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EMMISSION OF SULPHUR OXIDES FROM AGRICULTURAL SOLID BIOFUELS COMBUSTION

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ABSTRACT

In the aspect of the course and analysis of products of biomass fuels combustion in grill feed boilers, the combustion process of wheat straw and meadow hay were assessed taking into consideration conditions of SO_2 emission. Different types of briquettes used in the research not only had various chemical properties but also physical properties. In the aspect of assessment of energy and organic parameters of the combustion process, the sulphur content in biomass becomes a significant factor at its energy use. Registered emission during combustion of meadow hay biomass referred to wheat biomass was for A and B type briquettes correspondingly higher by ca. 320 and 120%. Differences in SO_2 emission at combustion of various biofuel forms in the aspect of the relation with the remaining combustion parameters including mainly with air flow require, however, further research that leads to development of low-emission and high-efficient biofuel combustion technologies in low-power heating devices.

Introduction

Plant biomass is a very popular energy carrier. However, its different chemical and physical properties (Demirbas, 2004; Denisiuk, 2020; Gorzelany et al., 2020), so clear for agricultural biomass, causes numerous problems with its use in individual heating devices. Besides the problems with sintering of ash, sediment and slag of which deteriorate the air flow causing uncontrolled emission and corrosion related to the content of chloride in biomass, participation of nitrogen and sulphur in its structure is also significant. During combustion, they form oxides of these elements and free to atmosphere (Boman et al., 2011; Dyjakon, 2012; Jenkins et al., 1998; Wang et al., 2011).

Among these compounds, emission of nitrogen oxides is a subject of many research (Juszczak, 2014; Kjällstrand and Olsson, 2004; Nussbaumer, 2002; 2003; Olsson and Kjällstrand, 2006). In case of solid biofuels relations concerning occurrence of fuel nitrogen above

0.6% of weight participation are noticeable, which is particularly important for grasses (Boman et al., 2011; Demirbas, 2004; Jenkins et al.; 1998; Nussbaumer, 2002; 2003; Obernberger et al., 2006). Some studies also show that NO_x emission depends on the sulphur content in biomass (Demirbas, 2005). The problem of NO_x emission is so serious that the admissible level of the emission is regulated with relevant provisions (Directive 2001/80/EC, EcoDesign Directive, Resolution 2005).

In case of emission of sulphur oxides, the use of biomass is favourable in comparison to replacement of fossil fuels which contain considerable amounts of this element and cause emission of these oxides. Calculated per sulphur the content of this element from the fossil fuels combustion processes more or less corresponds to the amount of sulphur freed in natural processes that take place in the environment e.g. decomposition processes of volcanos' emission. Sulphur emission in the form of sulphur dioxide in exhaust gases from fossil fuels occurs in 95% as a result of oxidation of this element that is contained in fuel. Exhaust gases contain also sulphur trioxide, but it is a small 0.5% amount and this compound condensates fast on the surface of the boiler forming sulphuric acid with water vapour. Moreover, co-combustion of biomass with carbon, which reduces SO_2 emission, is also interesting (Kordylewski, 2008).

The sulphur content in biomass is unfavourable due to formation of sulphur oxides (SO_2 and SO_3) which cause in some conditions, corrosion of steal parts of the boiler (Bukowski and Hardy, 2006; Jewiarz et al., 2012; Kordylewski, 2008; Hardy et al., 2009; Shao et al., 2012).

Sulphur contained in fuel in the combustion process is combined in ash or frees to atmosphere together with exhaust gases mainly in the form of sulphur dioxides (SO_2) and more rarely of sulphur trioxide (SO_3). Due to cooling of combustion gases in a part of the boiler, SO_x forms sulphates and condenses on the surface of fine volatile ash. It also forms particles on the surface of a heat exchanger or reacts directly with particles of volatile ash deposited on the surface of a heat exchanger (sulphation). Nussbaumer (2002) indicates that even from 60 to 90% of sulphur included in timber and bark in the combustion process combines with ash. In case of wheat straw combustion, there was only 40-55% of its content in fuel. The remaining sulphur included in fuel is contained in combustion gases in the form of aerosols in the form of SO_2 (in small amounts as SO_3). Thus, at timber combustion due to low concentration of sulphur, SO_2 emissions are usually non-significant. According to Obernberger (2003), problems related to sulphur emission should be expected at the concentration in fuel above 0.2% of weight. It may be important in case of agricultural biomass combustion such as rapeseed straw and grass. Classification of SO_2 emission from biofuel combustion made of such agricultural biomass through timber biomass seems to be incorrect. Moreover, there are no relevant rules that would control admissible values of such emission in heating devices of low-power which as a dispersed energy generation source with its amount participation in the produced thermal energy may constitute a considerable source of local emission. Supervision over SO_2 emission is carried out only for big objects for energy combustion (Directive 2001/80/EC).

