

Preliminary Study on The Reduction of Mercury Content in Steam Coal by Using a Pneumatic Vibrating Concentrating Table

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Abstract

Mercury content in hard coal is relatively small. However, considering the large amounts of coal burned in Poland, considerable quantities of mercury are released to the atmosphere. Mercury occurs mainly in pyrite and marcasite included in mineral matter in coal, but it is also present in coal organic matter. Certain, sometimes substantial, amounts of mercury are found in the roof and bottom layers of coal fields, which pass to run-of-mine coal in the mining process. It is possible to remove impurities with gangue coming from these layers and impurities originating from stone inserts in coal fields. It is also possible to remove the liberated iron sulphide particles. But, as practice shows, wet enrichment methods (jigging, flotation) are often imprecise. The concentrates conta-in certain amounts of gangue and sulphur compounds which results in the passing mercury compounds to commercial products. The Katowice Branch if the Institute of Mechanized Construction and Rock Mining has been conducting research on method for deshaling of dry run-of-mine for several years. In the study a system equipped with a pneumatic vibrating concentrating table was used. The system, when properly adjusted, allows effective removal of these contaminants. It is therefore possible to deshale the excavated material and also to remove mercury compounds occurring in the high density fractions. This article discusses the sources and emissions of mercury, its content in hard coal and legal considerations regarding mercury emissions. Also presented are the preliminary results of research on mercury removal using a pneumatic vibrating concentrating table.

Keywords: steam coal, dry separation, removing mercury from raw coal, pneumatic vibrating concentrating table

Introduction

The goal of the study undertaken by the Institute of Mechanised Construction and Rock Mining was to investigate the possibility of removing mercury from run-of-mine steam coal and from commercial products obtained using traditional (wet) coal preparation methods: in this study a pneumatic vibration concentrating table available in the Institute was used. The studies conducted thus far by using dry separation of coal products allow us to argue that, in addition to removing stone and ash, it is possible to reduce the level of mercury occurring in mineral components present in the ceiling and floor layers of coal seams and in coal intergrowths.

Area description

Sources of mercury emissions

Mercury is released into the environment from natural and anthropogenic sources. Annually, approximately 6,200 Mg of mercury are released from natural sources, whereas about 2,600 Mg originate from anthropogenic sources. The contribution of natural and anthropogenic mercury emissions is shown in Table 1 (Pirrone et al. 2010; Sloss 2015; UNEP 2013).

A decrease in mercury emission is currently observed in Europe and North America. This is due to the introduction of modern flue gas cleaning methods as well as systems for pollutant emission inspection. Poland is one of major mercury emitting countries in the European Union. In 2010–2014 Poland's mercury emissions ranged from 9.6 Mg - 10.3 Mg per year. A breakdown of Poland's mercury emissions by sources is given in Table 2. (KOBIZE 2013–2016).

From the data in Table 2, it can be noted that fuel firing accounts for almost 94% of mercury emissions whereas energy generation alone accounts for 55%. Considering that coal firing is one of major sources of anthropogenic mercury emissions in Poland and in the world, many countries have undertaken programs to curb this emission. This is of particular importance in Poland, where the electricity and heat generating industries use mainly hard coal as fuel.

Mercury content of hard coal

Mercury content of hard coal is relatively low, ranging from tens to hundreds of μ g/kg. Analysis of the investigation results shows that mercury occurs in coal in various forms (Meij & Winkel 2009; Strezov et al. 2010; Yudovich & Ketris 2005; Zhang et al. 2009). Mercury occurs in both mineral matter and organic matter. In mineral matter mercury occurs mainly in pyrite and marcasite. Its content may reach as high as 10,000 μ g/kg (Hower et al. 2008). In organic matter, mercury

