

Cyclone furnace as a way for mercury removal from lignite

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Abstract. The power plants will have to meet the requirements regarding permissible Hg emission levels by 2021. A particularly unfavourable situation is observed for front of lignite, which contains more mercury than hard coal, and in addition its combustion also causes more CO₂ emissions. The paper presents the proposal to use the cyclone furnace in the process of heat treatment for the process of the release of mercury from lignites. The structure and methods of operation of the cyclone furnace were presented. The paper also discusses the methodology of the process of numerical modelling of combustion and gasification of coal dust. The results showed that changing a fuel stream fed to the cyclone furnace allows for controlling the temperature and degree of fuel devolatilization. This leads to heating of the fuel to the desired temperature due to the expected level of mercury removal from the fuel. This is very important especially in the case of fuels containing significant amounts of moisture, such as lignite. The experiments performed in the study confirmed the results of numerical calculations for heating fuel and showed the possibility of over 90% removal of mercury from the fuel. The results confirmed the laboratory workplace rotary kiln.

1 Introduction

Processes of fossil fuel combustion for the purposes of electricity and heat generation contribute to the degeneration of the natural environment. The contaminants removed from flue gas include dust, sulphur oxides and nitrogen oxides. Technologies are currently being developed to separate carbon dioxide from flue gas in order to store it underground [1, 2]. Limitations of heavy metal emissions (such as mercury) are expected to be introduced in the nearest future. Mercury is an element which is more difficult to be removed from flue gas compared to the previously mentioned compounds as its content in carbon and, consequently, in flue gas, is insignificant. Medium content of mercury in Polish energy coals is around 80 ng/g [3, 4]. This insignificant mercury content in carbon, combined with huge amounts of burnt coal, results in emissions of mercury at the level of 50 Mg mercury per year per one power plant [5]. Total mercury emissions from anthropogenic sources in Poland is 11 Mg/year [6]. Of this number, over 9 Mg is caused by combustion of carbon fuels. Around 1.1 Mg is emitted as a result of non-ferrous metals production and 0.7 Mg - from cement production [7].

Limitation of mercury emissions can be reached by flue gas cleaning or through removal of mercury from coal before combustion. Removal of mercury vapour from flue gas (post-combustion methods) is burdensome since the amount of sorbents used is very high and

content of mercury in the flue gas stream is very small. Furthermore, fly ash is also contaminated with activated carbon, mostly used to capture mercury vapours together with the adsorbed mercury and non-reacted sorbent. A promising option is to remove mercury from fuel before combustion (pre-combustion methods) which allows for cleaning the coal and than use it in various areas of the industry. One of these methods is low-temperature thermal processing, with its efficiency reaching over 90% at temperature of ca. 400°C, which was demonstrated in numerous publications [8-10]. Another concept of mercury removal from fuel was proposed in the paper [11]. Overheated steam was used as a heat carrier in this solution. The concept related to fluidized-bed furnaces and allowed for limitation of waste caused by low-temperature processing.

This study presents the proposal of thermal processing of fuel using the cyclone furnace.

2 Cyclone furnace

Cyclone furnaces are understood to mean energy equipment where a strong eddy flow of gases is used to support the process of solid, liquid or gaseous fuel combustion [12, 13]. Cyclone furnaces also allow for solid fuel gasification in the atmosphere of flue gases or water vapour [14-18] and vitrification of fly ashes [19, 20]. Apart from these function, cyclone furnaces can be used for thermal processing of fuel that consists in its

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drying and controlled devolatilization to the expected level. In this case one can perform the controlled process of fuel heating that allows for heating coal grains to the required temperature while releasing a certain amount of volatile parts and removing mercury in the form of vapour. Details of the assumptions of low-temperature fuel processing for the needs of mercury removal have been discussed in studies [8-10].