The aim of the research was the analysis of sulphur oxides emission from the combustion process of two types of briquettes that differ with regard to geometrical properties, which for each type A and B were made of wheat straw and meadow hay.

Material and Methods

Raw materials and agglomeration

Biomass used during the research came from holdings in Lubelskie Voivodeship. Two types of green biomass were selected for analyses: wheat straw and meadow hay to differentiate the research material with regard to the sulphur content. Samples of the selected raw materials with the mass of 25 kilo were collected from the field, then fragmented with a hammer mill H111 with 10 mm sieves. So prepared raw material was subjected to the briquetting process in the hydraulic briquetting machine by Por company Junior model with a diameter of a sleeve of 50 mm obtaining A briquette and in the screw briquetting machine by EnEco model JW-08 obtaining briquette type B. The following methods were used for determination of chemical properties:

- carbon, hydrogen, nitrogen - measurement with an automated analyser acc. to the standard PN EN ISO 16948:2015;
- sulphur - measurement with the automatic analyser according to the standard PN EN ISO 16994:2016;
- calorific value - according to the standard PN-EN ISO 18122:2016;
- ash- according to the standard PN-EN ISO 18122:2016.

While, at determination of physical properties, the following methods were used:

- diameter and length - taking a direct measurement with a calliper of 10 randomly selected briquettes according to the provisions of the standard PN-EN ISO 16127:2012;
- moisture - with a gravimetric method according to the standard PN-EN ISO 18134-3:2015;
- density - calculated based on the geometrical properties in selected samples and the biofuel portion mass using the following formula 1:

$$\rho_w = \frac{4 \cdot 10^6 \cdot m}{\pi \cdot d^2 \cdot l} \; (\mathrm{kg \cdot m^{-3}}) \qquad (1)$$

where:

ρ_w – density of briquette, ($\mathrm{kg \cdot m^{-3}}$)
m – briquette portion mass, (g)
d – average diameter of briquettes, (mm)
l – sum of the briquettes length in fuel portion, (mm)

Combustion tests

Combustion process of the so prepared fuels was carried out in a grill heating device supplied periodically with fuel portions with the mass of 1 kilo. This process was analysed on account of the following parameters: composition of exhaust gases, process timing and combustion gases temperature. Recording of these parameters was constant i.e. from the moment of ignition initiation to the moment of reaction extinction, which is reduction of temperature of combustion gases to the value of 100°C. It corresponded to obtaining a stabilized layer of heat constituting a relevant base for initiation of flame during everyday exploitation at supplementing fuel.

Air in the combustion process was supplied under the grill through a fan with a speed of 1 $\mathrm{m \cdot s^{-1}}$. Fumes were collected from a chimney. A measuring probe was combined with a combustion gas drier, from which exhaust gases got to the exhaust gas analyser. During the tests, a mobile exhaust gas analyser operating based on infrared sensors NDIR was applied for the following gases: CO, CO_2, NO, SO_2 and electrochemical – O_2. Temperature was measured with the use of a thermocouple K type which was located in the exhaust gas collection probe. Results were registered automatically for the data base of the analyser every 2 seconds with a uniform recording of the data registration time. SO_2 concentration in exhaust gases was referred to the dry exhaust gases volume with the oxygen content of 10% in normal conditions ($mg \cdot m^{-3}$) 0°C and 1013 mbar, according to the guidelines included in the standard PN-EN 303-5:2002.