Tab. 1. Natural and anthropogenic mercury emissions

| No. | Natural emissions | | Anthropogenic emissions | | | | |
|-----|--|--|---|-----|--|--|--|
| | Source of emission | [%] | Source of emission | [%] | | | |
| 1. | Oceans | Oceans 52 Fossil fuel firing for energy generation | | | | | |
| 2. | Lakes | 2 | Cement | 9 | | | |
| 3. | Forests | 7 | Production of metals (including steel metallurgy) | 18 | | | |
| 4. | Tundra, meadows, savannas, prairies | 9 | Chlorine-alkali industry | 1 | | | |
| 5. | Deserts | 10 | Artistic handicraft and gold production | 37 | | | |
| 6. | Agricultural land | 2 | Waste utilisation | 5 | | | |
| 7. | Mercury release after its accumulation | 4 | Other | 6 | | | |
| 8. | Biomass firing | 13 | | | | | |
| 9. | Volcanoes and geothermal areas | 2 | | | | | |

Tab. 2. Mercury emissions in Poland in 2010-2014

| Source of emission | Emissions [kg] | | | | | | | |
|---|----------------|---------|----------|----------|---------|--|--|--|
| Source of emission | 2010 | 2011 | 2012 | 2013 | 2014 | | | |
| Combustion processes in the energy production and transformation sector | 5,640.4 | 5,588.2 | 5,776.6 | 5,687.2 | 5,210.2 | | | |
| Combustion processes outside industry | 1,782.5 | 1,476.4 | 1,546.5 | 1,112.2 | 1,011.2 | | | |
| Combustion processes in industry | 2,095.4 | 2,271.7 | 2,390.2 | 2,655.3 | 2,817.2 | | | |
| Manufacturing processes | 552.6 | 600.9 | 588.7 | 517.4 | 519.6 | | | |
| Waste utilization | 45.0 | 43.4 | 55.7 | 55.1 | 34.8 | | | |
| Other | 0.1 | 0.1 | 0.1 | 0.1 | 0.1 | | | |
| Total | 10,116.0 | 9,980.7 | 10,357.8 | 10,027.3 | 9,593.1 | | | |

occurs in compounds containing sulphur: mainly bound with thiol groups. (R-SH).

In Poland, depending on the location and type of mined hard coal seams, mercury content ranges from 10 to 800 μ g/kg. Table 3 shows the ranges of mercury content in selected hard coal mines in Poland (Bukowski & Burczyk 2008; Michalska & Białecka 2012; Smoliński 2007).

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In recent years several research projects have been completed, in which mercury content of Polish coals was studied. The most important of them is entitled "Developing a database on mercury content of Polish coal and technical guidelines for its further reduction and determining benchmarks for national mercury emission levels" with the acronym Baza-Hg.

Altogether 179 samples of steam coal from a number of hard coal mines were studied. Analysis of the results confirmed that mercury content of Polish coal is relatively low (Białecka & Pyka 2016).

