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# Assessment of Heating and Cooling of a Spontaneous Fire Source in Coal Deposits—Effect of Coal Grain Size

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**Abstract:** Fire hazard assessment in coal mines is performed on the basis of concentrations of particular gases emitted from the heating coal deposit, but more precise criteria and indicators are needed to assess fire hazard properly—both during the temperature rise phase and in the coal bed cooling phase. In the paper the impact of coal grinding on hazard assessment of spontaneous fire development in the coal deposit during heating and cooling the fire source was analyzed. The intensity of desorption of ethane, ethylene, propane, propylene, acetylene, carbon monoxide and hydrogen is the resultant of temperature and grinding of coal samples. The results proved that the ratio of concentrations emitted by standard versus coarsely crushed coal for each of the gases, changed both in the growth phase as well as in the temperature drop phase. It was found that as the temperature rose, the effect of coal grinding on the release of ethane, ethylene, propane, propylene and carbon monoxide decreased. The greatest effect of coal grinding was observed in the case of ethane and propane, while the lowest in the case of hydrogen and carbon monoxide.

Keywords: coal; self-heating; fire hazard; coal grain size

## 1. Introduction

The assessment of fire hazard in hard coal mines is conventionally performed using chromatographic analysis. The following gases are monitored in the mine air: ethane, ethylene, propane, propylene, acetylene, carbon monoxide and hydrogen. Their presence may testify to the heating of coal, which in turn may lead to the occurrence of spontaneous fire [1-3]. This is accompanied by a decrease in oxygen content in the mine air [1,4]. The processes of self-heating and self-ignition of coal are widely discussed in the literature, especially in aspects related to the desorption of hydrocarbons and other gases [5–9]. In literature, there are no research results concerning the effect of coal grinding on the desorption of these gases during coal heating and cooling. Despite the development of fire prevention measures in coal mines, new fires are still occurring which cause safety concerns and financial losses [10,11]. Self-heating of coal and other fuels is also considered to be a safety hazard in other industrial applications, including raw materials production, storage and transport or biomass handling in energy applications [12–14]. Self-heating tests in a larger scale were performed by Mao et al. [15] on a sample of 1.5 Mg tested in 39 days. Low-temperature tests of coal oxidation (at 40, 60 and 90 °C) aiming at the assessment of the combined effect of process temperature, grain size and petrographic properties of Turkish coals were carried out by Baris et al. [16]. The self-heating characteristic of lignite depending on gas flow rates, coal and mine air moisture content, and coal grain

size was assessed by Kucuk et al. [17]. The effect of coal particle size on emission of carbon monoxide, carbon dioxide, alkanes and alkenes during oxidation at the temperature range of 30–500 °C was tested by Zhao et al. [18]. In Polish hard coal mining, the maximum permissible concentration of carbon monoxide is 26 ppm [19], while the concentrations of the other tested gases are not included in the regulations. In the initial phase of carbon monoxide emission, the stream most probably originates from carbon monoxide physically adsorbed on the surface of coal grains [20–22]. The extend of sorption process and sorption capacities of coal and coal derived carbon materials were previously reported to be depending largely on pressure and temperature, as well as gas atmosphere of partial processing of coal under elevated pressure and temperature [23–26]. The degree of self-heating development is commonly assessed on the basis of measurements of concentrations of hydrocarbons and other gases in a mine air [27–30]. The study on oxygen consumption and emission of gaseous products from coal samples heated to 230 °C resulted in determination of the self-heating index [31]. The intrinsic properties and spontaneous combustion tests of selected coal and coal-shale samples were conducted and a relationship between the two has been established. An increase in carbon, moisture, hydrogen, volatile matter, nitrogen and a decrease in ash content indicate an increased proneness to self-heating [32,33].

Some parameters are determined indirectly and in an approximate way, which is why determining the state of heating or cooling is a difficult issue which requires further research. Coal is crushed during mining and transport, while the remaining coal residues in the gobs are also crushed by the pressure of the overburden rock. Different types of coal with greater susceptibility to crushing and different types of overburden rock cause coal fragmentation to various extent. The mechanization of mining causes the percentage share of coal with grains below 10 mm to increase. The problem of the impact of coal grinding is of great importance when assessing the fire hazard. The lower the coal grain size the more carbon monoxide, being the main gaseous indicator applied in self-ignition index calculations, was reported to be released at ambient temperature [34].