Statistical analysis

The obtained results with the use of the packet for analysis of data Statistica 13.1 PL were subjected to Anova analysis for factor systems. Before these analyses compliance of results with a normal distribution with the use of Shapiro-Wilk method was verified, and the uniformity of variance was estimated with Brown-Forsyth test. Pearson correlation test was used for description of the relation between particular variables. Observed differences were recognised as statistically significant at the significance level $p<0.05$.

Results and Discussion

Chemical properties of raw materials were presented in table 1 and physical properties of briquettes in table 2. Raw materials used for research had similar features within the content of carbon, hydrogen, and calorific value. While, with regard to the content of nitrogen, sulphur and ash, meadow hay in comparison to wheat straw had higher values respectively by 78, 916 and 59% – Table 1. A chemical composition of the biomass does not differ from the data described in the literature. However, it differs in comparison to fossil fuels. Even though carbon content in the analysed biomass was comparable, carbonification of this biomass is considerably lower than fossil fuels. Differences in higher content of nitrogen and sulphur in hay than in wheat straw is also reflected in literature data (Demirbas, 2004; Mc Kendry, 2002; Obernberger et al., 2006).

Table 1.
Chemical properties of used raw materials

Parameter (Average values)	Unit	Wheat straw	Hay
C	(%)	47.70	46.04
H	(%)	5.50	5.64
N	(%)	0.77	1.37
S	(%)	0.06	0.61
Calorific value in the working state	($MJ \cdot kg^{-1}$)	16.32	16.14
Ash content in the working state	(%)	2.31	6.21

Table 2.
Physical properties of the produced briquettes

Parameter (Average values)	Unit	Briquettes			
		From wheat straw		From hay	
		A type	B type	A type	B type
Length	(mm)	15.5	66	23.4	55
Diameter	(mm)	50.7	87	50.7	87
Volumetric density	(kg·m^{-3})	956	397	965	385
Total water content	(%)	9.69	10.23	10.01	9.78

Bigger variability of the fuels used during the tests concerned geometrical properties. The use of two different briquetting machines caused that A type briquettes were shorter and had a smaller diameter but had greater thickness than B type briquettes. However, differences in these parameters between the used raw materials were comparable, only the length of briquettes in the raw material group was slightly different. Whereas the water content for all agglomerates was comparable and amounted to ca. 10% - Table 2.

At the same time, briquettes used for research as a rule meet the requirements of the quality standard PN-EN ISO 17225-7:2014-8 with regard to the diameter and length. According to the norm of length of briquettes, it should be within 40 to 400 mm or more, thus the agglomerate from wheat straw within this scope was below the norm. The remaining physical properties (density and water content) also were within the range provided in the relevant literature (Denisiuk, 2009; Niedziółka et al., 2013).

Figures 1-2 present time distribution of SO_2 content in exhaust gases registered during combustion of biofuels from wheat straw and hay.

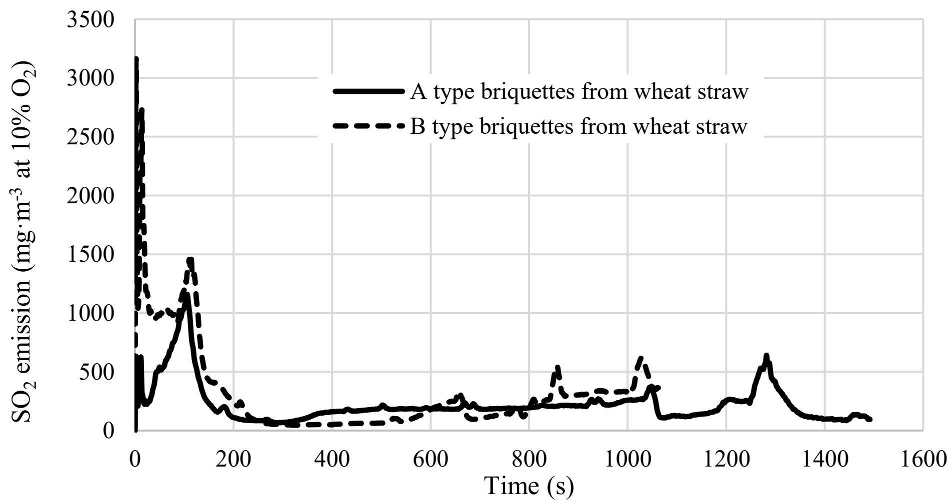

Figure 1. Changes of SO_2 concentration during combustion of briquettes from wheat straw