| Company name | Coal mine | Mercury content range [µg/kg] |
|-------------------------------------|--|----------------------------------|
| | Ruch Jankowice | no data available |
| Polska Grupa Górnicza S.A. | Ruch Chwałowice | no data available |
| KWK ROW | Ruch Marcel | 15-113 |
| | Ruch Rydułtowy | no data available |
| Polska Grupa Górnicza S.A. | Ruch Bielszowice | no data available |
| KWK Ruda | Ruch Halemba | 1-758 |
| KWK Kuud | Ruch Pokój | no data available |
| Polska Grupa Górnicza S.A. | Ruch Piast | 99–126 |
| KWK Piast-Ziemowit | Ruch Ziemowit | 45-169 |
| Polska Grupa Górnicza S.A | KWK Bolesław Śmiały | no data available |
| Polska Grupa Górnicza S.A | KWK Sośnica | 95-130 |
| Katowicki Holding Węglowy S.A. | KWK "Murcki - Staszic" Ruch Staszic | 20-120 |
| Katowicki Holding Węglowy S.A. | KWK "Murcki - Staszic" Ruch Murcki | 20-160 |
| Katowicki Holding Węglowy S.A. | KWK "Mysłowice -Wesoła" Ruch Mysłowice | 68–101 |
| Katowicki Holding Węglowy S.A. | KWK "Mysłowice - Wesoła" Ruch Wesoła | 37-73 |
| Katowicki Holding Węglowy S.A. | KWK "Wieczorek" | 30-50 |
| Katowicki Holding Węglowy S.A. | KWK "Wujek" | 95-260 |
| Jastrzębska Spółka Węglowa S.A. | KWK "Borynia – Zofiówka – Jastrzębie, Ruch Borynia | 73–185 |
| Jastrzębska Spółka Węglowa S.A. | KWK "Borynia – Zofiówka – Jastrzębie, Ruch Zofiówka | 43–173 |
| Jastrzębska Spółka Węglowa S.A. | KWK "Borynia – Zofiówka – Jastrzębie, Ruch Jastrzębie | no data available |
| Jastrzębska Spółka Węglowa S.A. | KWK "Budryk" | 153-202 |
| Jastrzębska Spółka Węglowa S.A. | KWK "Knurów-Szczygłowice" | no data available |
| Jastrzębska Spółka Węglowa S.A. | KWK "Krupiński" | 5-195 |
| Jastrzębska Spółka Węglowa S.A. | KWK "Pniówek" | 90-202 |
| TAURON Wydobycie S.A. | ZG "Janina" | 18-198 |
| TAURON Wydobycie S.A. | ZG "Sobieski" | 24–74 |
| TAURON Wydobycie S.A. | ZG "Nowe Brzeszcze" | 11-518 |
| WĘGLOKOKS | | |
| KRAJ Sp. z o. o. WĘGLOKOKS | KWK "Bobrek" | no data available |
| WĘGLOKOKS | KWK "Piekary" | 51-71 |
| KRAJ Sp. z o. o. | | 51-71 |
| PG Silesia Sp. z o.o. | - | 6-113 |
| Lubelski Węgiel Bogdanka S.A. | - | 18-561 |
| Spółka Restrukturyzacji Kopalń S.A. | KWK "Kazimierz – Juliusz" | 136-159 |
| Spółka Restrukturyzacji Kopalń S.A. | KWK "Anna" | 1-83 |
| Spółka Restrukturyzacji Kopalń S.A. | KWK "Jas-Mos" | 5-123 |
| Spółka Restrukturyzacji Kopalń S.A. | KWK "Murcki-Staszic" Ruch Boże Dary | 75-120 |

Legal conditions pertaining to mercury emissions

In Poland, as in the entire European Union, there are currently no legal regulations or standards concerning mercury content of coal and mercury emissions by coal-fired power stations, however all EU countries maintain a European Pollutant Release and Transfer Register (E - PRTR), whose goal is to keep records of pollutant emissions (including mercury) from industrial plants. Currently the register maintains data from almost 30,000 industrial plants of various types, including those of the power sector (E-PRTR 2014). Data for this register come from the National Pollutant Release and Transfer Registers which all EU member countries are obligated to maintain on the strengths of Regulation (EC) No 166/2006 of the European Parliament and of the Council (Regulation 2006). In the case of mercury and its compounds, to reporting of their emissions are obliged entities whose annual emissions exceed 10 kg. In Poland the register of pollutants, including mercury, is maintained by the National Centre for Emissions Management (KOBiZE).

Although there are no EU standards concerning mercury emissions from coal-fired power stations (such standards are already in force in the Netherlands, USA and Germany has announced that they will soon be introduced). Table 4 specifies permissible levels of mercury emissions from coal-fired power stations in those countries. The USA has also introduced nationwide standards for curbing pollutant emissions, includ-

Tab. 4. Mercury emission standards for coal power plant

| Country | Mercury emission standard [µg/Nm ³ flue gas] | | | | |
|-------------|--|------------|--|--|--|
| | Existing plants | New plants | | | |
| USA | 5.0 | 3.0 | | | |
| Netherlands | 2.8 | 2.8 | | | |
| Germany | 3.0 (planned) | | | | |

Tab. 5. BAT-levels of mercury emission to air from coal combustion

| Coal type | Plant capacity [MW] | Permissible mercury level in flue gas [µg/Nm ³] | | | |
|------------|------------------------|--|-----------------|--|--|
| | | New plants | Existing plants | | |
| Hard coal | < 300 | <3 | <9 | | |
| fiald coal | > 300 | <2 | <4 | | |
| Lignite | < 300 | <5 | <10 | | |
| Liginic | > 300 | <4 | <7 | | |

ing mercury, from coal-fired power stations – MATS (Mercury and Air Toxics Standard). These regulations are binding on all power stations with capacities greater than 25 MW (EPA 2015). Their goal is to reduce current mercury emissions in the USA by 90% (Gołaś & Strugała 2014).

