



Analysis of Mercury Content in Feed Coal and Rejects for Selected Hard Coal Cleaning Processes

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1. Introduction

Mercury is commonly found in hard coal (Ketris & Yudovich 2009) and its combustion is one of the main sources of the anthropogenic mercury emission into the atmosphere (Pacyna et al. 2016). Due to the toxicity of mercury (Li & Tse 2015) a reduction of its emission is the objective of many legislative actions, among others the BAT conclusion for large combustion plants adopted by the European Commission (BAT-LCP 2017). In the case of the large combustion plants, there are well-known and widely used methods allowing for the removal of mercury from flue gases (the post-combustion stage), among others a sorbent injection into the flue gases stream (Wierońska et al. 2018). In the case of households and small-scale combustion installations these methods are not used. This is caused by both lack of technical possibilities as well as high investment costs. The most effective solution for this group of customers is the removal of mercury from coal (the pre-combustion stage), which can be obtained with the use of various methods (Dziok 2018).

The removal of mercury from hard coal can be achieved, among others, through the cleaning process (Baic & Blaschke 2018, Dziok & Strugała 2017, Dziok 2018). This process yields: (i) clean coal with a reduced ash content and an increased calorific value, (ii) middling products that are used in the power industry, and (iii) rejects. The rejects are directed to a landfill or are used as a substitute for the natural aggregates (Baic & Blaschke 2013). Thus, they are not directed to the thermal processes and, therefore, mercury contained in the rejects is not released into the atmosphere during coal combustion.

The washing processes are commonly used in hard coal preparation plants. Recently, the dry separation methods allowing for the efficient deshaling of raw coal have become more popular (Baic et al. 2014, 2014a, 2015, 2015a, Blaschke et al. 2016). The Authors' scientific works confirmed the possibility of mercury removal from coal in these processes, although the effectiveness of mercury removal together with rejects varied from 8 to 96% (Dziok & Strugała 2017, Dziok 2018). This should be explained by the difference in the mode of mercury occurrence in individual coals. In Polish hard coals mercury is mainly found in pyrite (Dziok et al. 2019), and the effectiveness of mercury removal from coal in the washing processes increases along with the growth in the amount of removed pyrite (Dziok et al. 2015). Effective pyrite removal from hard coal, and, consequently, also mercury removal, may be achieved with the use of the pneumatic vibrating separators (Baic & Blaschke 2017, 2017a, 2018, Dziok & Strugała 2017). The aim of the study was to assess the effectiveness of mercury removal from hard coal with the rejects in the washing and dry deshaling processes.

2. Methodology

The coal cleaning processes which are commonly used in Polish coal preparation plants were analyzed, i.e. dense media separation, grain and fine coal jigging, flotation, as well as dry deshaling using the pneumatic vibrating FGX type separator. The first four are the washing processes and the last one is the dry separation process. In the case of the washing processes, samples for analysis were derived from industrial installations located in the Polish hard coal preparation plants. In the case of the dry deshaling process, samples were obtained from a pilot test plant equipped with the pneumatic vibrating FGX type separator with a capacity of 10 Mg/h. The feed coals were supplied by different mines and therefore were characterized by different parameters.

For each of the analyzed processes, the samples of feed coals and rejects were investigated. In all samples the mercury content was determined with the use of the MA-2 analyzer (Nippon Instruments Corporation), based on cold vapor atomic absorption spectroscopy (CVAAS). The measurements were carried out in compliance with the EPA 7473 Method. The obtained results are given in Table 1.

In order to determine the effectiveness of the analyzed processes, the RF factor was used. RF was calculated as the ratio of the mercury content in the rejects ($Hg_{rejects}$) to the mercury content in the feed coal (Hg_{feed}) – Eq. (1). RF values higher than 1 indicate the accumulation of mercury in the rejects and those lower than 1 indicate a lower mercury content in the rejects in comparison to the mercury content in the feed coal.