Therefore, the main objective of the conducted research is to show the impact of changes in the grinding of hard coal samples on the amount of released gases accompanying the process of self-heating of coal and, as a consequence, spontaneous fire. Coal grinding is a parameter related to the individual characteristics of hard coals. It depends on the petrographic composition of coal, the composition of the mineral substance and the degree of metamorphism. There are no standards that explicitly allow the classification of coals according to their grinding susceptibility [35], but it is well recognized that more porous coals are softer and more prone to crushing. The paper presents the results of chromatographic analyses of gas concentrations from analyzed coal samples taken from hard coal seams in eight mines in two variants of grinding (standard ones and so-called coarse ones) and in two process phases, of heating and cooling. The first variant of coal grinding was achieved at the opening of the crusher gap of about 2 mm, while the second one at the opening of about 5 mm. Coal is crushed in gobs which increases its contact surface area with mine air, and creates conditions for an intensive desorption of gases. The studies on the effect of coal grain size on the release of particular gases would allow the assessment of coal susceptibility to oxidation in gobs. The results are considered to be contributing to a more in-depth assessment of the process of coal heating in gobs as well as fire hazard conditions in a fire field which is cooled down.

#### 2. Materials and Methods

For the tests, eight samples of hard coals from various hard coal mines were collected in accordance with the relevant standard [36]. Coal samples were taken from currently exploited mine longwalls of the mines of the Upper Silesian Coal Basin in Poland. Basic physical and chemical parameters were determined for the collected samples (see Table 1). Elemental analysis for C, H, N and S content as well as technical analysis for moisture, ash and volatile matter were carried out. Moreover, the heat of combustion and the calorific value of the samples were determined. The sample was taken from a freshly exposed coal seam, from places representing the average properties of the seam with respect to its spontaneous fire hazard susceptibility. Collected coal samples in the form of solids of the greatest

mass possible were placed in a hermetic container. The collected solids were then sprinkled with additional fine coal and sealed in a container. The coal collected for testing was subsequently crushed in a disk coal crusher with screw feeder. Solids with a grain size less than 2.5 mm and a weight of 6 kg were used as samples intended for testing. The samples thus prepared were again placed in a metal container with nitrogen blowdown. Two types of samples with different grain distributions were prepared for each of the tested coals. The first sample was crushed in the crusher at standard settings (at the opening of the crusher gap of about 2 mm), while the second sample was crushed at the maximum opening of the crusher gap (5 mm). The results of the sieve analysis of the percentage share of grains in the analyzed analytical samples are presented in Figure 1.







(b)

Figure 1. Grain analysis of coal samples: (a) standard and (b) coarsely crushed.

Sample No.	Transient Moisture, % w/w	Moisture, % w/w	Ash, % w/w	Volatiles, % w/w	Carbon, % w/w	Sulphur, % w/w	Hydrogen, % w/w	Nitrogen, % w/w	Oxygen, % w/w	Self-Ignition Index *A, kJ/mol	Self-Ignition Index **Sz <sup>a</sup> , °C/min	Self-Ignition Group
1	3.6	4.4	8.2	32.1	72.7	0.9	4.7	1.3	7.6	52	105	IV
2	1.2	0.8	4.2	23.0	87.8	0.5	3.2	1.1	8.3	60	42	Π
3	4.9	3.1	7.8	28.7	72.9	0.6	4.1	1.0	8.2	54	109	IV
4	2.7	2.6	3.9	27.9	78.7	0.4	4.4	1.4	8.3	52	97	III
5	8.8	9.2	4.9	34.1	70.6	1.1	4.7	0.8	12.3	47	135	V
6	1.8	1.1	4.1	22.9	88.1	0.5	4.6	1.2	4.2	66	45	Ι
7	1.6	1.2	4.7	26.5	84.5	0.4	4.4	1.6	4.6	69	53	Ι
8	1.7	1.3	7.5	33.9	83.8	1.0	5.4	1.4	3.4	65	62	Π

**Table 1.** Physical and chemical parameters of coal samples.

\*A—Activation energy of oxidation; \*\*Sz<sup>a</sup>—Self-ignition index.

