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**Author:** Zbigniew Mirkowski, Iwona Jelonek

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Petrographic composition of coals and products of coal combustion from the selected combined heat and power plants (CHP) and heating plants in Upper Silesia, Poland

Z. Mirkowski⁎, I. Jelonek

University of Silesia, Faculty of Earth Sciences, 41-200 Sosnowiec, Będzicka 68, Poland

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ABSTRACT

Coal samples and by-products resulting from the combustion process collected from seven combined heat and power (CHP) plants and heating plants located in Upper Silesia, southern Poland, were subjected to petrographic analysis. The coal used as a fuel in these plants was collected from mines of the Upper Silesian Coal Basin; it occurs in a wide range of coalification and has variable quality parameters. The coal is dominated by macerals from the vitrinite group; however, a high content of macerals from the inertinite group has also been observed. Based on petrographic analysis of ash and slags, a highly variable mineral matter content, confirming variable combustion efficiency, has been found. In the case of samples collected from large CHP plants with pulverized and fluidized bed boilers, the mineral matter content is high. The share of mineral matter in combustion products clearly decreases in the case of smaller power plants and heating plants, especially those using grate boilers. The increased content of unburned coal can be explained by the fact that coals of higher rank are often used as fuels in the mentioned plants.

The greatest diversity of char forms can be observed in the case of samples collected from small CHP plants. In the case of three samples of slag, collected from small heating plants using grate boilers, additional presence of coke, thermally altered, and unaltered coal has also been found.

1. Introduction

The combustion of coal generates by-products, including fly ashes, bottom ashes, slags, and flue gas desulfurization products. Fly ash, being the finest fractions, are separated from a stream of exhaust gases by electrostatic precipitators or bag filters. The bottom ashes in fluidized bed boilers and slags in pulverized and grate boilers are cooled with water and collected from the lower part of the boiler using a conveyor belt (Miller, 2010; Breeze, 2015).

The organic by-products of combustion are referred to as unburned coal (carbon), carbonaceous matter, unburned organic matter, loss-on-ignition, or coke; however, in coal petrography, the most commonly used term is char. The presence of unburned carbon in the residues of the combustion process confirms the loss of fuel and reduced degree of efficiency of the process; in addition, it may also prevent the use of ashes (Hower et al., 2017). The amount of unburned coal depends, to a large extent, on the combustion technology (boiler type, temperature and pressure in the boiler, the boiler load, the air to coal ratio) (Yan and Li, 2009; Bartoňová, 2015; Dindarloo and Hower, 2015; Hower et al., 2017). In the case of the most common pulverized boilers, the amount of unburned carbon in the ashes ranges from 2 to 30% depending on technical parameters (Serre and Silcox, 2000). The greatest amount of unburned carbon (35% on average and up to 45%) is recorded in ashes collected from grate boilers (Brown and Dykstra, 1995; Fang et al., 1999). In the case of ashes collected from fluidized bed boilers, the amount of unburned carbon may be comparable or slightly lower (from a few to a dozen or more %) than in the case of pulverized boilers (Bartoňová et al., 2007; Jelonek and Mirkowski, 2015). There is a possibility of a reduction in the amount of unburned carbon in the ashes through the appropriate supply of coal and air to different parts of the boiler (Gao et al., 2013). Measures aimed at reducing the NOx emissions from boilers may contribute to increased amounts of unburned coal as a result of reduced air supply and lowered temperature (Hurt and Gibbins, 1995; Hower et al., 1997; Jones et al., 2010). In order to decrease the amount of unburned carbon, the used coal shall be ground to a finer size (Styszko-Grochowiak et al., 2004).

The unburned carbon matter content is strongly dependent on the degree of coalification and the maceral composition of the burned coal.

⁎ Corresponding author.
E-mail addresses: zbigniew.mirkowski@us.edu.pl (Z. Mirkowski), iwona.jelonek@us.edu.pl (I. Jelonek).

