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Movement of Chemical Elements in Ash on the Example of a Process of Combustion Corn Straw Briquettes in a Low Power Boiler

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1. Introduction

In Poland, the use of plant biomass for energy purposes is gaining importance. At the same time, the popularity of dendromass, despite changes in regulations increasing its supply for energy purposes, promotes an increased interest in a biomass of agricultural origin. This trend is associated not only with professional energy industry, that is obliged to use biomass for energy production in the processes of its direct combustion or co-combustion with fossil fuels, but also depends on the increase in demand for different assortments of solid biofuels in single-family housing or municipal buildings, where coal boilers are replaced by modern heating devices powered by a biomass under the "anti-smog" program (Kamińska 2018, Wąsowicz 2018).

The barrier with the use of solid biofuels in the aspect of emission standards for low-power heating devices, is their diversification in terms of volatile parts and the contents of carbon, hydrogen, nitrogen and sulfur. These parameters depend on the physiological characteristics of different plant species and varieties, as well as their development phase, plant parts, habitat, date and method of harvesting, transport, storage and other factors (Demirbas 2004, Graham et al. 2016, Jenkins et al. 1998, Krzyżaniak et al. 2014, Szyszlak-Bargłowicz et al. 2006, Wang et al. 2011).

The plant biomass also contains some amount of mineral substance, which after combustion is the main part of the ash, not exceeding 6% (Demirbas 2004, Kalembasa 2006, Kowalczyk-Juśko 2017, Shao et al. 2012). Chemical composition of ash (tendency to form sinters) affects the course of the combustion process, the choice of technology, emission of solid particles and after combustion – manner of its use. However, the management of ash is determined by its chemical properties. Ashes obtained from the combustion of straw, compared to biofuels with dendromass, contain relatively little calcium, a lot of potassium and silicon (Kraszkiewicz et al. 2017, Obernberger et al. 2006, Róg 2011, Uliasz-Bocheńczyk & Mokrzycki 2018). At the same time, longer vegetation period of trees than other plants used for energy purposes is conducive to the accumulation of other metals. Among wide range of biomass fuels, corn straw is an attractive raw material. Maize is one of the main cereal crops grown in Poland, and its straw is considered an interesting source of biomass for energy purposes (Karcz et al. 2013, Niedziółka & Zuchniarz 2006). While analyzing the combustion process and explaining the mechanism of burning straw from maize on a fixed bed contributes to its clean and efficient use as an energy source.

Metals contained in the biomass, evaporate or remain in ash during the combustion process and consequently get into the natural environment (Li et al. 2015, Rybak 2006). Kalembkiewicz and Chmielarz (2013) [after (Ram & Masto 2010, Pandey & Singh 2010, Johnson et al. 2010, Ahmaruzzaman 2010, Pandey et al. 2009, Reijnders 2005, 2007, Onisei et al. 2012, Dong et al. 2010, Syc et al. 2012)] indicate in their work that various applications of ash (reclamation, construction of embankments, production of building materials, flue gas desulphurization, production of ceramics, membranes and geopolymers) cause that knowledge of heavy metals distribution within ash as a mobile fraction is necessary before their use. The problem of heavy metals division between waste streams during combustion of Virginia mallow also draws attention of Szyszlak-Bargłowicz and Zając (2015), who indicate the need to analyze the problem in low power installations using biomass fuels, referring at the same time to the results obtained in this area by researchers dealing with industrial installations, in which fossil fuels were combusted.

Combustion is a complex phenomenon. It consists of many physical and chemical phenomena of thermal decomposition and combustion of fuel, occurring in a given space and time. Analysis of the combustion process requires knowledge of the properties of fuels and their impact on its course (Van Loo & Koppejan 2008, Villeneuve et al. 2012, Juszczak 2014, Liu et al. 2013, Ozgen et al. 2014). The type of pollution generated in the biomass combustion process depends not only on the process factors, heating equipment used, but also on the type of biomass being combusted (Zając et al. 2017, Konieczyński et al. 2017).

The aim of the conducted research was to assess the migration of elements within the ash during the combustion process of maize straw briquettes in the low power boiler in the aspect of their impact on the natural environment, and taking into account directions and possibilities of potential management of ashes.