The cyclone furnace used in the experimental and numerical examinations (Fig. 1) is composed of two chambers. Fragmented fuel is supplied by tangentially designed nozzles to the upper chamber PC2. The process of heating, drying and initial fuel devolatilization is performed in this chamber. The PC1 chamber is located below the upper chamber and used to continue the process of heating, fuel devolatilization, combustion and gasification. In order to ensure undisturbed flow of fuel from the PC2 to PC1 chamber, a part in the form of the plunger that separates the flow of fuel grains from PC2 to PC1 from the flow of gas from PC1 to PC2 was installed. The PC1 chamber is made of several cylinders with decreasing diameters that form the steps at the places of changes in PC1 chamber diameter. A set of nozzles was designed tangentially to the cylinder generatrix of the PC1 chamber in order to introduce the driving gas at high speed. The aim of the tangentially supplied driving gas is firstly to generate a strong eddy motion of fuel grains and to ensure adequate composition (air, oxygen, carbon dioxide, water vapour) due to performance of the combustion process, gasification and thermal fuel processing aimed to control heating, fuel devolatilization and mercury removal. With PC1 chamber diameter that decreases with the vertical flow of coal grains, the centrifugal force that allows for maintaining a substantial fuel mass in strong eddy current increases while time of remaining the grains necessary to heat the fuel to the required temperature increases (thermal processing of the fuel).

This concept of the cyclone furnace allows for the controlled heating and devolatilization of the fuel swirling in the furnace. The heat needed for the process of heating and devolatilization is obtained from combustion of a certain part of gases generated during fuel devolatilization. The control of thermal processing of fuel can be performed by:

- changing the stream of fuel mass supplied to the cyclone furnace at invariable parameters of driving gas: possibility to achieve the conditions of fuel combustion in the furnace, gasification and controlled heating and fuel devolatilization;
- change in the speed of the driving gas (PC1 chamber): changes in the centrifugal force (control of time of fuel remaining in the furnace) and changes in the amount of heat generated from oxidation of volatile matter;
- change in the composition of the driving gas (PC1 chamber): possibility to control temperature inside the PC1 chamber by changing the stream of oxygen needed for oxidation of volatile matter (change in the amount of heat generated).

The above presented methods allow for determination of optimal process parameters that ensure the required conditions of thermal processing of fuel due to the expected level of mercury removal.

During the experimental examinations, the process of thermal processing of fuel was controlled by changing the stream of fuel mass over a wide range from combustion conditions through gasification to the state when stable thermal fuel processing conditions are obtained: partial fuel devolatilization that ensures the heat for maintaining the process of fuel heating to the assumed temperature.

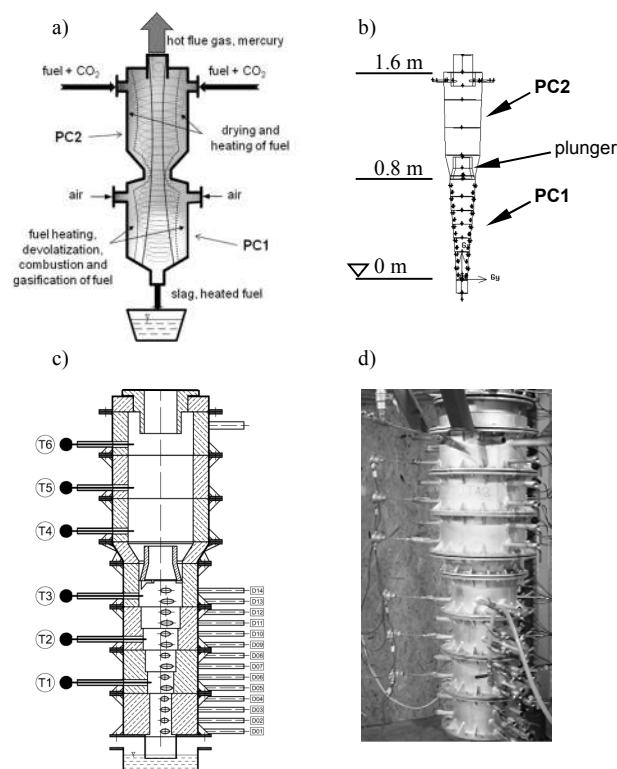


Fig. 1. Design a), geometry b), cross-section c), view of cyclone furnace d).