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The increase in unburned carbon is observed with the increasing degree of coalification; this applies to lignites, subbituminous, and bituminous coals (Bartoňová et al., 2007; Craig et al., 2013). Changes taking place with the increase in coalification of bituminous coals, manifested mostly by coking properties of coal, have a strong influence on the behavior of macerals from the vitrinite group in the combustion process. As a result of the combustion of coking coals, the resulting ashes contain coke grains with textures typical for metallurgical cokes (Jasienko, 1978; Gray and Devaney, 1986; Hower, 2017). The increase in coalification of bituminous coals can also cause an increase in grain size (Varma et al., 2014), increase in dense (Ribeiro et al., 2011) and increase in anisotropy of forms (Hower and Mastalerz, 2001) of the unburned carbon contained in the ashes.

The significant influence of the maceral composition of coals on the quantity and nature of the unburned carbon matter stems from the varying susceptibility of individual macerals to pyrolysis. Macerals from the liptinite group are the most susceptible to thermal decomposition; their rapid complete combustion is the reason why unburned residues are not commonly seen in optical microscopic pictures (Bartoňová, 2015). During the decomposition, liptinite produces high amounts of volatiles, contributing to flame stability (Smith et al., 1993). Precursors of unburned forms of coal matter include macerals from the vitrinite and inertinite groups (Alonso et al., 2001; Wu et al., 2006). The results of numerous studies suggest the special importance of macerals from the inertinite group; macerals with strong high reflectance are crucial for the increased amount of unburned carbon in ashes and slags (Vardis et al., 1977; Tsai and Scaroni, 1987; Crelling et al., 1988; Suárez-Ruiz and Crelling, 2008; Valentin et al., 2013). The high content of macerals from the inertinite group is not always a reason for an increase in unburned carbon (Jelonek and Mirkowski, 2015) When it comes to resistance to pyrolysis, macerals from the inertinite group can be divided into reactive and non-reactive (Kruszewska, 1990). Reactive macerals from the inertinite group are subjected to degassing and combustion. Semi-reactive macerals change but retain morphological characteristics of the original macerals. Unreactive macerals from the inertinite group, even in optimal conditions for the combustion process, are not completely burned (Jelonek, 2003; Choudhury et al., 2008). In high-efficiency furnaces, macerals from the vitrinite group and reactive macerals from the inertinite group are completely burnt. The combustion process occurs in several phases (Solomon et al., 1993) and is initially associated with the moisture loss, which is followed by the plastic stage and devolatilization of coal particles (Saito et al., 1987). As a result of the devolatilization process, coal particles are transformed into porous char particles, which in the final stage are fragmented and slowly burned (Smoot and Pratt, 1979). A high content of macerals from the inertinite group contributes to the increased content of higher density grains in unburned carbon (Varma et al., 2014).

Based on microscopic observations of unburned carbon and differences in morphology of individual grains, the classification of forms of unburned carbonaceous matter was carried out. The first classifications used morphological characteristics, the porosity, and wall thickness of individual grains of unburned carbon (Jones et al., 1985; Shibooka, 1985; Tsai and Scaroni, 1987; Bailey et al., 1990; Alvarez et al., 1997; Taylor et al., 1998; Misz, 2002; Cloke et al., 2003). Subsequent classifications also took into account other features, such as optical texture, structure, and nature; however, they were focused on ashes from pulverized boilers (Hower and Mastalerz, 2001; Suárez-Ruiz and Crelling, 2008). The Fly-Ash Working Group, Commission III of the ICCP has developed a petrographic classification of fly ash components, taking into account the origin of organic components and various combustion technologies (Suárez-Ruiz and Valentim, 2015). The above mentioned working group has developed the most recent classification that takes into account multiple levels of authentication, including the origin of organic components (Suárez-Ruiz et al., 2017) with type of particle (char) according to Lester et al. (2010).

The aim of the presented study was to investigated the impact of the coal rank, maceral content of coal and the technology of combustion on the amount and the content of morphological type of particle of unburned carbon in combustion by-products.