Among legal regulations concerning mercury emissions, an important document is the mercury convention, known as the Minamata Convention, which was adopted in 2013 (Adamska 2014; Chmielarz 2014). The provisions of the Convention were also supported by Poland in 2014. Its main goal is to protect human health and the environment against anthropogenic emissions of mercury and its compounds. The mercury convention regulates the supply and trade of mercury, mercury emissions and release to the environment, products and processes using mercury, mercury-containing waste, mercury storage and contamination of land, legal aspects, financing and technical assistance as well as the exchange of information, research and development.

As a result of the introduction of the mercury convention, the European Union formulated BAT Conclusions on mercury emissions from combustion of solid fuels (mainly hard coal and lignite) in power generation units: this document will be the basis for determining mercury emissions limits. Table 5 shows the BAT-related permissible mercury limits in flue gas of 2016 (Wdowiak & Henc 2016).

Methods for reducing mercury content of coal

Methods for reducing mercury content of coal before its use for energy generation are known as precombustion methods in contrast to postcombustion methods which involve mercury elimination from flue gas or post-process gases (Krzyżyńska et al. 2011, Wdowin et al. 2015).

The efficiency of precombustion methods for mer-

cury elimination depends on the form of its occurrence in a coal deposit. Basic pre-combustion methods for mercury elimination from coal include:

- mechanical preparation of coal,
- preliminary thermal preparation of coal,
- extraction by water in sub-critical conditions,
- chemical treatment of coal,
- dissolution of pyrite with SO₂,
- biological methods,
- the Hyper-Coal method.

Mechanical coal preparation

Hard coal extracted in mines, known as ungraded coal, is composed of grains of various sizes, gangue, coal-gangue intergrowths and coal shale. It is not usable in the original form and must be subjected to preparation. In the case of steam coal, if quality parameters meet the requirements of potential users (net calorific value as well as ash, moisture and sulphur content) it suffices to classify it into narrow grain size grades. In most cases, however, steam coal does not fulfil quality requirements of users. On the other hand, coking coal cannot be used in its raw state. In order to meet users' quality requirements, coal must be subjected to mechanical preparation to remove gangue, shale, part of coal-gangue intergrowths, backfill sand and pyrite. Such processes are called beneficiation. Coal can be beneficiated in aqueous or air medium. Beneficiation processes employ differences in physical properties of coal and mineral matter: these include such properties as actual density or surface properties (Blaschke 2009). In addition to the elimination of components unwanted from the viewpoint of power generation parameters, beneficiation processes also contribute to the reduction of noxious chemical elements, such as mercury and sulphur (Baic et al. 2015a, 2015b, 2015c). The efficiency of mercury reduction differs for various types of coal.

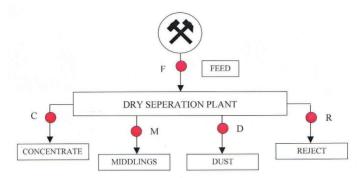


Fig. 1. A simplified flowsheet of pilot plant for dry separation of steam coal (black dots indicate the points of sampling)