3 Experimental examinations and numerical analysis of thermal processing of fuel in a cyclone furnace

The below presented results of numerical computations and experimental examinations were performed for brown coal with physicochemical parameters as presented in Table. 1. The examinations were conducted for fuel with medium grain size (1.5 mm). The fuel was supplied to the PC2 chamber (upper) with two tangent channels in the CO₂ stream at the speed of 3 m/s and temperature of 25°C. The stream of fuel supplied was changed from 2 to 16 g/s. Air with temperature of 25°C was supplied to the PC1 chamber at the speed of 3 m/s, being the driving gas used to induce strong eddy current of fuel in the PC1 chamber. For the fuel stream of 2 g/s, air-fuel ratio is $\lambda=1.2$ and ensures complete fuel combustion.

For the above presented process conditions, numerical analysis of the combustion process and fuel devolatilization were performed for variable stream of fuel mass.

Table 1. Physicochemical fuel parameters (all the data in the analytical state).

M	A	VM	HHV	C	H	N	S	Hg
[%]	[%]	[%]	[kJ/kg]	[%]	[%]	[%]	[%]	[ng/g]
25.3	40.2	38.8	13112	33.2	4.4	0.6	2.5	180.7

A - ash

M - moisture

VM - volatile matter

HHV - higher heating value

Geometry and the grid used for computation of the cyclone furnace was developed using the Gambit software (Fig. 1b). Computation of the fuel grain flow, devolatilization process and combustion of volatile matter was performed by means of the ANSYS FLUENT 14 software. Calculations of fuel grain and gas flow were performed using the Reynolds Stress turbulence model. Modelling of flow of coal grains was based on the Discrete Phase Model, whereas coal dust combustion was performed using Species Transport model, which allows for modelling chemical reactions both in the solid phase and gaseous phase [21-25]. The Single-Rate fuel devolatilization model was employed. Calculations were based on the radiation model termed Discrete Ordinate (DO). A numerical model of the process allows for computation of combustion, fuel devolatilization and fuel gasification in the atmosphere of air, O₂, CO₂ and water vapour.

Calculations of the process of heating, devolatilization, gasification and combustion of fuel with replacement diameter of 1.5 mm were carried out for the fuel with physicochemical parameters as presented in Table 1. For simplification purposes, it was adopted that the fuel does not contain sulphur. Reaction rate constants were derived from the studies [22, 23].

Calculations for combustion and gasification of coal dust were described with the following six reactions:

- Reaction of volatile matter (VM) combustion

$$VM = C_{0.13}H_{2.93}O_{1.55}N_{0.0422}$$

$$VM + 0.02 O_2 = 0.13 CO + 1.46 H_2O + 0.0211 N_2 \quad (1)$$
- Reaction of oxidation of carbon oxide

$$CO + 0.5 O_2 = CO_2 \quad (2)$$
- Reaction of oxidation of fixed carbon (FC)

$$C_{(s)} + 0.5 O_2 = CO \quad (3)$$
- Boudouard's reaction

$$C_{(s)} + CO_2 = 2 CO \quad (4)$$
- Synthesis of water gas

$$C_{(s)} + H_2O = CO + H_2 \quad (5)$$
- Hydrogen oxidation reaction

$$H_2 + 0.5 O_2 = H_2O \quad (6)$$

The numerical model takes into consideration the heat transfer through furnace walls to its surroundings.

Actual thickness of individual components of the laboratory furnace (Fig. 1d) [26, 27] with coefficients of heat penetration and conduction was also taken into

consideration for the materials used. It was adopted that air temperature around the furnace was 25°C.

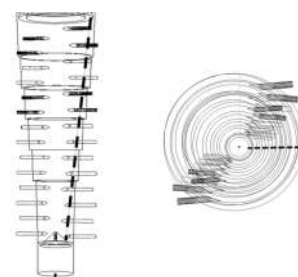


Fig. 2. Location of the line used to determine profiles of temperature, fuel concentration and fuel devolatilization rate.