2. Material and methods

Maize straw briquettes were used during the tests. The material for the study was obtained from an agricultural farm in the eastern part of Lublin province. Compaction process was carried out in a hydraulic briquetting machine with a cylindrical barrel with a diameter of 50 mm. For the obtained biofuels, basic physical and chemical properties were determined in three repetitions, applying the following methods:

- moisture content – by weight method according to the norm PN-EN 18134-3:2015,
- density – calculated on the basis of the mass, diameter and length of the briquettes from the randomly selected sample of 10 pieces, using the following formula:

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

where:
ρ_w – density of briquttes ($kg \cdot m^{-3}$),
m – mass (g),
d – diameter (mm),
l – length (mm).

- carbon, hydrogen, sulfur – by means of IR absorption;
- nitrogen – by the katharometer method;
- net calorific value – calculated after previous determination of the heat of combustion according to the norm PN-EN 18125:2017;
- ash – according to the norm PN-EN18122:2016;
- chemical composition of the ash was made by plasma spectrometry using the Thermo iCAP 6500 Duo ICP device.

The combustion tests of the collected research material were carried out using a test bench (Figure 1), the integral element of which was a upper-combustion boiler with a fixed grate periodically loaded, in which the fan directed the air stream under the grate with the speed of 1.5 $m \cdot s^{-1}$.

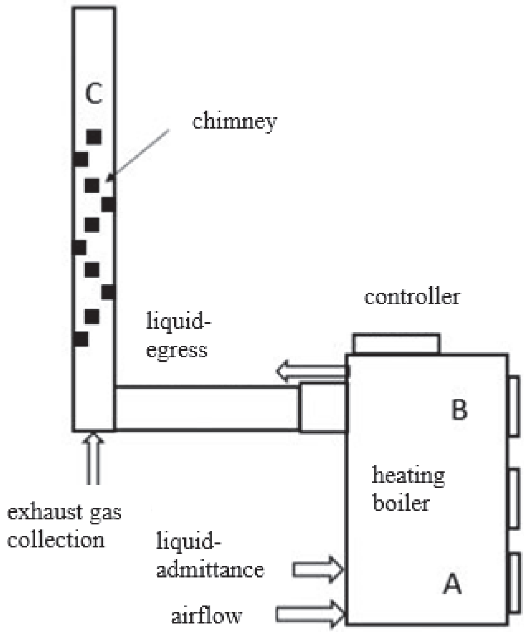

Fig. 1. Schematic of the measurement system (place for ash sampling: A ash pan, B heat exchanger, C chimney shelves marked with a black square)

Capacity of the combustion chamber to the exchanger part was 35.1 dm^3 (0.26 m × 0.3 m × 0.45 m). Above the combustion chamber, there was a heat exchanger with horizontal partitions with water channels. The ash chamber was below the water grate, in which the ash container was placed. The fuel was loaded and ash removed manually. Water capacity of the water jacket was 30 dm^3, and the storage tank 400 dm^3. Such a system was supposed to ensure similarity to real chambers and enable combustion process, as in low-power heating devices. The exhaust gases were taken through the chimney at a distance of 1 m from the boiler flue. The measuring probe was connected to the exhaust gas drier PGD-100 (Madur Eljack Electronics), from which the exhaust gases were sent to the flue gas analyzer. During the tests, a Photon portable gas analyzer, from the same company as the gas dryer, was used. It is a device operating on the basis of infrared sensors (NDIR) for the following gases: CO, CO_2, NO, SO_2 and electrochemical sensor for O_2. The temperature measurement was carried out using a K type thermocouple. The first of them was located in the middle part of the combustion chamber height at its outer wall, while the second one was to measure the temperature of the flue gas in the chimney and was located near the flue gas collection point.

The tests consisted of burning 20 portions of 1 kg of previously mentioned briquettes. Measurements of the exhaust gas composition were carried out continuously from the moment of the fuel lining to the stabilized layer of heat until the reaction extinguished. Results of the discussed parameters were automatically recorded by the analyzer's database every 4 s, with the simultaneous recording of the data recording time. The database created in this way was transferred to a PC computer after the tests were completed.

The ash for laboratory tests was taken from three points of the combustion installation: from the ash pan of the boiler, heat exchanger and chimney, in which 10 metal shelves of 20 mm × 20 mm were arranged on the circumference of the chimney every 120° on the 50 cm section, were installed to collect the ash (Figure 1). Prepared portions of ash, after previous preparation (digestion and dilution), were tested using SpectroBlue ICP OES spectrometer at the Regional Center for Environmental Research, Agricultural Technology and Innovation, the Pope John Paul II State School of Higher Education in Biała Podlaska. The analytical curves were prepared by diluting the VHG SM68-1-500 Element Multi Standard 1 in 5% HNO_3. Operating parameters were as follows: number of measurements: 3; pump speed: 30 rpm; coolant flow: 12 $dm^3 \cdot min^{-1}$; auxiliary flow: 0.90 $dm^3 \cdot min^{-1}$; nebulizer flow: 0.78 $dm^3 \cdot min^{-1}$.