2. Samples and analytical procedures

Samples of coal, ash and slag were collected from seven plants generating electricity and heat localized in the Upper Silesia (southern part of Poland). The selected CHP and heating plants represented different technologies and capacities; in addition, they used coals of different quality and petrographic composition. Some samples were collected from large CHPs with capacities in the range from 250 to 800 MW and using pulverized and fluidized bed boilers. The majority of samples was collected from small, local CHP plants and heating plants with capacities in the range from 70 to 150 MW, using pulverized and grate boilers. The C1, FA1, and BA1 samples were collected from a fluidized bed boiler with a capacity of 153 MW (Table 1). The largest number of samples was obtained from four pulverized boilers, with capacities ranging from 64 to 120 MW. Due to technical reasons, the only samples collected from the three grate boilers with capacities ranging from 11.6 to 37.7 MW were slag samples.

The coal samples were collected from landfills, conveyor belts, or automatically from the boiler feeder, depending on the technical capabilities of a given facility.

The samples of slag and bottom ash were collected from the scraper conveyor located under the boiler, while the samples of fly ash were collected directly from electrostatic precipitators. The samples of coal, ash, and slag were then used to prepare pellets using epoxy resin (ISO 7404-2:2005, n.d.). Once hardened, the pellets were ground and polished.

Petrographic analyses of the coal and ash and slag samples were carried out in reflected white light using a Zeiss Axio Imager M2 m microscope combined with a Swift point counter fitted with an automated stage. Maceral analysis and vitrinite reflectance were completed on the coal samples in accordance with ISO (ISO 7404:3:2009, n.d. and ISO 7404:5:2009, n.d.). The ash samples were analyzed using the ICCP classification (Lester et al., 2010; Suárez-Ruiz et al., 2017) in reflected white light using a 500× objective and immersion oil.

3. Results and discussion

3.1. The maceral composition of coal

Bituminous coal used as a fuel in the analyzed CHP plants and heating plants is produced in mines exploiting coal deposits within the Upper Silesian Coal Basin. These coals originate from Upper Carboniferous deposits and represent different ranks of coal, ranging from low to middle rank coals. The lowest rank of coal (Rt about 0.5%) is typical for coals burned in large pulverized and fluidized bed boilers, while the highest degree of coalification (Rt about 1.0%) is reported for coals used in smaller pulverized and grate boilers (by-products resulting from coking coal enrichment process).

The ash content in coals (PN-ISO 1171, 2002) subjected to

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### Table 1: Type and capacity of boilers with number of samples.

| Type of boiler | Capacities of boiler [MW] | Number of samples |
|---------------|--------------------------|------------------|
| Fluidized     | 153.0                    | C1, FA1, BA1     |
| Pulverized    | 120.0                    | C2, FA2, S2      |
| Pulverized    | 112.0                    | C3, FA3, S3      |
| Pulverized    | 64.0                     | C4, FA4, S4      |
| Pulverized    | 120.7                    | C5, FA5, S5      |
| Grate         | 37.7                     | C6, S6           |
| Grate         | 25.0                     | C7, S7           |
| Grate         | 11.6                     | C8, S8           |

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The content of macerals from the vitrinite group in the analyzed samples (Table 3) ranges from 55.8 to 69.7% (mmf). Coal samples with the highest content of macerals from the vitrinite group (61.2–69.7% mmf) are characterized by the lowest content of macerals from the inertinite group (21.1–30.0% mmf). Coal samples with the lowest content of macerals from the vitrinite group (55.8–56.0% mmf) have the highest content of macerals from the inertinite group (> 39%). The content of macerals from the liptinite group ranges from 3.9 to 9.2% (mmf).