Table 1. Mercury content in analyzed samples

Case no.	Coal-cleaning process	Particle size [mm]	Mercury content Hg ^d [μg/kg]	
			feed coal	rejects
1	dense media separation	>20	57	62
2			54	62
3			114	249
4			67	116
5	grain coal jigging	0.5-70	148	79
6			83	71
7	fine coal jigging	0.5-20	99	88
8			75	55
9			131	190
10			134	114
11	flotation	<0.5	66	148
12			218	136
13			56	69
14			137	115
15			127	127
16			93	93
17	dry deshaling (pneumatic vibrating FGX type separator)	0-25	113	319
18			146	121
19			88	162
20			131	132
21			65	78
22			124	160

$$RF = \frac{Hg_{rejects}}{Hg_{feed}}, [-] \quad (1)$$

where:

RF – factor of mercury accumulation in the rejects separated from coal in the washing/deshaling processes [-],

$Hg_{rejects}$ – mercury content in the rejects [μg/kg],

Hg_{feed} – mercury content in the feed coal for the coal washing/deshaling processes [μg/kg].

3. Results and discussion

The mercury content in the feed coals for the coal cleaning process varied from 54 to 218 $\mu\text{g}/\text{kg}$ (the average of 106 $\mu\text{g}/\text{kg}$) and was smaller than the mercury content in the rejects separated from coal – from 55 to 319 $\mu\text{g}/\text{kg}$ (the average of 125 $\mu\text{g}/\text{kg}$). For the entire population of analyzed samples a significant correlation between the mercury content in the feed coal and in the rejects was not found (Fig. 1a). Such a relationship was obtained only for the beneficiation in dense media separators (Fig. 1b). The significance of the correlation was verified with the use of the F-Snedecor test at the confidence level of 0.95. The lack of correlation for the entire population may be caused by, both, differences in the mode of mercury occurrence in individual coals (Dziok et al. 2015) as well as by differences in mercury content in the grains of pyrite and other sulfides which occur even within a single grain (Dziok et al. 2019). The results of the preliminary statistical analysis obtained for the dense media separation may indicate that for coarse size fractions of hard coal the dominant mode of mercury occurrence is the adventitious inorganic constituents which undergoes separation very well. However, due to the small number of samples (4 cases), the results obtained should be verified in further investigations.

A comparison of mercury content in the rejects derived from the analyzed processes is shown in Fig. 2. The highest mercury content and the highest dispersion of results were recorded for the rejects derived from the dense media separation as well as from the dry deshaling process. The lowest values were obtained for the grain coal jigging. In the case of the coal jigging, low mercury content may be related to its accumulation in the middling products, which was pointed out in our previous work (Dziok et al. 2019). The mercury content in the analyzed middling products was 313 and 246 $\mu\text{g}/\text{kg}$ (dry basis), respectively. Their examination using an electron probe microanalyzer (EPMA) showed the occurrence of mercury mainly in the grains of pyrite, marcasite as well as chalcopyrite and the mercury content in them reached 0,1%.