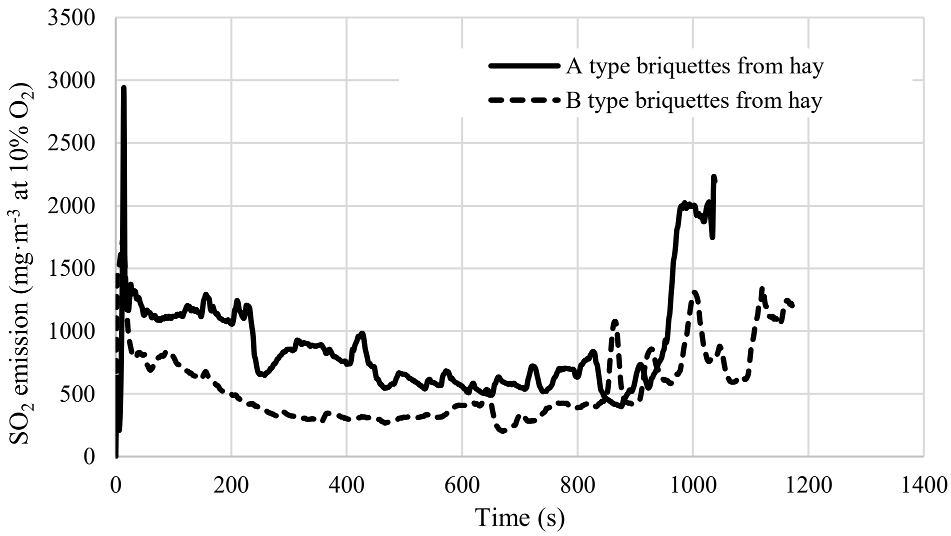

Figure 2. Changes of SO_2 concentration during combustion of briquettes from hay

During combustion of the analysed briquettes made from wheat straw and hay, differences in time of their combustion were reported. Type A briquettes from wheat straw had the slowest combustion, while portions of the remaining briquettes combusted by 5 minutes faster - figure 1-2. During the tests, directly after the start of combustion of a fuel portion, an increased SO_2 emission to atmosphere with the value of ca. 3000 $mg \cdot m^{-3}$ at 10% O_2 was reported. Together with intensification of combustion, reduction of emission was reported, which for wheat straw biofuels was within 50-300 $mg \cdot m^{-3}$ at 10% O_2, while for hay it was within 200-700 $mg \cdot m^{-3}$ at 10% O_2. Simultaneously during combustion of briquettes, the increase of emission was observed in the final stage of combustion which was higher during the use of hay briquettes – fig. 1 and 2. Variability of the geometrical form of combusted briquettes also indicates the impact on the registered emission. In the basic combustion phase, which in the test conditions was between 300 and 800 second it was observed that B type briquettes fire with a lower SO_2 emission. While in the remaining combustion periods, these differences were not so clear. The statistical analysis that was performed showed significant differences in SO_2 emission both during combustion of briquettes from wheat straw and hay and between the discussed types of agglomerate. Average values of the analysed variables with a division into quality factors of the used raw material and type of briquettes were presented in figure 3.