3.2. The petrographic analysis of fly ash, bottom ash and slag

In the analyzed samples of ashes and slags, the content of unburned organic matter is highly variable and related to the combustion technology and properties of the combusted coal. The lowest content of unburned carbon was found in combustion by-products (especially fly ash) resulting from the combustion in fluidized bed boilers. In the case of ashes and slags resulting from the combustion of coal in pulverized boilers, the content of unburned coal is highly variable and ranges considerably from 12.0 to 51.6% (Table 4). Such high variability can be explained by the combustion technology - modern technological solutions used in high-power boilers favor more effective combustion of coal. The high content of unburned carbon is typical for some pulverized and grate boilers, which are still used by smaller, local heating and power plants. Another factor affecting the content of unburned carbon is the rank of coal. Large CHP plants generally use steam coals of lower rank, often mixed with biomass, while smaller CHP plants use coals of higher rank. The content of unburned coal may exceed 50%; this applies particularly to samples collected from grate furnaces using coking coals with a high degree of coalification. The combustion of coking coals may seem irrational, but the discussed coals do not meet the quality (high ash content) requirements for coals used in the production of metallurgical coke.

According to the latest ICCP classification (Suárez-Ruiz et al., 2017), fused and porous forms dominate (except samples S6-S8) over unfused and dense forms in the analyzed samples of ash and slags (as confirmed based on the designated levels characterizing the optical character and structure). Isotropic and anisotropic (optical texture) forms are present in equal, or nearly equal proportions, however, in some samples the isotropic forms are dominant (Table 4). The number of anisotropic form increases with a higher degree of coalification of combustion coals. In the case of individual char morphotypes significant variability can be observed (Table 5). The lowest variability was reported in the case of products resulting from the combustion in fluidized bed boilers, especially when it comes to fly ash. In the bottom ash fragments are

Table 2
Quality parameters of coal samples.

| Sample no. | Moisture [%] | Sulfur [%] | Ash [%] | Net Calorific Value [MJ/kg] |
|------------|-------------|-----------|--------|-----------------------------|
| C1         | 16.9        | 1.0       | 17.3   | 19.7                        |
| C2         | 17.7        | 0.9       | 22.4   | 17.5                        |
| C3         | 8.4         | 0.5       | 19.9   | 23.0                        |
| C4         | 8.3         | 0.5       | 29.4   | 23.5                        |
| C5         | 8.5         | 0.5       | 27.6   | 24.1                        |
| C6         | 7.0         | 0.7       | 28.6   | 23.6                        |
| C7         | 7.8         | 0.6       | 10.3   | 27.8                        |
| C8         | 8.2         | 0.5       | 10.6   | 26.9                        |

Table 3
Result of the determination of maceral group composition (mmf, vol,%) of coal and mean vitrinite reflectance (%).

| Sample no. | Vitrinite | Liptinite | Inertinite | Vitrinite reflectance (Ri) |
|------------|-----------|-----------|------------|---------------------------|
| C1         | 69.7      | 9.2       | 21.1       | 0.5                       |
| C2         | 61.2      | 8.8       | 30.0       | 0.5                       |
| C3         | 57.2      | 5.8       | 37.0       | 0.9                       |
| C4         | 55.8      | 4.6       | 39.6       | 1.1                       |
| C5         | 56.0      | 4.6       | 39.4       | 1.1                       |
| C6         | 68.9      | 3.9       | 27.2       | 1.0                       |
| C7         | 46.1      | 8.9       | 45.0       | 1.0                       |
| C8         | 66.7      | 3.8       | 29.5       | 0.9                       |

Table 4
Optical components using ICCP classification (Suárez et al., 2017) of fly ash (FA1-FA5), bottom ash (BA1) and slag (S2-S8) (vol%).
accompanied by inertoid particles, and, to a lesser extent, solid and tenui- and crassinetworks. Fragments (grains < 10 μm) present in the ash are crushed particles of other forms (Fig. 1a, b), mostly inertoids, spheres, and networks (Goodarzi and Hower, 2008). The proper mixture of fuel with air and recirculation of carbon particles contribute to a better combustion and grinding of combustion products, which can be clearly observed in the case of fly ashes from fluidized bed boilers (Vleeskens et al., 1988). In this case, the coal fed to the boiler is characterized by a low degree of coalification and a relatively low inertinite content (sample C1). The increase in the degree of coalification and inertinite content may lead to a decrease in the content of fragments and higher variability of char forms in ashes from fluidized bed boilers (Valentim et al., 2006a, 2006b).