The tests into oxidation and cooling processes were carried out in a metal reaction chamber in the shape similar to the sphere (see Figure 2). Inside the sphere, crushed coal was placed. Synthetic air (during the coal heating stage) and nitrogen (during the coal bed cooling stage) were introduced into the reaction chamber with a flow of 1.11 cm<sup>3</sup>/s, respectively. The coal bed was heated by means of a heating element made of a spiral heating cord with a diameter of 40 mm and a height of 40 mm. The space between the rolls of the heating cords was filled with ground coal, which formed a cylindrical sample at the temperature set by the temperature control system. Tests of the coal heating process were carried out in the temperature range from 35 to 300 °C. Two channels were provided to the place of coal heating, ensuring the inflow of synthetic air (in the heating stage) and nitrogen (in the cooling stage of the deposit). Each experiment has been conducted for 50 hours. Thermal losses were determined on the basis of the measured values of temperatures in six measuring points: at the source of heat, and at 40, 60, 80, 100 and 120 mm from the heater (sensors 1–6). With the sufficiently large thermal capacity used (mass of the coal sample 6 kg) the temperature fluctuation was suppressed. A temperature sensor was installed in the apparatus, which controls the process and was used for its adjustment. The temperature measurement of coal during the reaction was recorded on-line with an accuracy of 0.2 °C.



**Figure 2.** Diagram of a test stand for testing the process of coal heating: 1—synthetic air, 2—nitrogen, 3—gas flowmeter and flow regulator, 4—thermocouples No. 2–6, 5—temperature, flow rate and gas sampling controller, 6—temperature recording, 7—air inlets, 8-termocouple No. 1, 9—heating zone, 10—gas outlets, 11—outlet gas flowmeter, 12—automatic gas sampling device, 13—Tedlar bag, 14—gas chromatographs, 15—gas concentrations recording.

Synthetic air with a flow of 4.0 dm<sup>3</sup>/h was fed into the coal sample placed in the reaction chamber. The tests included both the heating phase (stage I) and coal bed cooling (stage II). In stage I the increase of the maximum temperature in the source (sensor 1) was analyzed, as well as temperatures at five remaining measuring points. Gas concentrations in an atmosphere containing 20.5% vol. oxygen, i.e., values close to the oxygen content in the circulating current of mine longwalls in hard coal mines were recorded. The exhaust gases were analyzed for ethane, ethylene, propane, propylene, acetylene,

oxygen, carbon dioxide, methane and hydrogen at temperatures of 35, 50, 100, 150, 200, 250 and 300 °C. In stage II, the temperature drop during cooling the heated coal bed was analyzed, analogously monitoring temperatures on sensors 1–6 and analyzing gas concentrations in a nitrogen atmosphere, i.e.. an atmosphere corresponding to a perfectly nitrogenized gobs space during the cooling of a fire source in the mining gobs. The experiment was carried out until the coal sample reached a final temperature of 30 °C.

#### 3. Results

The assessment of coal self-heating in mines is commonly performed on the basis of measurements of concentration of particular gases, including ethylene, propylene, acetylene, carbon monoxide and hydrogen applied in calculations of fire indices [1,3,37]. The use of Graham index as well as several other indices based on concentrations of carbon monoxide, hydrogen, ethylene, propylene and acetylene in a mine air is legally required in Poland [38]. Other indices used include, for example, the ratio of carbon monoxide to carbon dioxide in a mine air [39]. The temperature of heated coal is determined on the basis of the fire indices values and compered with the temperature of a standard coal sample thermally oxidized under laboratory conditions. Changes in concentration of particular gases and their ratios significantly affect the values of fire indices and the fire hazard assessment. The effect of coal grain size on concentrations of particular gases applied in the determination of fire indices, and therefore on the fire hazard assessment, was performed in this study.

Ethane accompanies methane which is released during coal mining. Between the tested coals there are very large differences in the concentrations of released ethane (see Figure 3). Considering the large amount of ethane released on the fragmented coal samples, it can be concluded that the process of coal crushing affects its compact structure. The lowest value of 8.28 ppm at 35 °C, i.e., the so-called background, was observed for a coal sample 4 with coarse grinding. The highest ethane value amounting to 730 ppm was found for a coal sample of 2 subjected to normal grinding. Such large differences in concentrations were found both during the heating and cooling stage. A correlation was observed between the concentration of the released ethane and the degree of sample coalification. Coals with higher carbon content release much higher amounts of ethane than coals less carbonified. For individual coals, increases in ethane concentrations were small and in the range from 3.6 for coal 3 to 12.0 for coal 4 (see Figure 3). Maximum ethane concentration was recorded for sample 2 at the temperature of 300 °C and it was 0.34% vol. (at the same time the maximum methane concentration was 2.26% vol). The lower explosion limit of ethane is 3.1% vol. and it is lower than the lower explosion limit of methane, which is 5.3% vol. Local accumulation of ethane and methane may create an explosion threat. For all examined coals, changes in the ratio of released ethane (in the case of coarse vs. standard coal crushing) with temperature increase were observed. In the coal bed cooling process, a decreasing tendency was observed. However, compared to the heating phase, the change in the concentration ratio of the released ethane proceeded much slower and did not reach the initial values of ethane concentration.