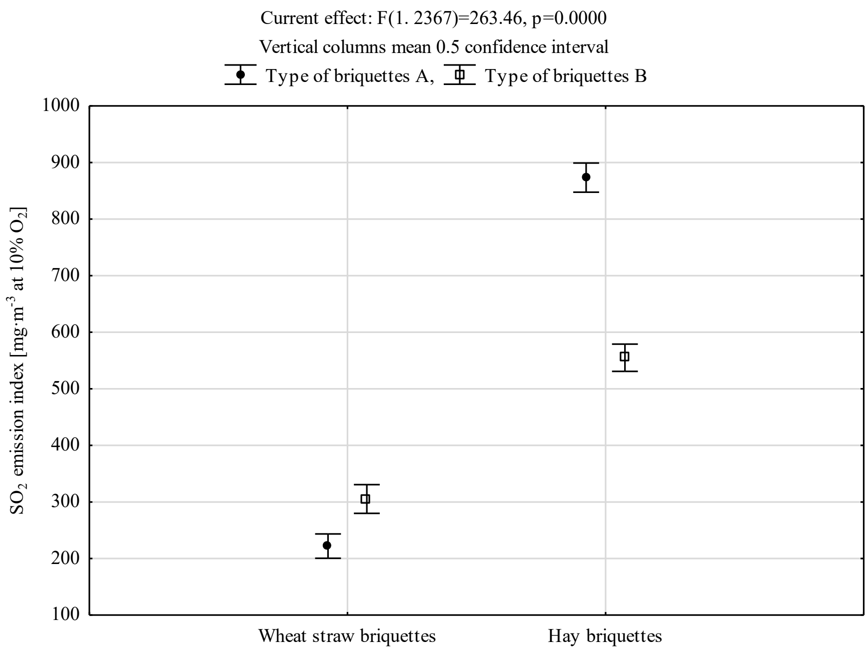

Figure 3. Average values of SO_2 concentrations during combustion of 1 kilo of briquettes from wheat straw and hay

Biofuels used in the study had various chemical and physical properties among which different geometrical properties are visible. In the aspect of the study range the sulphur content in fuel was significant which for meadow hay biomass referred to wheat straw biomass was higher by almost 916%. During the tests, an increased emission at combustion of briquettes from meadow hay was observed which for the A type briquettes was on average by 320% higher than for briquettes of this type made of wheat straw. While, for the B type briquettes emission at combustion was on average higher by 120%.

Table 3 presents relations between the emitted SO_2 and the remaining elements of exhaust gases (CO, NO) temperature of gases (T_{gas}) air excess index (λ).

SO_2 emission for wheat straw biofuels in relation to air excess in exhaust gases had weaker relations which with the use of combustion of briquettes from hay were statistically insignificant. This emission showed much stronger and in majority of cases - negative relations with exhaust gases temperature which reflected the temperature in the combustion chamber. The strongest positive relations of SO_2 emission were observed with CO emission. Whereas the analysed emission with NO emission at the use of wheat straw briquettes proved quite strong relation and at combustion of briquettes this relation became non-significant. But for A type briquettes these were positive relations and for B type -negative ones- table 3.

Table 3.
Pearson correlation coefficients between gas combustion products, temperature of gases and air excess

Variables	Wheat straw briquettes		Hay briquettes	
	A type	B type	A type	B type
SO_2 vs CO	-0.108	0.639	0.808	0.745
SO_2 vs NO	0.564	-0.622	0.115	**-0.070**
SO_2 vs T_{gas}	0.578	-0.770	-0.189	-0.453
SO_2 vs λ	-0.278	0.425	**-0.026**	**0.045**
CO vs NO	-0.593	-0.695	0.335	0.332
CO vs T_{gas}	-0.679	-0.542	**0.037**	**-0.058**
CO vs λ	0.276	0.173	-0.130	-0.370
T_{gas} vs λ	-0.516	-0.554	-0.370	-0.771

Values in bold are statistically insignificant

Analysing the collected data, one may state that the emission value is mainly affected by the sulphur content in fuel and conditions of the combustion process which favoured CO emission. Other researchers indicate the emission problems related to a higher sulphur content in some biofuels types, particularly these of agricultural origin (Demirbas, 2004; Juszczak, 2014; Obernberger, 2006; Verma, 2011a; 2011b). Nussbaumer (2002) says that problems with the increased SO_x emissions occur at the weight participation of sulphur from 0.2% and concerns mainly grass biomass. Simultaneusly, he indicates that the increased $S0_2$ emission is accompanied by CO emission. While Fournel et al., (2015) showed in their paper that delay in biomass harvest from autumn to spring reduced emission of NO_x (0-11%), SO_2 (11-54%) and PM (0-37%) which is probably due to the decrease of the nitrogen content, chloride sulphur by 20-60% in biomass as a result of washing out and leaching.

