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The release of mercury from polish coals during thermal treatment of fuels in a fluidized bed reactor

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ABSTRACT

The paper deals with mercury release during thermal treatment of Polish coals. Three coal types (two hard coals and a lignite) from Polish coal mines supplying fuel for the power generation industry were used in the experiment. The release of mercury was investigated in a fluidized bed reactor heated to moderate temperatures 170 °C–410 °C. The thermal treatment of mercury was conducted in various atmospheres (air, nitrogen and carbon dioxide). Except for mercury concentration in the flue gas the emission of CO, CO_2, and O_2 was also determined.

The experimental results indicated that the mercury release from hard coals strongly depended on the gas atmosphere. The highest mercury release was determined in CO_2 atmosphere, while the Hg release in air or nitrogen gas was significantly lower. In the case of lignite no effect of gas type on the mercury release was stated. The results of the investigation also indicated that regardless of the coal type and the process temperature the loss of chemical energy of the coal samples, calculated from the emission of CO and CO_2 was less than 5%.

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1. Introduction

As indicated in numerous papers [e.g. 1–4] mercury is a toxic element that negatively affects the environment and all living organisms including human beings. The total atmospheric emission of mercury is roughly estimated at over 5000 ton per year and roughly 50% of that amount is of anthropogenic origin [2,5,6]. The largest source of mercury emission in the world is hard coal and lignite fired power plants. This is mainly due to the amount of coal burned each year and quite significant concentration of Hg in coals (roughly 0.1—15 mg/g [7–9]). Since Poland burns quite significant amount of coal for power generation (roughly 96.5 mln tons in 2011 [10]) the issue of Hg emission in Poland has been carefully investigated. The research carried out by Wichliński and others [11,12] indicated that the mercury content in Polish hard coals is moderate, between 12 ng/g and 180 ng/g, while mercury content in lignite is somewhat higher roughly 50–230 ng/g. The results are similar to those obtained by other authors (e.g. Wisz and Wojnar [8], or Bojarska [13]).

So far there are two main ways to reduce the emission of mercury from coal-fired facilities. The first one, currently more commonly applied, is focused on postcombustion activities mainly associated with the removal of mercury from the flue gases. The mercury is, for example, removed in after-treatment devices such as electrostatic precipitator, fabric filters, wet flue gas desulphurization plants, etc. According to the U.S. EPA the efficiency of mercury removal with those methods may range from 10% to 90% depending on the type of equipment and the conditions under which combustion is carried out [14]. The mercury can be also removed by the injection of activated carbon, often impregnated with iodine or sulfur, into the exhaust stream before the electrostatic precipitator. This method allows to get rid of up to 90% of mercury [14], but the problem is the amount of active carbon that must be injected into the exhaust stream. The results reported by Sloss [14] indicated that a significant amount of carbon must be injected into the flue gas to capture a moderate amount of mercury. Similar results are also published in ref. [15] where the authors argue that almost 20 t of active carbon must be injected to capture 1 kg of Hg. Although the majority of the carbon is then captured by ESP device the process brings about significant contamination of the fly ash with the unburned carbon and is not welcomed due to significant difficulties with the use of the fly ash for commercial purposes [16].

The other way of mercury removal are precombustion methods, i.e. activities focused on mercury removal from the fuel before it is combusted. Those methods are mainly associated with thermal treatment and heating of the fuel to temperatures high enough to release moisture and mercury but at the same time low enough to prevent the devolatilization and significant decrease of the fuel heating value (HHV or LHV). Apart from the technical relative ‘simplicity’ the advantage of the precombustion methods is much higher mercury removal efficiencies, up to 95%, and much smaller volumes of the mercury-containing exhaust gases for eventual further treatment. Furthermore, the concentration of mercury vapor in the off-gases is higher thus making the cleaning process more efficient. Apart from the above, an advantage of such thermal treatment of the fuel is also the production of

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'clean' fly ash in the combustion facility i.e. the ash that is not contaminated with mercury compounds or activated carbon.

One of the precombustion methods to remove mercury from coal is the so-called mild pyrolysis [17] that involves slow heating of the sample in the absence of air up to 600 °C. Under such conditions coal undergoes partial decomposition and a part of some coal component, like mercury or bromine, are evaporated. The process is affected by many factors, e.g. the type of coal, particle size, temperature, coal residence time in the reactor, or the heating rate. The author [17] reported that with the increase in temperature more mercury was removed until a maximum at roughly 400 °C was reached; the further increase of the temperature decreased the amount of the mercury removed. The process was also affected by the sample residence time; 2 minutes were enough to remove roughly 55% of the mercury in coal, but the increase of the residence time up to 6 minutes brought about the removal of almost 80% of Hg. The thermal treatment of the fuel brought about, however, the loss of some hydrocarbon volatiles; the amount of gas losses for the residence time of 4 minutes and temperature of 400 °C were roughly 6.3%. The studies on mercury removal during pyrolysis were also conducted by e.g. Iwashita [18] who studied the removal efficiency of mercury from coals at various temperatures and reported that the amount of mercury removed at 200 °C was relatively low, roughly 20%, while it increased to almost 90% in the case the coal samples were treated at 400–600 °C. The temperature of the treatment in Iwashita's experiments depended on coal type [18]. Similar results were also obtained by Okhi [19], who investigated the release of mercury from coal during pyrolysis at 150–300 °C and reported the mercury removal efficiency of over 80%.