Figure 4 presents the changes in the ratio of released ethylene (coarse vs. standard coal crushing), which in the natural state occurs in coal in small quantities. For the tested coal samples, the release of ethylene at the temperature of 35 °C ranged from 0.02 to 0.15 ppm. In the conducted tests, an increase in the concentration of ethylene was observed as the temperature increased. At the highest temperature tested (300 °C), it reached several dozen ppm, with the maximum value of 56 ppm reported for sample 3. Ethylene release ratios in coarse vs. standard coal grain size changed to a greater extent than in the case of ethane (see Figure 3; Figure 4). During coal cooling a decreasing tendency was also observed, however compared to the heating phase, changes in the concentration ratio of the released ethylene took place much slowly and, as in the case of ethane, they did not reach the initial values.



**Figure 3.** The course of changes in the ratio of released ethane (coarse vs. standard coal crushing) in the stage of heating and cooling of coal samples.



**Figure 4.** The course of changes in the ratio of emitted ethylene (coarse vs. standard coal crushing) in the stage of heating and cooling of coal samples.

Propane, similarly to ethane, accompanies methane which is released during coal mining process (see Figure 5). The tested coals clearly showed different characteristics in terms of the amount of propane released during the heating process. The lowest concentrations of propane were observed at 35 °C and were treated as background. For coal sample No. 8, the concentration of propane was 0.70 ppm (coarse crushing). The highest concentration of propane was observed in the process of heating coal sample 2 (43.7 ppm standard crushing). With the increase of the heating temperature of the

tested coal samples, no significant changes in propane concentrations were observed. The maximum concentration of propane of 0.047% vol. was recorded for sample 2 at 300°C. Although the lower explosive limit of propane is 1.9% vol. it should be noted that propane-ethane mixture can constitute an explosion hazard. For all coal samples tested, changes in the released propane ratio (for both coarse and standard crushing) with increasing temperature were observed. During the cooling of the tested coal samples, a decreasing tendency was also observed. However, compared to the heating phase, the changes in the propane gas ratio in both options (coarse and standard) run much slower and do not reach the initial values.



**Figure 5.** The course of changes in the ratio of released propane (for both coarse and standard crushing) in the stages of heating and cooling of coal samples.

Propylene in the natural state is still present in smaller quantities than ethylene. For the examined coal samples, the concentrations of propylene at the temperature of 35 °C ranged from 0.02 to 0.09 ppm (Figure 6). As the temperature rose, the concentration of propylene increased several times reaching several ppm at 300 °C. The highest concentrations of propylene were observed for coal sample No. 8. For propylene, the proportions between standard and coarsely crushed coal changed as in the case of ethylene.

Acetylene is virtually absent in the natural state. For the tested coals its concentration at 35 °C amounted to 0.001–0.002 ppm, which is the same as the threshold of determination (see Figure 7). Acetylene concentrations at temperatures up to 100 °C were maintained for all tested coal samples at this level. Therefore, the ratio of released acetylene in coarsely crushed coal vs. standard crushed coal was equal to one. As the temperature rises, the differences between standard and coarsely crushed coal increased. At 300 °C, the ratio was from 62% for sample 2 to 98% for sample 8. For the cooling phase, the mentioned ratios were again close to being equal. One of the fire indices applied is based on the ratio of ethylene and acetylene concentrations. Changes in ratios of these gases will affect its value (see Figure 4; Figure 7).



**Figure 6.** The course of changes in the ratio of released propylene (for both coarse and standard crushing) in the stages of heating and cooling of coal samples.



**Figure 7.** The course of changes in the ratio of released acetylene (for both coarse and standard crushing) in the stages of heating and cooling of coal samples.

The effect of coal grain size on coal oxidation is hardly observable at lower temperatures, of about 40 °C, as previously reported by Baris et al. [16]. No such effect was also observed for coal grain size lower than 1 mm by Kaji et al. [6] and Wang et al. [31], who stated that the rate of carbon dioxide and carbon monoxide creation is rather chemically controlled than mass transport controlled. Lignite samples of lower coal grain size and higher moisture content were reported to be more susceptible to self-ignition by Kucuk et al. [17]. Coal grain size was also claimed to be affecting oxygen consumption

and release of particular gaseous components in coal heating phase by Wang et al. [31], with the release of hydrogen at 90–100  $^{\circ}$ C.