3. Results and discussion

The average values of the obtained results (from three replications) characterizing the biofuels used are presented in Tables 1 and 2.

Table 1. Physical and chemical properties of briquettes made of maize straw

Parameter	Symbol	Unit	Value
Length	L	mm	23
Diameter	D	mm	50
Density	–	$kg \cdot m^{-3}$	900
Total moisture	W_t^r	%	8.00
Elemental composition	C	%	45.0
	H	%	5.50
	N	%	0.70
	S	%	0.18
Calorific value	Q_s^a	$MJ \cdot kg^{-1}$	18.81
Net calorific value	Q_i^r	$MJ \cdot kg^{-1}$	17.58
Ash	A^a	%	6.77

Table 2. Chemical composition of ash from briquettes made of maize straw

Chemical element	Unit	Value
Si	$g \cdot kg^{-1}$ dry mass ash	270.67
P		16.24
K		147.70
Ca		41.71
Mg		10.08
Na		2.37
S		7.34
Fe		9.61
Al		13.80
Mn		0.70
Ba		0.18
Ti		1.47
Sr		0.18

The produced briquettes were characterized by comparable physico-chemical features to other biofuels based on maize straw, thus meeting the assumptions of appropriate quality standards (Demirbas 2004, Eisenbies et al. 2016, Obernberger 2006). The elemental composition of ashes obtained after combustion at 550°C by a standardized method (PN-EN 18122: 2016) varied. In the structure of elements determined with this method, particularly high silicon (Si) content was observed – 50% compared to the other elements and 28% potassium (K). Following elements had a few percentages: phosphorus (P), calcium (Ca), magnesium (Mg), sulfur (S), iron (Fe), and aluminum (Al). The remaining ash constituents made up small shares of less than 1% (Na, Mn, Ba, Ti, Sr). At the same time, the content of macro- and micronutrients in the ash from the analyzed biofuels was comparable to those from the literature data (Jewiarz & Kubica 2012, Kalembasa 2006).

The average values of CO, NO, SO_2, O_2 content in the flue gases and flue gas temperatures are presented in Table 3.

Interpretation of the obtained results concerning the content of CO, NO and SO_2 in the exhaust gas refers to the adopted combustion criteria, the characteristic feature of which was the use of a grate combustion system with fuel ignition from below and distribution of air under the grate. The obtained values of these compounds varied due to the periodicity of fuel feed to the boiler and phase of the combustion process. Immediately after lining the 1 kg of fuel along with a stabilized layer of heat, the temperature was increased and evaporation of volatiles from the fuel was mainly represented by maximum values of CO, NO and SO_2. Stabilization of the combustion process represented by minimum O_2 value led to a decrease in the value of these compounds in the exhaust gas, and for CO, it was acceptable for this type of boiler, falling into the 3rd class according to PN-EN 303-5: 2012. The analyzed components of exhaust gases during the tests were similar to the literature values (Jewiarz & Kubica 2012, Kordylewski 2008, Szyszlak-Bargłowicz et al. 2017).

Table 3. The content in the flue gas of CO, NO, SO_2, O_2 and the temperature of the exhaust during the combustion of the briquettes concerned

Parameter	Unit	Minimum	Maximum	Average	At min. O_2
CO	ppm	0	9925	3892	3396
CO at 10% O_2	$mg \cdot m^{-3}$	0	39311	7713	4408
NO	ppm	0	243	152	217
NO at 10% O_2	$mg \cdot m^{-3}$	0	413	288	303
SO_2	ppm	0	141	51	65
SO_2 at10% O_2	$mg \cdot m^{-3}$	0	382	172	194
O_2	%	10.44	20.95	13.66	10.44
Exhaust gases temperature	°C	9.8	397.2	285.0	389.8

Among the pool of 34 analyzed elements, for 12 (Cd, Ce, Cr, Gd, Ho, In, La, Lu, Na, Sm, Y, Yb) the recorded results were below the measuring range of the device. The remaining values of elements determined in ash are shown in Table 4.