Tab. 6.1. Test result of preliminary research on the possibility of removing mercury from raw coal granulation 20-0 mm

| Property | Symbol | Unit | Feed | Concentrate | Middlings | Reject | Dust |
|---------------------|------------------|-------|--------|-------------|-----------|--------|--------|
| Surface moisture | Wex ^r | % | 5.1 | 1.2 | 1.2 | 0.9 | 1.1 |
| Analytical moisture | Wa | % | 4.6 | 5.3 | 4.1 | 1.8 | 4.8 |
| Total moisture | Wtr | % | 9.5 | 6.4 | 5.3 | 2.7 | 5.8 |
| Ash content | Aa | % | 25.0 | 13.4 | 33.3 | 81.3 | 27.8 |
| Heating value | Qs ^a | kJ/kg | 21,979 | 26,431 | 18,530 | 2,566 | 20,942 |
| Calorific value | Qi ^a | kJ/kg | 21,105 | 25,411 | 17,747 | 2,315 | 20,109 |
| Mercury | Hgtd | µg/kg | 113 | 72 | 143 | 319 | 153 |
| Yield | γ | % | 100 | 60 | 27 | 7 | 6 |

Satisfactory results of mercury elimination are produced when pyrite-rich coals are beneficiated. Beneficiation allows us to reduce mercury content by almost 90%, and the efficiency of its removal is proportional to the efficiency of removing mineral matter. Beneficiation is largely ineffective for coals with low pyrite content and the process efficiency is not greater than 10%.

It should be emphasised here that the effectiveness of mercury removal does not depend solely on the pyrite content of coal, but also on the form of this mineral. Pyrite of epigenetic origin, or coarse grained pyrite, can be easily removed by beneficiation in contrast to syngenetic pyrite (fine-grained). Literature data indicate that the best results of pyrite removal can be obtained for epigenetic pyrite occurring in the form of large intrusions irregularly distributed throughout the coal structure preceded by fine grinding processes. (Aleksa et al. 2007).

Wet beneficiation

In Poland, coal is most often beneficiated using wet methods. These methods use differences in densities between mineral and organic matter of coal and represent gravity methods. Most often they are used for run-of mine coal with grain sizes greater than 10 mm or 20 mm, although they are also used for ROM coal with grains greater than 1mm or 3 mm. Coal with fine grains is beneficiated using physicochemical methods which employ differences in surface properties between coal and impurities. An example of such processes is flotation (Blaschke 2009).

In Poland, the most often wet methods include:

- dense media beneficiation,
- beneficiation using jigs,
- flotation beneficiation.

Dry beneficiation

Dry separation methods, also known as dry de-stoning, use no or little water, which favours environmental protection and reduces both capital investment and operating costs (Baic et al. 2015a, 2015b, 2015c). In some countries dry separation is used before coal is subjected to wet separation. Preliminary removal of gangue simplifies the process layout of a preparation plant, reduces the amount of feed and the number of required machines and equipment, reduces the consumption of energy and reduces the cost of wet separation. Additionally, the separated waste products, which had no contact with water, can be successfully used as a substitute for natural aggregate. For dry separation we can use, among others, pneumatic vibrating separation tables. (Baic & Blaschke 2013). A pneumatic vibrating concentrating table consists of a perforated working plate, vibration mechanism, an air chamber, a drive system and a mechanism for adjustment of the table tilt angle and vibration frequency. The raw coal feed is directed onto the plate which is set into vibration. Air for fluidizing the coal bed is supplied from under the table. The flow of air causes separation of the coal bed: low density material (coal) gathers on the bed surface

Tab. 6.1. Test result of preliminary research on the possibility of removing mercury from raw coal granulation 20-0 mm

| Property | Symbol | Unit | Feed | Concentrate | Middlings | Reject |
|---------------------|-----------------|-------|--------|-------------|-----------|--------|
| Surface moisture | Wexr | % | 1.6 | 1.9 | 1.6 | 2.1 |
| Analytical moisture | W ^a | % | 2.0 | 3.4 | 2.9 | 0.9 |
| Total moisture | Wtr | % | 3.6 | 5.2 | 4.5 | 3.0 |
| Ash content | A ^a | % | 50.5 | 8.1 | 24.3 | 86.2 |
| Heating value | Qs ^a | kJ/kg | 11,852 | 29,169 | 20,837 | 0 |
| Calorific value | Qi ^a | kJ/kg | 11,233 | 28,018 | 19,899 | 0 |
| Mercury | Hgtd | µg/kg | 65 | 51 | 90 | 78 |
| Yield | γ | % | 100 | 33.3 | 7.5 | 59.1 |