In the majority of samples of ashes and slags from pulverized boilers (samples S2, FA3, FA4, S4, FA5), the dominant components are fragments (Table 5). Other common forms in the examined samples with a

Table 5
Char forms using ICCP classification (Lester et al., 2010, Suárez et al., 2017) composition of fly ash (FA1-FA5) and bottom ash (BA1) and slag (S2-S8) (vol%).

| Sample no. | Mixed porous | Mixed dense | Inertoid | Solid/Fusinoid | Tenusphere | Crassisphere | Tenunetwork | Crassinetwork | Fragments | Mineroid | Coke | Coal |
|------------|--------------|-------------|----------|---------------|------------|--------------|-------------|---------------|------------|----------|------|------|
| FA1        | 0.0          | 0.0         | 0.4      | 0.0           | 0.0        | 0.0          | 0.0         | 0.4           | 99.2       | 0.0      | 0.0  | 0.0  |
| FA2        | 2.0          | 0.0         | 0.8      | 1.2           | 0.0        | 0.4          | 0.4         | 0.4           | 7.2        | 90.0     | 0.0  | 0.0  |
| S2         | 6.0          | 0.8         | 1.2      | 4.8           | 0.8        | 6.0          | 1.6         | 1.2           | 92.2       | 68.4     | 0.0  | 0.0  |
| FA3        | 10.0         | 0.0         | 1.2      | 1.2           | 6.0        | 11.2         | 1.6         | 1.6           | 9.6        | 57.4     | 0.0  | 0.0  |
| S3         | 9.2          | 1.2         | 1.6      | 1.6           | 0.0        | 5.6          | 2.0         | 6.8           | 0.0        | 72.0     | 0.0  | 0.0  |
| FA4        | 6.0          | 0.0         | 1.6      | 0.4           | 4.0        | 6.4          | 1.6         | 1.6           | 30.0       | 48.4     | 0.0  | 0.0  |
| S4         | 3.2          | 0.0         | 0.4      | 0.4           | 1.2        | 4.8          | 4.0         | 1.2           | 11.6       | 73.2     | 0.0  | 0.0  |
| FA5        | 2.0          | 0.0         | 0.4      | 0.4           | 1.6        | 2.0          | 1.2         | 0.8           | 20.8       | 70.0     | 0.0  | 0.0  |
| S5         | 4.4          | 0.4         | 1.6      | 1.6           | 2.8        | 2.8          | 4.8         | 2.8           | 0.0        | 78.0     | 0.0  | 0.0  |
| S6         | 4.4          | 0.0         | 0.8      | 4.0           | 1.2        | 2.4          | 1.2         | 6.7           | 58.3       | 13.9     | 5.9  | 5.9  |
| S7         | 2.7          | 0.0         | 13.6     | 0.4           | 0.0        | 0.0          | 0.4         | 0.0           | 57.7       | 22.9     | 2.3  | 2.3  |
| S8         | 1.2          | 0.0         | 2.8      | 0.0           | 0.0        | 0.0          | 1.2         | 3.6           | 68.4       | 17.2     | 5.6  | 5.6  |