Numerous researches were also conducted by the Western Research Institute, WRI [20]. The authors stressed high efficiency of mercury removal from coals and reported that they could get rid of roughly 70–80% of the mercury after coal thermal treatment at quite low temperatures, 150–290 °C (the remaining 20–30% of mercury was then released after heating the samples to 593 °C). For their experiments the authors [20] used a horizontal reactor that consisted of two zones. In the first one the coal was heated to 150 °C in order to remove moisture and then transported to another zone where the mercury was removed at 290 °C and the residence time of 12 minutes. The evaporated mercury was then carried away by a carrier gas. Although quite interesting, their process was complicated and not very efficient since it required long fuel residence time (over 4 minutes) and the mercury removal efficiency did not exceed 80%. The authors also conducted similar studies in a fluidized bed reactor and reported for their coals much more promising results which they explained by excellent heat transfer and mixing in the fluidized bed. In their tests the mercury release started just above 150 °C and almost 70% of mercury was removed at 230 °C. The maximum amount of the removed mercury was 79% and was reported for the experiments conducted at 315 °C.

Since no papers have been published so far, where the authors would investigate the efficiency of mercury removal from Polish coals due to their thermal treatment in a fluidized bed reactor, the intention of this paper is to provide some experimental data in that field. Particular attention is put to investigate the effect of temperature and gas atmosphere on the efficiency of mercury removal.

2. Research methodology

2.1. Coal data

For the present studies three coal samples were selected. Two of them, marked 'A' and 'B' were hard coals, while the third one, 'C' was lignite. The results of the coal analysis are shown in Table 1.

Table 1
Some main parameters of the coal samples used in the experiments (air dry).

	Unit	Coal A	Coal B	Coal C
Transient moisture	%	8.4	6.1	10.3
Air-dry (hygroscopic) moisture	%	3.1	7.1	15.9
Ash	%	24.8	24.8	5.6
Volatile matter	%	26.2	20.2	39.6
Fixed carbon	%	45.6	47.2	38.7
HHV	kJ/kg	26,246	24,800	21,522
LHV	kJ/kg	25,295	23,794	20,181
Sulfur	%	1.04	0.58	0.5
Carbon	%	51.5	53.0	53.3
Mercury	ng/g	131	79	231

2.2. Experimental facility

The experiments were conducted in a quartz tube fluidized bed reactor shown in Fig. 1. The tube internal diameter is 25 mm. The bed solids is composed of 40 g of quartz sand (particle size 250–500 μm), and 10 g of ceramic balls of particle size 1500 μm. For the experiments three gases, i.e. air, nitrogen, or carbon dioxide, were used as the fluidizing medium. Regardless of the type of the fluidizing gas, the gas flow rate was maintained constant at 0.00017 m^3/s. The temperature in the column was measured by a K-type thermocouple. The temperature in the column was controlled by adjusting the power of the electrical heaters surrounding the column. For each test 0.5 g of coal of particle size between 500 and 1000 μm was introduced into the bed.

After the coal was fed into the fluidized bed the mercury compounds in the sample decomposed and evaporated. The gas composition was then determined *on-line* by mercury spectrometer LUMEX RA-915+ and flue gas analyzer. The details on the mercury spectrometer are published elsewhere [21,22]. After the test was finished the reactor heating was turned off and the whole facility was cooled but the fluidization was still maintained. The remaining coal sample was then removed from the column and placed in a desiccator since it was later used for further analyses and the determination of the remaining mercury content.

2.3. Measurement method

The measurement setup was connected to the computer and thus enabled to measure, visualize and record *on-line* the concentration of mercury, as well as CO, CO_2 and O_2 in the flue gas during thermal treatment of the fuel particles. Example variations of the concentration of some chosen gas components for the case of coal heating and combustion are shown in Fig. 2 [23]. The *on-line* tracking of the mercury concentration

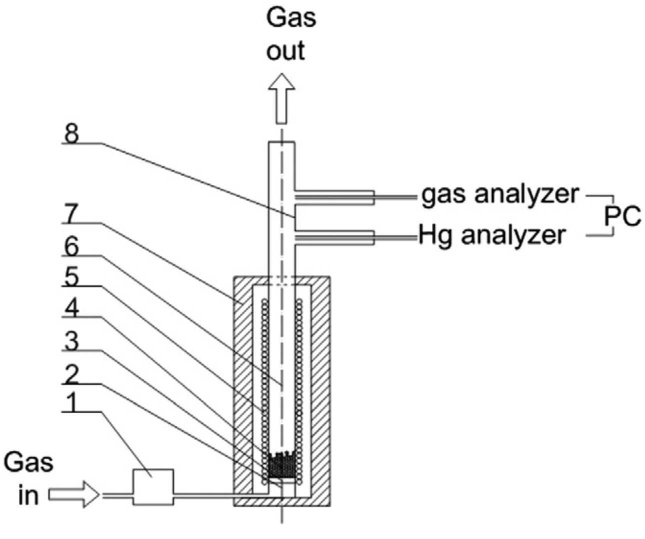

Fig. 1. Sketch of the fluidized bed column: 1—mass/gas flow controller, 2—gas distributor, 3—grate, 4—fluidized bed, 5—electric heater, 6—quartz tube, 7—thermal insulation, 8—measurement zone.

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