Figures 3, 4 and 5 present the time-averaged profiles of temperature, fuel concentration and fuel devolatilization rate for selected streams of fuel mass supplied to the cyclone furnace (from 2 to 16 g/s). Profiles (Fig. 3, 4, 5) were determined along the dashed line visible in Fig. 2. It is located at the distance from 10 to 25 mm from the PC1 chamber. Figures 6, 7 and 8 presented time-averaged distributions of temperature, fuel concentration and fuel devolatilization rate for the three selected fuel streams: 2, 6 and 10 g/s. Analysis of the temperature profiles presented in Fig. 3 reveals that for the fuel stream of 2 g/s, temperature at the level of $z = 0.3$ m reaches the level of 1300°C, which suggests the intensive fuel combustion in this zone of the PC1 chamber. This fact is confirmed by profiles of fuel concentration and fuel devolatilization rate (Figs. 4, 5) for the analysed case (2 g/s) and temperature distribution (Fig. 6a). While increasing the stream of the fuel supplied to the cyclone furnace, a temperature decline (Fig. 3 and 6) is observed, connected with substoichiometric conditions of the process and necessity to supply greater amount of heat to the heating and devolatilization process. Increasing the stream of the fuel supplied is noticeable in concentration profiles (Fig. 4) and concentration distributions (Fig. 7) An increase in fuel concentration can be observed near the PC1 chamber walls (Fig. 7b and 7c) compared to the case of reference (Fig. 7a, 2 g/s).

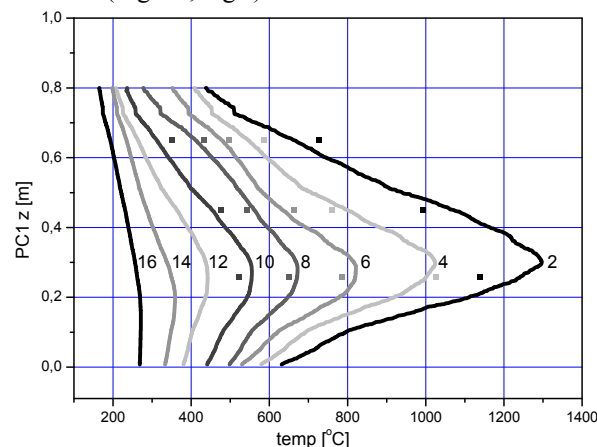


Fig. 3. Time-averaged temperature profiles near the PC1 chamber, points - results of experimental studies (T1, T2, T3).

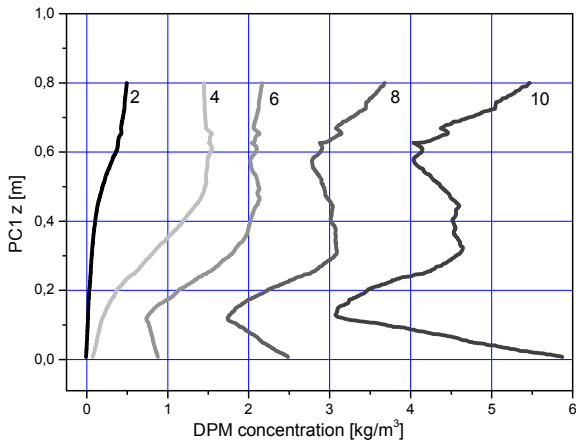


Fig. 4. Time-averaged profiles of fuel concentration near PC1 chamber walls.

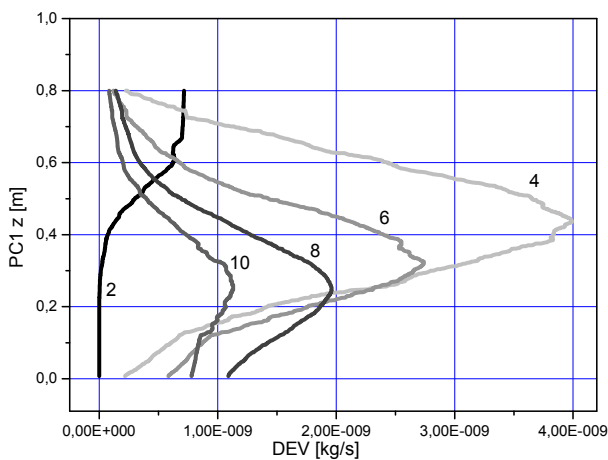


Fig. 5. Time-averaged profiles of fuel devolatilization rate near PC1 chamber walls.

