.

One of the limiting elements in combustion of biomass is the production of chlorine. During combustion, chlorine is transformed in large part in to gas form. In our biofuel samples analysis, chlorine was found in its original state. Other research work (Johansson et al., 2003) addresses the issue of chlorine concentration that varies at a high level in rape straw samples compared to wood. The importance of chlorine is based on the one hand on HCl emissions, in their possible effect on the formation of polychlorinated dibenzo/dioxins and furans (PCDD/F) and on the other hand on the corrosive effects of these elements or their other compounds (Launhardt et al., 1998).

Most of the sulphur is also transformed by combustion into the gas phase as SO_2 or SO_3. Emissions of sulphur in heating devices, with regard to the limit values, using solid fuels from renewable resources, do not constitute usually a problem (Kjällstrand and Olsson, 2004). This was not confirmed in our rape straw samples. Increased sulphur concentration in the samples may therefore be caused by contamination of the locality by sulphur oxides emissions. The corrosive behavior of sulphur is the most decisive factor for choosing biofuel from renewable resources and depends on sulphur concentration in biofuels. Other values of elemental analysis meet the optimal parameters for the energy use of biofuel samples.

Based on these analyzes, it can be concluded that the samples from rape straw pellets will significantly burden combustion device spaces during combustion process. Use of biofuels based on rape straw is thus not appropriate for municipal heating facilities.

The most decisive for the thermal utilization of solid fuels is the calorific value. It depends on the water and ash content in the fuel (Ruzbarsky, 2014). The great advantage of the pyrolysis processing is the fact that calorific value of these samples almost doubled in particular by reducing oxygen content in the fuel. Reduction of oxygen has a positive effect on the increase in total energy of the resulting pyrolysis samples. Oxygen is undesirable part of the fuel because it binds hydrogen and partly carbon on hydroxides, oxides and water (Malatak and Kucera, 2013). On the other hand, the proportion of volatile matter in this pyrolysis processed samples from rape straw pellets is significantly reduced. There is generally known that fuels with a high volatile content have long burning flame (Fiedler and Persson, 2009).

The processed samples do not yet have classical properties of fuels from biomass, but they do have similar properties as fossil fuels. One positive aspect is that the amount of water contained in the samples is low, because humidity affects behaviour during combustion and exhaust gas volume produced per unit of energy (Eskilsson et al., 2004). The ash content in the original samples is high compared to wood biomass up to five times quantity, and the amount of ash after the pyrolysis process increased to three times compared to original samples. Such large amount of ash significantly affects the thermal properties of assessed solid fuels and consequently affects both selection and adjustment of combustion equipment.

The resulting values of the stoichiometric analysis indicate a difference in thermal emission parameters of assessed pellet samples. From the results of the stoichiometric analysis is evident difference in consumption of oxygen and air and flue gas production for individual biofuel samples before and after pyrolysis processing that significantly affect the operation of power equipment.

The starting point is to determine the optimum excess air coefficient as shown by the average value of the thermal emission analysis (Table 3). This optimum guarantees such combustion conditions, where there is no occurrence of high emission concentrations, but also prevents heat loss (Johansson et al., 2003).

Similar research works written by international authors, e.g. Solantausta et al. (1995), point to the fact that changes of fuel combustion properties occurs after pyrolysis processing. Pyrolysis improves the energy utilization of biofuels. However, different combustion characteristics and also the right choice of suitable combustion devices must be taken in to account.