For 12 elements, the content in the ash from the chimney was much larger than in the ash from the other two sampling points within the boiler. These elements were: Al, Co, Cu, Fe, Mn, Ni, Pb, Rb, Sc, Tl, V and Zn. The observed differences were from several to several hundred percent. Observed results partially coincide with the classification of elements migration in ashes deposited at various places of the installation presented by Meij and Winkel (2007). This classification contains 3 groups, in which group 1 are elements accumulating in ash, group 2 are elements concentrating in fly ash, while the third group are the most easily evaporating elements, often present in the gaseous phase in the exhaust gases (they were not determined during the tests). In the conditions of own research, following elements were convergent with this classification in group 1: Ca, K, Mg, Sr, Th, while in group 2: Co, Cu, Ni, V, Pb, Tl, Zn. Differences that appeared in comparison with this classification (especially in group 1: Al, Fe, Sc) may indicate some anomalies that appeared during tests in the assumed combustion conditions.

Table 4. Chemical composition of ash depending on the sampling point

Chemical element	Sampling point			Chemical element	Sampling point		
	ashpan	exchanger	chimney		ashpan	exchanger	chimney
	in mg·kg^{-1} d.m. ash				in mg·kg^{-1} d.m. ash		
Al	4061.27	3693.95	**20916.93**	Ni	0.00	6.69	**91.41**
Ba	**3988.44**	22.93	186.20	P	14965.32	**15307.01**	7964.32
Ca	**35968.21**	32149.68	16940.10	Pb	564.16	75.48	**603.39**
Co	10.98	1.59	**90.89**	Rb	13.58	11.46	**368.23**
Cu	38.15	50.00	**125.26**	Sc	0.00	9.55	**383.59**
Er	0.00	**750.64**	375.52	Sr	**289.31**	247.77	149.22
Fe	4376.01	10716.24	**12162.60**	Th	30.92	**31.53**	24.22
K	**690.75**	660.19	0.00	Tl	33.53	35.99	**269.27**
Mn	623.41	733.76	**2211.46**	U	91.33	**142.36**	0.01
Mg	**12085.26**	11489.81	6425.26	V	51.73	49.36	**72.66**
Nd	**26.88**	21.02	14.58	Zn	1574.28	497.77	**20054.17**

* The maximum values are marked in bold

4. Summary

Under the test conditions, the fuel used in the form of maize straw briquettes did not differ from other fuels of this type. The combustion criteria adopted during the tests, the characteristic feature of which was the use of a grate combustion system with bottom fuel ignition and air distribution under the grate, was acceptable in the scope of CO emissions for this type of boiler, falling into the 3rd class according to PN-EN 303-5: 2012. Loading the environment with gaseous components: CO, NO and SO_2, is significantly affected by combustion technology, combustion conditions and nitrogen content in the fuel. Choosing biofuels with the lowest nitrogen content for combustion should contribute to the reduction of NO emissions, which becomes another criterion for allowing heating devices to be marketed (Commission… 2015).

Despite varied chemical composition, the ashes analyzed in terms of the macro-, micronutrients and heavy metals load do not tend to exceed the limits set by relevant standards. The experimental results indicate that in low-power boilers fed with briquettes of maize straw, the ash components move. Anomalies that appeared for Al, Fe, Sc are particularly important. This process is important from the point of view of environmental protection, boiler durability and the possibility of using furnace wastes. Low-power boilers, that do not have exhaust gas treatment installations, in which biomass fuels are used, may be a source of harmful emissions.