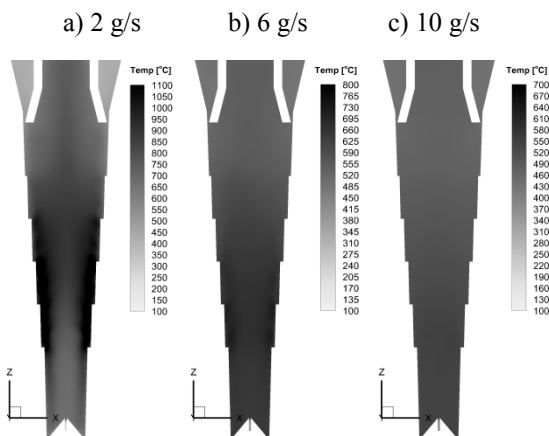


Fig. 6. Time-averaged temperature distributions in the vertical cross-section of the PC1 chamber.

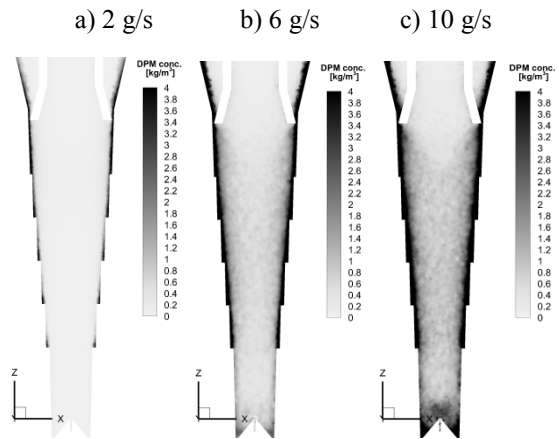


Fig. 7. Time-averaged fuel concentration distributions in the vertical cross-section of the PC1 chamber.

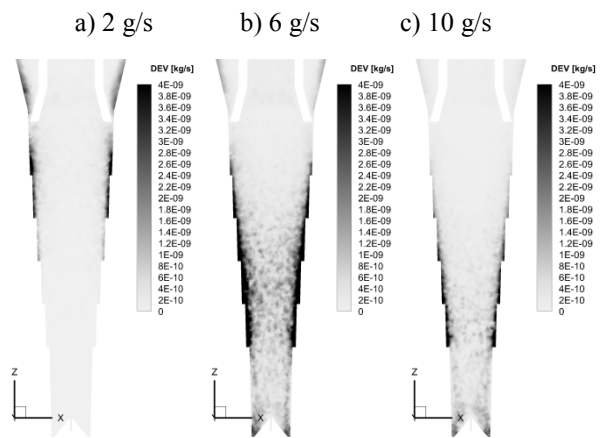


Fig. 8. Time-averaged fuel devolatilization rate distributions in the vertical cross-section of the PC1 chamber.

The increase in the fuel mass stream also requires greater energy (momentum) generated by the driving gas in order to maintain a strong eddy motion of the fuel. If the swirling fuel mass exceeds the boundary value of the driving gas stream, further stable swirling is impossible and fuel flows towards the lower part of the furnaces, which is reflected by the increased fuel concentration near the coordinate $z = 0$ m where the container that allows for collecting the heated and thermally processed fuel is located. The increase in the stream of the supplied fuel impacts significantly on its devolatilization rate (Fig. 5, 8). Temperature (Fig. 3, 6) and devolatilization rate (Fig. 5, 8) are closely related since the increase in temperature leads to the increase in devolatilization rate and therefore allows for generation of an adequate amount of volatile parts whose combustion allows for maintaining adequate temperature inside the PC1 chamber. The increase in volatile matter in the analysed cases that results from the increased devolatilization rate (Figs. 5, 8) and increased stream of volatile matter does not translate into increased temperature (Figs. 3, 6) due to maintaining constant stream of driving gas (oxidant). The increase in fuel stream requires increased heat stream for the purposes of heating the increased fuel mass.

In conclusion of the presented results of the numerical computations, maintaining constant stream of driving gas (oxidant) and variable fuel stream can ensure the conditions that allow for the controlled heating and fuel devolatilization for the fuel flowing to the container located in the lower part of the PC1 chamber.

Fig. 9a presents the instantaneous image of the fuel combustion process (2 g/s) in the PC1 chamber, Fig. 9b presents the interior of the PC1 chamber covered with the molten slag after the fuel combustion process. Fig. 9b illustrates the location of the thermocouples and the internal plunger that divides PC1 and PC2 chambers.