Fig. 1. Char forms in fly ash samples from fluidized boiler (a, b) and fly ash and slag samples from pulverized boilers (c, d, e, f) (oil immersion, x50 objectives and polarized): a, b - fragments (sample FA1), c - solid (sample FA4), d - crassisphere (sample FA3), e - mixed porous (sample S5), f - mixed porous (sample FA3).
relatively large share are spheres and networks (Fig. 1d); slightly higher contents of inertoids, solids (Figure 1c), and mixed forms (Fig. 1e, f) can be observed in some of the examined samples. A high content of spheres and networks is typical for ashes and slags resulting from the combustion of low- and middle rank coals in pulverized boilers (Bailey et al., 1990; Bend et al., 1992; Bartoňová, 2015). The lowest variability in char forms was recorded for FA2 sample (Table 5) probably resulting from the combustion of coal with a low degree of coalification (Rr 0.5%) and a relatively low inertinite content (sample C2. A totally different character of the unburned carbon was recorded in samples resulting from the combustion of coking coal in grate boilers (Table 5). The grain size of the unburned coal in these samples reaches up to 5 cm; the macroscopic characteristics are typical for metallurgical coke. In the case of slags from grate furnaces, the microscopically visible organic matter consists of various forms of coke and coal (samples S6 and S8) with a small proportion of typical morphological forms of unburned coal. When it comes to forms typical for metallurgical coke, fine-medium- and coarse mosaic were usually observed; domains were found in smaller quantities. In individual coke grains, small fragments of macerals from the inertinite and vitrinite groups were also observed.

The occurrence of forms typical for coke (Jasienko, 1978; Gray and Devanney, 1986; Hower et al., 2017) in by-products of the combustion process is possible during the combustion of coals of higher rank (coking coal), which go through the thermplastic stage, producing anisotropic-coke texture of unburned coal (Hower, 2012; Bartoňová, 2015).

The presence of coal grains in slags produced in grate boilers has been confirmed. The samples contain thermally unaltered inertinite, vitrinite, and microlithotype grains (Fig. 2a, e). The presence of grains of thermally altered vitrinite (sometimes inertinite - Fig. 3d), containing cracks typical for such forms (Fig. 3a, b), and light color rims around the mentioned cracks or vitrinite particle, has also been confirmed (Fig. 3a-c) Some of the vitrinite grains show traces of more complex thermal changes in the form of transition into the plastic phase with devolatilization pores (Fig. 3c, e-f). The thermally altered carbon grains have also been reported in the combustion products of coals of higher ranks (anthracites) (Hower et al., 2017). The most commonly observed thermal changes of coal were observed in samples obtained from self-heated coal waste dumps (Misz and Fabiańska, 2007; Suárez-Ruiz et al., 2012), or intrusive affected coal seams (Singh et al., 2007).

4. Conclusions

The aim of the study was to show the impact of the combustion technology, rank of coal, and the petrographic composition on the amount and nature of unburned carbonaceous matter in ashes and slags. The lowest amount of unburned coal was found in the ash from fluidized bed boilers (5.6–7.6%), in which the fuel circulates relatively...
longer in the combustion chamber; in addition, unburned coal is captured in cyclones and returned to the boiler. However, in the case of pulverized boilers, the range of values is very large and depends mainly on the type of coal burned and the use of technological solutions for the combustion of coke residues. The highest amounts of unburned coal were recorded, among others, in slag samples from grate boilers. In the mentioned grate boilers, coal with different grain size form the fuel bed; the primary air is supplied from the bottom of the grate, which is not favorable for good mixing of fuel with the air. The content of unburned coal clearly increases in the case of the combustion of coals with a higher degree of coalification (R_t about 1%) in pulverized and grate boilers.

The lowest variability of forms of unburned organic matter was reported for the by-products of combustion in fluidized bed boilers, especially in the case of fly ash, which contains only fragments. The mentioned fragments dominate in all of the examined samples obtained from pulverized bed boilers; in addition, higher amounts of spheres and networks can also be observed. This is a typical composition of char forms for both ashes and slags resulting from the combustion of low- and middle rank coals in pulverized boilers. The different composition of unburned carbon was found in the case of slag samples from grate boilers, in which coking coals were burned. The mentioned samples contain forms typical for metallurgical coke (mosaic, domain, pyroilitic coal), grains of unchanged carbon, and vitrinite grains showing characteristic thermal changes (cracks, rims, plastic phase, and pores).

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Fig. 3. Thermally altered coal in slag sample (S6) from grater boilers (oil immersion, x50 objectives and incident light): a - particle of coal with cracks and light color rims, b - vitrinite with craks and zoned rim, c - vitrinite with initial stage of devolatilization, d - inertinite with rims, e, f - particles of coal with final stage of devolatilization.
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