When analysing SO_2 emission during biofuels combustion, the assumed combustion system is also important. Combustion of solid biofuels on the grill causes uncontrolled emissions of CO to atmosphere and according to this research also of SO_2, particularly visible at the initial combustion phase, directly after placing the fuel portion on the heat layer and at the end of its combustion. Similar problems during periodical combustion of a fuel portion was observed by Juszczak (2013) when he analysed emission of CO and NO_x during combustion tests of wood pellets in a chamber with the power of ca. 25 kW with a overfeed-channel furnace during which at periodic dosing together with the increase of the break in feeding pellets he observed a considerably bigger distribution of the CO concentration value than during continuous feeding. The relation he observed, he explained with the fact that at the constant air stream which was not reduced during the break in periodical dosing of pellets, the temperature in the combustion chamber and oxygen concentration changed more than during continuous feeding.

During the own research, SO_2 emission took place in similar conditions like in CO emission which is proved by quite strong correlations. A clear impact of exhaust gases temperature on the registered emission of SO_2 is observed. When the combustion process is fully efficient (between 300 and 800 second – fig. 1 and 2 - at a quite high temperature of combustion and respectively low air excess) the registered emission was lower. A low temperature in the combustion chamber at the beginning and end of the period results in secretion of such compounds and uncontrolled emission. However, air feeding within this time does not result in combustion improvement. A dimension of the combusted particles and movement of temperature inside them is crucial. As a result of the studies carried out by Yang et al. (2015) it was observed that a smaller size of fuel particles influences the CO concentration increase and the remaining gas compounds in exhaust gases that leave the bed. It partially explains a higher emission for smaller A briquettes made of hay. However, this relationship is not compliant with data for biofuels made of wheat straw. Therefore, confirmation of this rule requires more thorough tests.

Conclusions

A literature study performed during realisation of this research shows that variability of chemical properties is a characteristic feature of biomass and depends on the state and methods of processing of physical properties. Values of these parameters are different in relation to biomass type and size grade. Standardisation of fuels can regulate, first, the water content and geometrical properties. However, to a small extent, and most of the times, it does not influence the chemical composition of fuel at all. In the aspect of assessment of organic and energy parameters of the combustion process, the sulphur content in biomass becomes a significant factor that determines its energy use. The applied combustion process is not without meaning. This research shows that periodical, in portions, combustion on the grill of briquettes with a higher content of sulphur (made of hay) has a small ecological effect influenced also by various changes of temperature on the combustion chamber that results in a considerable CO emission and correlated with it SO_2 emission. However, the analysed relations of emission of these compounds with fuels dimensions and air flow require further research leading to development of low-emission and high-efficient technologies of biofuels combustion in heating devices of low power.

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EMISJA TLENKÓW SIARKI Z PROCESU SPALANIA BIOPALIW STAŁYCH POCHODZENIA ROLNICZEGO

Streszczenie. W aspekcie przebiegu i analizy produktów spalania paliw biomasowych w rusztowych kotłach zasypowych, oceniono proces spalania słomy pszennej i siana łąkowego pod kątem warunków powstawania emisji SO_2. Wykorzystane do badań różne rodzaje brykietów, charakteryzowały się nie tylko odmiennymi cechami chemicznymi ale również i właściwościami fizycznymi. W aspekcie oceny parametrów ekologiczno-energetycznych procesu spalania, zawartość w biomasie siarki staje się istotnym czynnikiem przy jej energetycznym wykorzystaniu. Rejestrowana emisja podczas spalania biomasy siana łąkowego w odniesieniu do biomasy pszennej była dla brykietów typu A i B odpowiednio większa o około 320 i 120%. Różnice w emisji SO_2 przy spalaniu różnych form biopaliw w aspekcie relacji z pozostałymi parametrami spalania w tym głównie z przepływem powietrza wymagają jednak przeprowadzenia dalszych badań prowadzących do opracowania niskoemisyjnych i wysoko sprawnych technologii spalania biopaliw w urządzeniach grzewczych małej mocy.

Słowa kluczowe: biomasa roślinna, spalanie, emisja, ditlenek siarki