The ratio of the released carbon monoxide during the heating tests of coarsely crushed coal vs. standard crushed coal changed at a temperature of 35 °C from 45% for sample 2 to 73% for sample 6 (see Figure 8). At 300 °C, these ratios were from 73% for sample 3 to 93% for sample 8. For the cooling phase, the ratio of carbon monoxide released during coal heating phase of coals subjected to coarse vs. standard crushing decreased again reaching values from 60% for sample 6 to 79% for sample 8.



**Figure 8.** The course of changes in the ratio of released carbon monoxide (for both coarse and standard crushing) in the stages of heating and cooling of coal samples.

Hydrogen concentration in the natural state is very low and amounts to about 0.5 ppm, as it was for all tested coal samples at 35 °C. With a temperature increase to 200 °C, hydrogen concentrations steadily increased (see Figure 9). However, with a further rise in temperature, the dynamics of the concentration increase dropped. During the cooling phase, hydrogen concentration again dropped to background value, or 0.5 ppm. For all samples tested, a slight impact of grain size variations on the change of the ratio between standard and coarsely crushed coal was observed. The ratio of hydrogen emission in the studies of coarsely crushed coal vs. standard crushed coal changed from 77% for sample 2 to 97% for sample 8.

The susceptibility of coal to crushing is an important aspect of fire hazard assessment, since it results in lower size of coal particles, which in turn is favorable for development of higher sorption capacity and for intensification of gaseous components release. The correct assessment of fire hazard requires therefore the analysis of the effect of coal grain size on fire indices values for various coal seams to be performed.

Figure 10 highlights the average differences between coals subjected to normal and coarse crushing for all eight coals tested, for the heating and cooling phase separately. The largest spread of average values was noted for ethane and propane during both the heating and cooling phases. For the heating phase, the smallest differences were observed for hydrogen, carbon monoxide and acetylene. However, for the cooling phase, the smallest differences were observed for carbon monoxide and hydrogen.



**Figure 9.** The course of changes in the ratio of released hydrogen (for both coarse and standard crushing) in the stages of heating and cooling of coal samples.



**Figure 10.** Distribution of average differences between standard and coarsely crushed coal for the heating (h) and cooling (c) phase.

The results presented prove that the correct selection of fire indices taking into account coal type, the effect of temperature in coal heating and cooling phase, as well as coal grain size range is required for a precise assessment of fire hazard in coal mines.

### 4. Conclusions

In the analysis of the effect of grindability and coal grain size on the fire hazard assessment, it was found that:

1. For all tested coal samples, as the temperature rose, the differences in ethane, ethylene, propane, propylene and carbon monoxide concentrations between the standard and coarsely crushed coal decreased during the heating phase. In the cooling phase, the differences between standard and coarsely crushed coal increased again, but did not reach the original level.

2. In the initial stage of heating, there were no differences in acetylene concentrations because this gas was only emitted above 100 °C. Hydrogen concentrations in the initial heating phase were at the limit of quantification, while significant differences in hydrogen concentrations were observed only at temperatures above 100 °C. During the cooling phase, the differences of acetylene and hydrogen decreased again, reaching the trace values.

- For the heating phase, the dispersion of average values of the gases tested between coals subjected to standard and coarse crushing were the highest for ethane and propane, while the lowest for hydrogen, carbon monoxide and acetylene. For the cooling phase, the distribution of average values between standard and coarse coal grains were also the highest for ethane and propane, while the lowest for carbon monoxide and hydrogen.
- The average values of differences between standard and coarse coal grains were lower for the heating phase than for the cooling phase.
- Coal crushing had a significant impact on the differences in gas concentration, which directly translates into an assessment of the fire hazard both during heating and cooling of coal. The selected fire indices are calculated on the basis of the amounts of gases released from crushed coal in a function of temperature. These indices are employed in determination of self-heating temperatures of coal in a coal mine. The determination and selection of a proper coal grain sizes of coal samples tested is important in terms of the representativeness of the results of these analyses which have been proven in the study presented and should be the subject of further research. The laboratory scale testing of coal samples crushed to the degree reflecting the natural coal crushing conditions in a coal mine would allow for better representation of natural conditions of the phenomena tested under laboratory conditions and would improve the correctness and the credibility of the results.

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