Table 3 Resulting average parameters from measurements of thermal-emission parameters on the combustion device (dry flue gas)

Rape straw pellets (Ø 8 mm)		Average	SD	Maximum	Minimum
$T_{flue\text{-}gas}$	℃	184.78	15.88	218.00	156.00
O_2	%	16.53	0.75	18.20	15.27
n (excess air coefficient)		4.83	0.86	7.50	3.66
CO_2	%	3.25	0.55	4.18	2.02
CO (O_2=11%)	mg/m^3	6015.50	1825.25	12082.14	3405.90
NO (O_2=11%)	mg/m^3	398.31	69.47	529.04	255.19
SO_2 (O_2=11%)	mg/m^3	78.95	80.60	276.21	0.00
NO_x (O_2=11%)	mg/m^3	610.68	106.50	811.10	391.25
Biochar: rape straw pellets (Ø 8 mm)		Average	SD	Maximum	Minimum
$T_{flue\text{-}gas}$	℃	166.68	24.41	213.00	111.00
O_2	%	16.87	2.54	20.95	12.34
n (excess air coefficient)		15.94	58.63	420.00	2.42
CO_2	%	3.09	1.54	6.33	1.26
CO (O_2=11%)	mg/m^3	10046.50	2716.11	195069.64	641.34
NO (O_2=11%)	mg/m^3	545.67	174.86	12587.86	89.47
SO_2 (O_2=11%)	mg/m^3	1.40	8.70	61.05	0.00
NO_x (O_2=11%)	mg/m^3	836.60	2679.74	19299.15	137.18

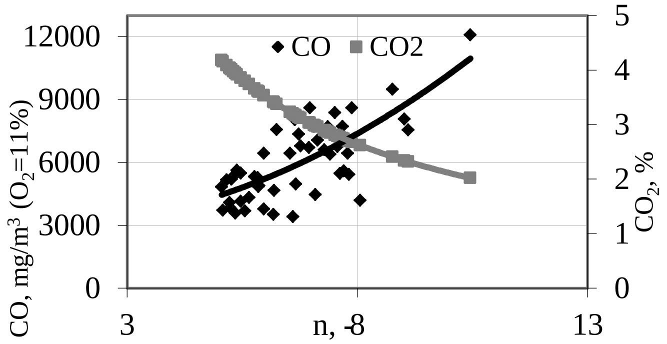

Figure 2 Dependence of carbon monoxide and carbon dioxide on the excess air coefficient during combustion of the pellets from rape straw

Burning of rape straw pellets under increased coefficient of quantity of excess air (see Figure 2) leads to an increase in the concentration of carbon monoxide according to the Equation (1):

$$CO\ (mg/m^3) = 86.878n^2 – 144.67n + 2966.8 \qquad (1)$$

The confidence level $R^2 = 0.53$.

The concentration of carbon dioxide is reduced according to the Equation (2):

$$CO_2\ (\%) = 21.157n^{-1.001} \qquad (2)$$

These concentrations were established within experiments over ten times the excess air coefficient. Above the higher excess air coefficient decreases combustion process and reduces combustion temperature below 160 °C (as shown in Figure 3). This flue gas cooling can be defined by the Equation (3):

$$T_{flue\text{-}gas}\ (^oC) = 0.4359n^2 – 14.864n + 264.12 \qquad (3)$$

The confidence level $R^2 = 0.44$.

The rapid increase of nitrogen oxides emissions was mainly caused due to the large amount of combustion air supply. It is depending on the amount of combustion air supply and under these conditions nitrogen reacted with oxygen to form nitrogen oxides. The effect of nitrogen oxides concentration in relation to the excess air coefficient is described by the Equation (4):

$$NO_x\ (mg/m^3) = 125.56n^{0.8307} \qquad (4)$$

The confidence level $R^2 = 0.67$.