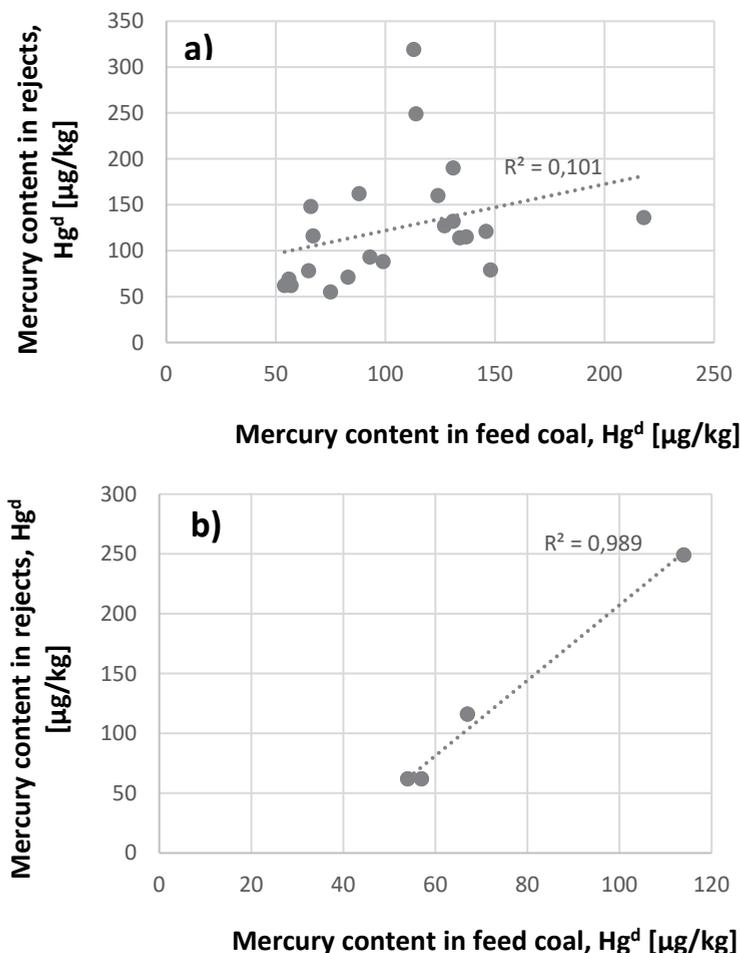


Fig. 1. Relationship between mercury content in feed coal and rejects derived from the coal washing/deshaling processes: a) all analyzed cases, b) dense media separation

In Fig. 3 a comparison of the RF values determined for the analyzed hard coal washing process was presented. The influence of grain size on the mercury content in the rejects is noticeable. Only in the case of beneficiation in dense media separators for each of the analyzed cases the RF was higher than 1 (from 1.09 to 2.18 with the average at the level of 1.54). This shows, as previously mentioned, a very good separation of mercury from the raw coal of large grain size in the coal washing process in the dense media separators (coal grain size >20 mm). In the case of the grain coal jiggling (coal grain size 0.5-70 mm) for

each of the analyzed cases the mercury content in the rejects was lower than in the feed coal. The RF value varied from 0.53 to 0.89 with the average of 0.69. A relatively low value of RF was obtained for the fine coal jigging as well (coal grain size 0.5-20 mm): from 0.85 to 1.45 with the average of 1.10. This may suggest difficulties in the separation of grains rich in mercury during these processes or the lack of such grains in feed coals.

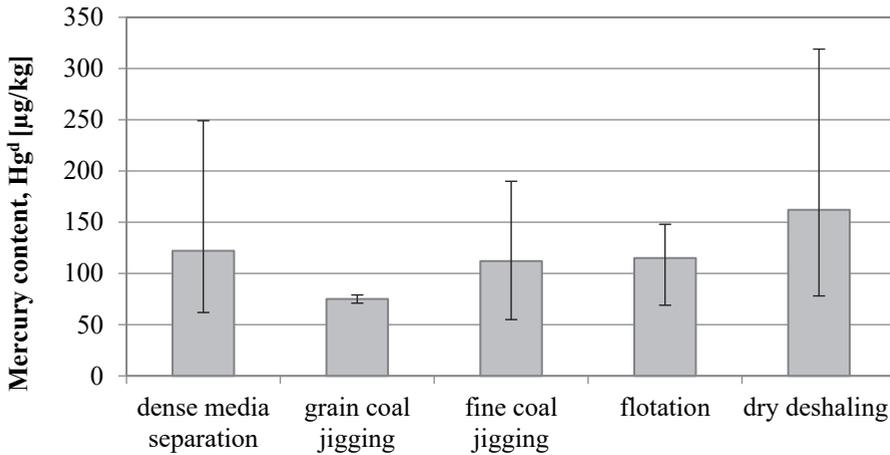


Fig. 2. Comparison of mercury content in the rejects derived from individual cleaning processes (the whiskers represent the dispersion of the obtained results)