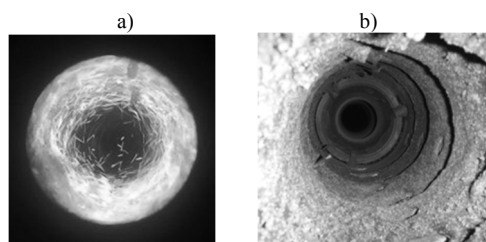


Fig. 9. The view of the fuel burnt in the PC1 chamber (2 g/s) a), interior of the PC1 chamber covered with molten slag after the combustion process b).

Analysis of the combustion and fuel devolatilization process was also performed using the cyclone furnace model (Figs. 1c, 1d) for five fuel rates (2, 4, 6, 8, 10 g/s). Experimental conditions were consistent with the conditions of numerical computations. The obtained results of the experimental examinations allowed for determination of mean values of temperature T1, T2 and T3 (Fig. 1.c) along the height of the PC1 and were added to the Figure 3 in the form of points. Comparison of the temperature distributions obtained from numerical computations and experimental examinations revealed that maximal differences between the values did not exceed 100°C. The experimental examinations for variable stream of fuel mass allowed for determination of the value of temperature to which fuel can be heated for the fuel mass stream. Table 2 presents mean values of fuel temperature which, after thermal processing in the cyclone furnace was supplied to the container located in the lower part of the furnace.

Table 2. Fuel temperature after thermal processing in the cyclone furnace.

Fuel flow rate [kg/s]	2	4	6	8	10
Fuel temperature [°C]	-	612	363	292	255

4 Laboratory examinations of the process of mercury removal in rotary furnace station

The laboratory examinations of the process of mercury removal from brown coal were conducted on a rotary furnace station (Fig. 10).

Fig. 10. Rotary furnace station.

The examinations were performed for the temperatures of 200-400°C. These temperatures were chosen because the temperature of 200°C allows for removal of a substantial amount of mercury from brown coal [10]. Furthermore, maximal temperature was determined at the level which guarantees removal of over 90% of mercury from coal but it is low enough not to cause a substantial degradation of brown coal as a fuel. The examinations of the rotary furnace station were performed in the air atmosphere. Time of remaining the samples in the furnace for individual temperature was chosen so that the sample temperature at the outlet from the furnace was 200, 250, 300, 350 and 400°C for individual tests. After obtaining the required temperature by the samples leaving the rotary furnace, they were cooled in the atmosphere of nitrogen and next the examinations of mercury content were performed in the coal samples. The difference between the content of mercury in the samples before and after thermal processing allowed for computation of the amount of mercury removed for individual temperatures. Mercury content was evaluated using the Lumex RA-915+ spectrometer with the RP-91 device. The methodology of measurements of mercury content in solids using the Lumen spectrometer has been extensively discussed in previous publications [3].

The results indicated that it is possible to remove around 18% of mercury from brown coal already at the temperature of 200°C whereas at the temperature of 250°C, this efficiency reached over 50%. In the case of higher temperatures (300 and 350°C), degree of mercury removal was 78% and over 90%, respectively. With maximal temperature of 400°C, mercury removal degree was 98% (Fig. 11).

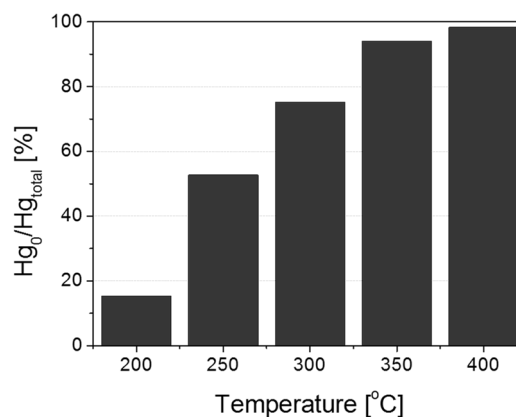


Fig. 11. Amount of mercury removed at individual temperatures for brown coals in rotary furnace station.