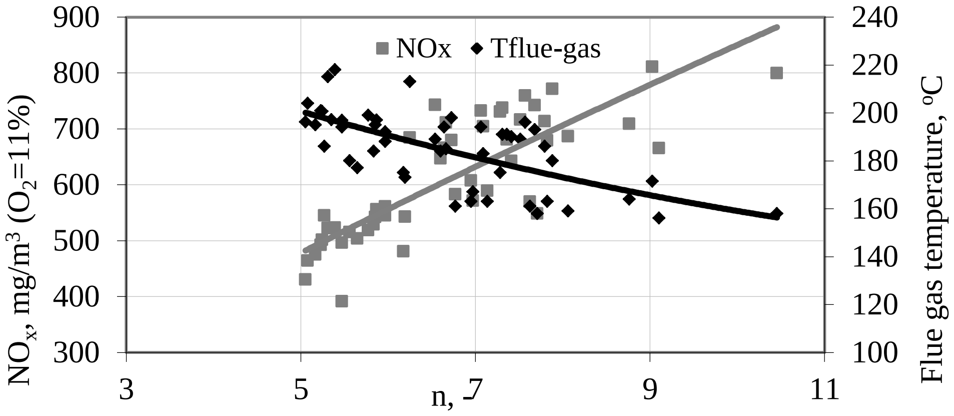

Figure 3 Dependence of nitrogen oxides and flue gas temperature on the excess air coefficient during combustion of pellets from rape straw

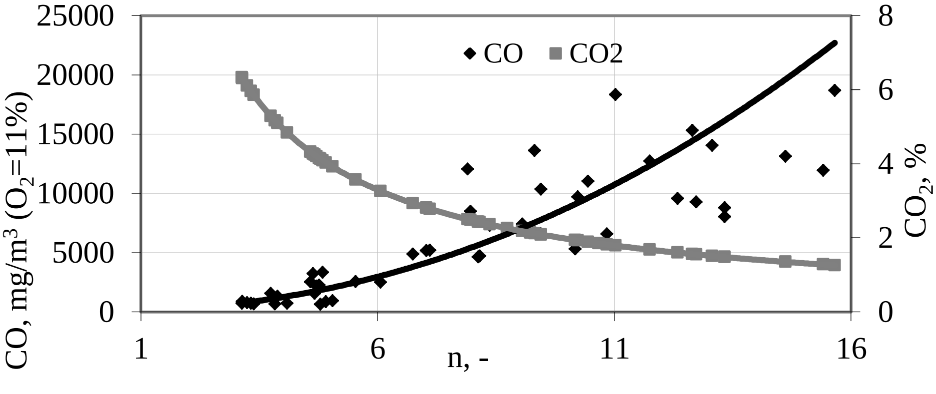

Figure 4 Dependence of carbon monoxide and carbon dioxide on the excess air coefficient during combustion of biochar from rape straw pellets

During combustion experiments of biochar samples (see Figure 4) very high concentrations of carbon monoxide emissions in areas of increasing excess air coefficient were measured. The measured data value increased when excess air coefficient rose according to the Equation (5):

$$\text{CO (mg/m}^3\text{)} = 65.936x^{2.1235} \quad (5)$$

The confidence level $R^2 = 0.75$.

The course of the concentration of carbon monoxide was measured up to the value of the coefficient of excess air 16. The combustion process was then stopped. Carbon dioxide has a decreasing course depending on the amount of supplied combustion air according to the Equation (6):

$$CO_2\ (\%) = 19.889x^{-1.003} \quad (6)$$

With increasing excess air coefficient is decreased combustion process and reduced combustion temperature below 150 °C (as shown in Figure 5). This flue gas cooling can be defined by the Equation (7):

$$T_{\text{flue-gas}}\ (^{o}\text{C}) = 24.622x^{1.4109} \quad (7)$$

The confidence level $R^2 = 0.60$.

Significant quantities reached the nitrogen oxides emission concentrations as shown in Figure 4. There in particular during increasing amount of combustion air (excess air coefficient) was increasing NO_x concentration according to the Equation (8):

$$NO_x\ (\text{mg/m}^3) = -0.0859x^2 - 2.9239x + 196.8 \quad (8)$$

The confidence level $R^2 = 0.44$.

As far as carbon monoxide and nitrogen oxides emissions are concerned, the combustion device should work in nominal parameters (as evidenced by other research studies). Any uncontrolled change of the combustion material and air flow leads to high emissions and combustion temperatures reduction (Fiedler and Persson, 2009).