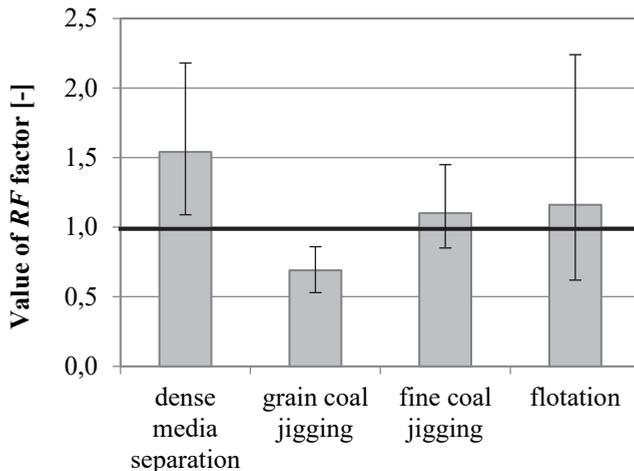


Fig. 3. Comparison of the RF values determined for the analyzed hard coal washing process (the whiskers represent the dispersion of the obtained results)

For the flotation (coal grain size <0.5) the obtained results were not obvious. For individual cases very low or very high values of RF were observed. This can be explained by the different mode of mercury occurrence in the analyzed coals.

In Fig 4 a comparison of the RF values determined for the analyzed cleaning processes was presented. In the light of the obtained results, it can be concluded, that mercury shows a tendency to occur in higher amounts in the rejects when compared to the feed coal both in the washing process (RF from 0.53 to 2.24 with the average of 1.15) and in the deshaling process (RF from 0.83 to 2.82 with the average of 1.50).

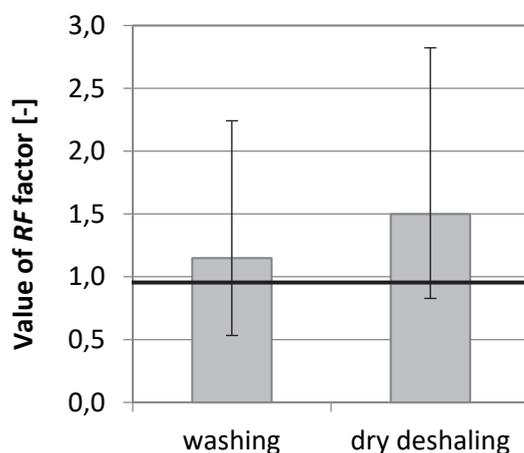


Fig. 4. Comparison of the RF values determined for analyzed hard coal washing and deshaling processes (the whiskers represent the dispersion of the obtained results)

This shows the possibility of the effective removal of mercury occurring in the adventitious inorganic constituents of the analyzed Polish hard coals. However, it should be noted, that the obtained results varied within a relatively high range, which should be explained by the difference in the mode of mercury occurrence in individual coals (Dziok et al. 2015, Zheng et al. 2008). This variability may cause low effectiveness of mercury removal for some coals. For such coals, the solution may be the thermal pretreatment of clean coals derived from the washing and deshaling processes at the temperatures of 200-300°C. This process allows for the removal of mercury occurring both in the organic matter as well as in the inorganic constituents characterized by a relatively low temperature of decomposition (Dziok & Strugała 2017). However it should be noted that the choice of an appropriate solution will depend on: (i) the investment and operating

costs, (ii) on the quality requirements for hard coal including ash content and calorific value, as well as (iii) the technological properties of coal, including the forms of mercury occurrence. When compared to the washing methods, the pneumatic vibrating FGX type separators are characterized by lower investment and operating costs (Baic et al. 2015).

4. Conclusions

The mercury content in the feed coals for coal cleaning varied from 54 to 218 $\mu\text{g}/\text{kg}$ (the average of 106 $\mu\text{g}/\text{kg}$) and was smaller than the mercury content in the rejects separated from coal – from 55 to 319 $\mu\text{g}/\text{kg}$ (the average of 125 $\mu\text{g}/\text{kg}$). The highest mercury content and the highest dispersion of results were recorded for the rejects derived from the dense media separation as well as from dry deshaling using the pneumatic vibrating separators. The lowest values were obtained for the grain coal jigging.

In the light of the obtained results, it can be concluded that mercury shows a tendency to occur in higher amounts in the rejects when compared to the feed coal both in the washing process (RF from 0.53 to 2.24 with the average of 1.15) and in the deshaling process (RF from 0.83 to 2.82 with the average of 1.50), while the higher RF values were obtained for the dry deshaling process. This shows the possibility of effective mercury removal from the Polish hard coals. However, the choice of an appropriate solution will depend on: (i) the investment and operating costs, (ii) the quality requirements for hard coal, including ash content and calorific value as well as (iii) the technological properties of coal, including the forms of mercury occurrence.