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Abstract

In Poland, the use of plant biomass for energy purposes is gaining importance. At the same time, the popularity of dendromass, despite changes in regulations increasing its supply for energy purposes, promotes an increased interest in a biomass of agricultural origin. This trend is associated not only with professional energy industry, but also depends on the increase in demand for different assortments of solid biofuels in single-family housing or municipal buildings, where coal boilers are replaced by modern heating devices powered by a biomass under the “anti-smog” program. Biomass combustion is treated as a process neutral to the environment through the prism of CO_2 emissions. However, under certain conditions, the combustion of biofuels in individual heating systems can cause serious risks on the local scale for the environment and human health. The aim of the study was to assess the migration of elements in the ash during the burning process of maize straw briquettes in the low power boiler in the aspect of their impact on the natural environment, taking into account the directions and potential for the potential management of ashes. In the test conditions, the fuel used in the form of briquettes from maize straw does not differ from other fuels of this type. The combustion criteria adopted during the tests, whose characteristic feature was the use of a grate combustion system with fuel ignition from below and distribution of air under the grate, at minimum content O_2, was acceptable in the scope of CO emissions for this type of boiler, falling into 3rd class according to the standard PN-EN 303-5: 2012. The burning of the gas components CO, NO and SO_2 is significantly affected by combustion technology, combustion conditions and nitrogen content in the fuel. Choosing to burn biofuels with the lowest nitrogen content should contribute to the reduction of NO emissions, which becomes another criterion for allowing heating devices to be marketed. Despite the varied chemical composition, the ashes analyzed in terms of the macro environment, microelements and heavy metals do not tend to exceed the limits set by the relevant standards. The experimental results indicate that in low-power boilers fed with briquettes of maize straw, the ash components move. Anomalies that appeared for Al, Fe, Sc are particularly important. This process is important from the point of view of environmental protection, boiler durability and the possibility of using furnace wastes. Low-power boilers, which do not have exhaust gas treatment installations in which biomass fuels are used, may be a source of harmful emissions.

Keywords:
biomass combustion, emission, ash composition

Przemieszczanie się pierwiastków w popiele na przykładzie procesu spalania brykietów ze słomy kukurydzianej w kotle małej mocy

Streszczenie

W Polsce wykorzystanie biomasy roślinnej na cele energetyczne zyskuje na znaczeniu. Jednocześnie popularność dendromasy, pomimo zmian w przepisach zwiększających jej podaż na cele energetyczne, sprzyja zwiększonemu zainteresowaniu biomasą pochodzenia rolniczego. Trend ten wiąże się nie tylko z energetyką zawodową, ale również koresponduje ze wzrostem zapotrzebowania na różne sortymenty biopaliw stałych w budownictwie jednorodzinnym lub komunalnym gdzie wymieniono kotły węglowe na nowoczesne urządzenia grzewcze zasilane biomasą na mocy programu „antysmogowego”. Spalanie biomasy, jest traktowane jako proces neutralny dla środowiska w kontekście emisji CO_2. Jednak w pewnych warunkach spalanie biopaliw w indywidualnych systemach grzewczych, może powodować poważne zagrożenie w skali lokalnej dla środowiska i zdrowia ludzi. Celem przeprowadzonych badań była ocena przemieszczania się pierwiastków w popiele podczas procesu spalania brykietów ze słomy kukurydzianej w kotle małej mocy w aspekcie ich wpływu na środowisko przyrodnicze uwzględniając tym samym kierunki i możliwości potencjalnego zagospodarowania popiołów. W warunkach badań wykorzystane paliwo w postaci brykietów ze słomy kukurydzianej nie odbiegało od innych paliw tego typu. Przyjęte podczas badań kryteria spalania, którego charakterystyczną cechą było wykorzystanie rusztowego systemu spalania z zapłonem paliwa od dołu i dystrybucją powietrza pod ruszt, przy minimalnej zawartości O_2, była akceptowalna w zakresie emisji CO dla tego typu kotła mieszcząc się w 3 klasie według normy PN-EN 303-5:2012. Na obciążenie środowiska składnikami gazowymi CO, NO i SO_2 wyraźny wpływ ma technologia spalania, warunki spalania i zawartość azotu w paliwie. Wybór biopaliw o jak najmniejszej zawartości azotu powinien przyczynić się do zmniejszenia emisji NO, która staje się kolejnym kryterium dopuszczającym urządzenia grzewcze do obrotu. Pomimo zróżnicowanego składu chemicznego analizowane popioły pod kątem obciążenia środowiska makro-, mikroelementami i metalami ciężkimi nie wykazują tendencji do przekroczenia wartości granicznych określonych odpowiednimi normami. Wyniki eksperymentalne wskazują, że w kotłach małej mocy zasilanych brykietami ze słomy kukurydzianej zachodzi przemieszczanie się składników popiołu. Szczególnie istotne są anomalie, które pojawiły się dla Al, Fe, Sc. Proces ten jest istotny z punktu widzenia ochrony środowiska, trwałości kotłów oraz możliwości wykorzystania odpadów paleniskowych. Kotły małej mocy, nieposiadające instalacji oczyszczania spalin w których stosuje się biopaliwa stałe mogą być źródłem emisji substancji szkodliwych.

Słowa kluczowe:
spalanie biomasy, emisja, skład popiołu