5 Examinations of mercury content in coal samples

After completion of the examinations in the rotary furnace station, the temperature ranges were determined for the cyclone furnace. Also in this case, the temperature reached by individual coal samples leaving the cyclone furnace station was significant (Table 2). Mercury content was also evaluated for coal brown samples using the Lumex spectrometer. The results are presented in Fig. 12.

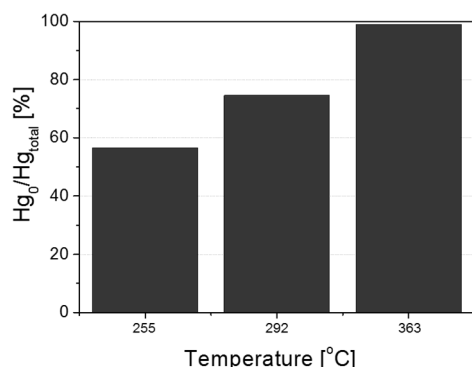


Fig. 12. Amount of mercury removed at individual temperatures for brown coals in cyclone furnace station.

In the cyclone furnace, the amount of mercury removed was nearly 60% for the initial temperature of 255°C (10 g/s). For higher temperatures of 292 (8 g/s) and 363°C (6 g/s), degree of mercury removal was very high i.e. over 75%, and nearly 99%, respectively.

In addition to the degree of mercury removal, fuel degradation degree which occurs at a specific process temperature is also important. Degree of fuel degradation was computed as a difference in the content of volatile matter before and after thermal processing. For the examinations performed on the rotary furnace station, the loss of volatile matter is presented in Fig. 13. For the lowest temperature (200°C), the amount of lost volatile matter was 15%, whereas for the temperature of 250°C, this was slightly over 15%. For the temperature of 300 and 350°C, the amount of lost volatile parts was 20 and 27%, respectively. For the highest temperature (400°C) the amount of lost volatile matter was over 32%.

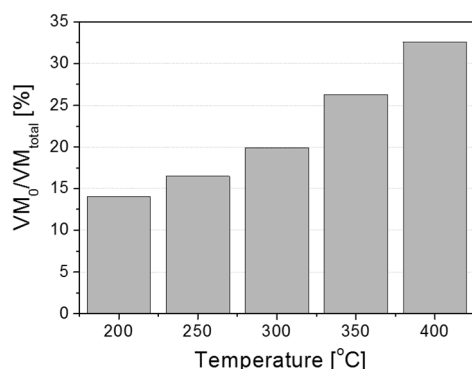


Fig. 13. Amount of volatile matter lost at individual temperatures for brown coals in rotary furnace station.

Similar examinations of the amount of lost volatile matter was performed for the cyclone furnace station. The results are presented in Fig. 14.

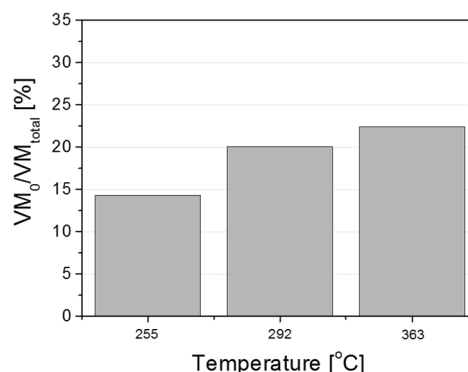


Fig. 14. Amount of volatile matter lost at individual temperatures for brown coals in the cyclone furnace station.

For the cyclone furnace, the amount of lost volatile matter was insignificantly lower than for the rotary furnace station. This is likely to have been caused by more efficient processes of heat transfer inside the cyclone furnace, which translated into faster reaching the expected temperature by the sample, which reduced the loss of volatile matter. For the initial temperature (255°C) (10 g/s) the amount of lost volatile matter was over 14%. For higher temperatures of 292 (8 g/s) and 363°C (6 g/s), this was 20% and 22%, respectively.