Tab. 6.3. Test result of preliminary research on the possibility of removing mercury from raw coal granulation 25-6 mm

| Property | Symbol | Unit | Feed | Concentrate | Middlings | Reject |
|---------------------|-----------------|-------|--------|-------------|-----------|--------|
| Surface moisture | Wexr | % | 6.4 | 7.0 | 5.6 | 6.3 |
| Analytical moisture | Wa | % | 9.7 | 9.6 | 9.7 | 8.3 |
| Total moisture | Wtr | % | 15.5 | 15.9 | 14.8 | 14.1 |
| Ash content | Aa | % | 8.4 | 6.1 | 7.6 | 14.8 |
| Heating value | Qs ^a | kJ/kg | 25,893 | 26,624 | 26,096 | 23,802 |
| Calorific value | Qi ^a | kJ/kg | 24,752 | 25,464 | 24,957 | 22,840 |
| Mercury | Hgtd | μg/kg | 88 | 61 | 110 | 162 |
| Yield | γ | % | 100 | 66.5 | 18.5 | 15 |

whereas the high density material (waste) gathers in the bottom part of the bed. As the table is transversally tilted, the low density material present on the bed surface tends to move over the surface under the force of gravity and pass over baffle located at the edge of the table. The material of greater density gathers in the bottom part of the bed and moves towards the waste outlet (Baic & Blaschke 2013). The possibility of reducing the mercury content of coal by using a pneumatic vibrating concentration table was confirmed by American scientists (Honaker 2007): in dry separation of coal the mercury content was decreased by approximately 67%.

Research methodology

Many samples were prepared and examined in the study. These were samples of the feed (run-of-mine coal and commercial products) and samples or products of dry separation (concentrate, middlings, dust and reject) obtained by using the pneumatic vibrating concentrating table. The samples were studied to assess how the dry separation process reduces the mercury content of hard coal. A simplified diagram of the set-up for testing dry separation of hard coal equipped with the FGX-1 vibrating concentrating table is presented in Fig. 1 showing also points of sample collection. All the samples were subjected to proximate and ultimate analysis.

Mercury content of the samples was measured us-

ing an MA-2 mercury analyser manufactured by the Japanese company Nippon Instruments Corporation, following the 7473 EPA Method. The instrument uses a Cold Vapour Atomic Absorption Spectroscopy (CVAAS) in conformity with the currently binding Polish Standard PN-ISO 15237:2007 – Solid Fuels – Determination of total mercury content of coal.

This is a generally used method for determining the mercury content of solid samples, including coal (Lopez-Anton et al. 2012). The analyser has a broad measurement range with a detection limit of $2 \cdot 10-6 \ \mu g$ and the upper detection limit of $1 \ \mu g$ mercury in the sample and has highly linear characteristics (R2 = 0,999).

Results of mercury content of steam coal after dry separation

The goal of the study on dry separation of coal using the vibrating concentrating table was to:

• assess the possibility of removing mercury from steam coal with this method,

• determine distribution of mercury among separation products.

In Tables 6.1 and 6.3 the results of the study are shown.

Conclusion

Preliminary studies have shown that it is possible to remove mercury from run-of-mine steam coal and from its beneficiation products by using dry separation. The use of a vibrating concentrating table enabled us to remove only 30% of the total sulphur contained in the studied steam coal samples. The study has shown that the effectiveness of mercury removal depends on the amount of reject obtained in the dry separation process (the greater the amount of reject, the higher the efficiency) and on the form of mercury in coal (whether it accompanies organic or mineral matter).

In the authors' opinion the effectiveness of mercury removal can be increased by modifying the operating parameters of the vibrating concentrating table and by extending the scope of analyses to take into consideration the form of mercury occurrence in coal.

The final effect of the planned research work will be a proposal of a process set-up for the removal of mercury from steam coal accompanied by a specification of conditions for conducting the process effectively.

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