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Abstract

Due to the toxicity of mercury a reduction of its emission is the objective of many legislative actions. In the case of power plants, there are well-known methods allowing for the removal of mercury from flue gases (post-combustion). In the case of households and small-scale combustion installations these methods are not used, which is caused by high investment costs. The most effective solution for this group of customers is the removal of mercury from coal (pre-combustion). This can be obtained in the washing and deshaling processes.

The coal-cleaning processes which is commonly used in Polish coal preparation plants were analyzed, i.e. dense media separation, grain and fine coal jigging, flotation, as well as dry deshaling using the pneumatic vibrating FGX type separator. The first four are the washing processes and the last one is the dry separation process. The effectiveness of the coal cleaning process was assessed with the use of the *RF* factor (the ratio of the mercury content in the rejects to the mercury content in the feed coal). The obtained values of the *RF* factors show that mercury has a tendency to remain in the rejects, while higher values of the *RF* factors were obtained for the dry deshaling process (from 0.83 to 2.82 with the average of 1.15) than for the washing process (from 0.53 to 2.24 with the average of 1.50). This shows the possibility of the effective removal of mercury occurring in the adventitious inorganic constituents of the analyzed Polish hard coals. However, it should be noted, that the obtained results varied within a relatively high range, which should be explained by the difference in the mode of mercury occurrence in individual coals.

Keywords:

hard coal, washing, deshaling, mercury, removal

Analiza zawartości rtęci w nadawie i odpadach dla wybranych procesów wzbogacania węgla kamiennego

Streszczenie

Emisja rtęci, z uwagi na jej toksyczne właściwości jest przedmiotem wielu działań legislacyjnych których przykładem jest m.in. przyjęcie w UE konkluzji BAT dla dużych obiektów energetycznego spalania. W przypadku dużych energetycznych instalacji znane i stosowane są różne metody usuwania rtęci ze spalin (etap post-combustion), natomiast w przypadku użytkowników domowych i instalacji energetycznych o małej mocy

te metody nie są stosowane. Jest to spowodowane w głównej mierze wysokimi kosztami inwestycyjnymi. Najskuteczniejszym rozwiązaniem dla tej grupy użytkowników węgla jest usuwanie rtęci z węgla (etap pre-combustion), co umożliwia uzyskanie węgla o niskiej zawartości rtęci. Taki węgiel może być przygotowany w wyniku jego wzbogacania lub odkamieniania.

Analizie poddano procesy wzbogacania węgla kamiennego stosowane w polskim sektorze przerobczym: wzbogacanie w płuczkach zawieszinowych cieczy ciężkich, wzbogacanie w osadzarkach miałowych i ziarnowych, flotację a także proces odkamieniania przy wykorzystaniu separatora powietrzno-wibracyjnego. Cztery pierwsze procesy są stosowane do wzbogacania węgla kamiennego na mokro, a ostatni do suchej separacji. Dla analizowanych procesów przebadano próbki nadaw kierowanych do wzbogacania oraz odpady. Dla oceny efektywności procesu wzbogacania wykorzystano wskaźnik RF, wyznaczony jako stosunek zawartości rtęci w odpadzie do nadawy kierowanej do wzbogacania. Uzyskane wartości RF wskazują na tendencję do pozostawiania rtęci w odpadach zarówno w procesie wzbogacania na mokro (od 0,53 do 2,24 przy średniej 1,15) jak i suchego odkamieniania (od 0,83 do 2,82 przy średniej 1,50), przy czym wyższe wartości wskaźnika uzyskano dla suchego odkamieniania. Świadczy to o możliwości efektywnego usuwania rtęci z badanych polskich węgla kamiennych. Należy zaznaczyć, że uzyskane wyniki wahały się w dość szerokim zakresie, co należy tłumaczyć różnicami w formach występowania rtęci w poszczególnych węglach.

Słowa kluczowe:

węgiel kamienny, wzbogacanie, odkamienianie, rtęć, usuwanie