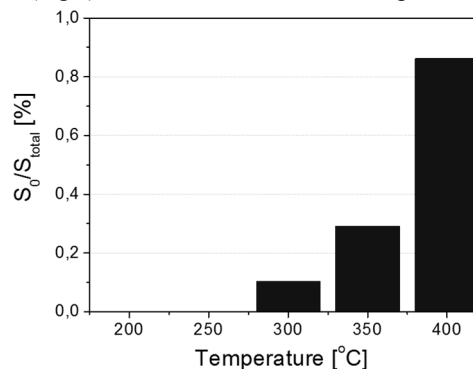


Fig. 15. Changes in sulphur content at individual temperatures for brown coals in rotary furnace station.

Apart from the evaluation of lost volatile matter, the change in the sulphur content in the brown coal was also examined for individual temperatures. Sulphur content was lower for the temperature range of 300-350°C both for rotary furnace and cyclone furnace. The results for the rotary furnace station are presented in Fig. 15. For initial temperatures of 200 and 250°C, no changes in sulphur content was found for the samples after thermal processing. A noticeable reduction in sulphur content was observed in brown coal samples for higher temperatures of 300, 350 and 400°C. Sulphur content for the highest temperature (400°C) was reduced by over 0.8%. Similar results were observed for coal subjected to thermal processing in the cyclone furnace. For these samples, changes in sulphur content (0.1%) were observed only for the temperatures of over 292°C (8 g/s). For higher temperature of 363°C (6 g/s), this was around 0.2% (Fig. 16).

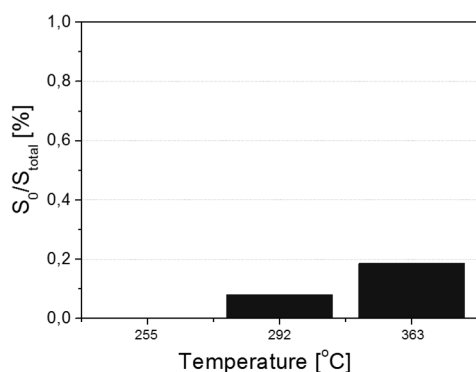


Fig. 16. Changes in sulphur content at individual temperatures for brown coals in cyclone furnace station.

6 Conclusions

The solution for the process of fuel thermal processing using cyclone furnace allows for controlled thermal processing of fuel and mercury removal from the fuel.

With strong eddy motion of fuel, cyclone furnaces allow for intensive mixing and heating of the fuel, thus allowing for high efficiency of thermal processing. Consequently, the cyclone furnace can be scaled to the dimensions that allow for thermal processing of fuel for the purposes of actual energy furnaces. The level of removed mercury can be controlled by changing the stream of fuel supplied to the furnace, changing the aerodynamics (velocity of the driving gas) or oxygen concentration in the driving gas. These parameters translate into the value of temperature inside the cyclone furnace and time of remaining (thermal processing) of fuel, which results in heating the fuel to the desired temperature that ensures the required level of the removed mercury.

The gases obtained during thermal processing, composed of the flue gas, volatile matter and water vapour and containing mercury vapour after cooling and condensation of humidity can be supplied to the absorber that allows for removing mercury vapour from gases. Cleaned gases can be next supplied to the furnace in order to be burnt. This technology of thermal processing of fuel and mercury removal does not lead to chemical energy loss and reduced performance of power units.

The results of the experiments conducted on the cyclone furnace station for the temperatures of 255-363°C showed that the amount of Hg was from nearly 60% to 99%. These results are similar to those obtained for the rotary furnace station, which demonstrated the opportunities for reducing mercury emissions from 50 to over 90% in the temperature range of 250-350°C. The amount of lost volatile matter for brown coal after processing on the cyclone furnace ranged from 15% (for temperature of 255°C) to 22% (for temperature of 363°C). Similar results were obtained for the rotary furnace station, with values ranging from 17% (temperature of 250°C) to 27% (temperature of 350°C). Thermal processing of coal also resulted in a reduction of sulphur content, which in the case of the cyclone furnace was from 0.1% to 0.2% for temperatures of 292 and 363°C. These results were confirmed in the rotary

furnace, with sulphur content for these temperatures reducing from 0.1% to 0.3% for the temperatures of 300 and 350°C.

The results obtained for mercury content after thermal processing in the cyclone furnace lead to the conclusion that it is possible to remove substantial amounts of mercury already at the temperature of ca. 300°C (75%), with the loss of volatile matter at the level not exceeding 20%.

Acknowledgements

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