Every type of combustion device has its own characteristic behaviour of carbon monoxide emissions. The greatest emissions occur mainly during the ignition and stopping of combustion device (Friberg and Blasiak, 2002). Largest emission concentration of carbon monoxide and nitrogen oxides is achieved at high excess air coefficient. The high amount of combustion air cools the combustion chamber and results in high emissions of carbon monoxide in the flue gas as has been confirmed by experimental work of author Johansson et al. (2003).

From the content of carbon dioxide (CO_2) we can determine the quality (effectiveness) of the combustion process (Fiedler and Persson, 2009). In the case of a low excess air process (complete combustion), involving the highest possible concentration of CO_2, the losses caused by combustion products (flue gases at the same temperature) are minimal. For each liquid and solid fuels, there is a maximum achievable proportion of

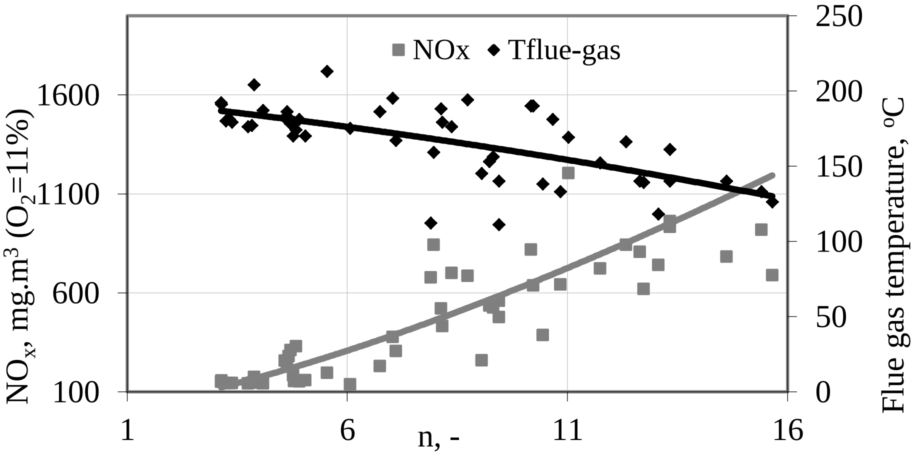

Figure 5 Dependence of nitrogen oxides and flue gas temperature on the excess air coefficient during combustion of biochar from rape straw pellets

carbon dioxide CO_2 (so-called stoichiometric CO_{2max}) in the flue gas, which is given by the elemental composition of the fuel combustible part. This value is not within the scope of our experimental measurement.

4 Conclusions

Experimental work includes the determination of elemental analysis of individual samples used for solid biofuels, determining stoichiometric analyses that are the basis for the assessment of thermal-emission properties of samples with the interaction of the combustion device with the lower fuel supply. The resultant values derived from elemental and stoichiometric analyses show that modified pyrolysis fuel samples have different properties then unmodified samples, which in turn affect their future energy recovery.

One of the greatest advantages of the modified samples was the fact that the calorific value of those samples almost doubled. On the other hand, the proportion of volatile matter in these pyrolysis processed fuels significantly reduces the water content. Pyrolysis processed samples have not already classical characteristics of biomass fuels, but have similar characteristics as coal.

The outcomes of our work can be very useful in solving problems related to suitability assessment and optimization of combustion processes, before and after pyrolysis processing of solid samples from rape straw pellets in combustion devices, particularly with reference to the thermal emission concentrations of CO, CO_2 and NO_x. The mass flows, emissions factors and characteristics of solid particles depending on the excess air coefficient are specified for each sample. These results show non-uniformity in the final thermal-emission parameters.

For optimising energy efficiency in combustion of modified fuels it is important to keep a stable supply of combustion air, in order to prevent or minimise losses of combustion of residual chemical imperfections and loss of heat. Differential thermal-emission parameters of assessed biofuel samples were defined during experimental measurements of thermal-emission concentration at the selected combustion device. Results from stoichiometric analysis have shown that an increase of air quantity is necessary for complete combustion of the biofuel samples, after pyrolysis processing. These final values of thermal-emission measurements are described by statistical regression analysis, where the regression equation is determined with a confidence level of each compared parameters.

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