



Analysis of adsorption tests of gases emitted in the coal self-heating process



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ABSTRACT

The analysis of the results of sorption absorbcency of hard coals with respect to gases relevant in assessing the fire hazard (carbon monoxide, carbon dioxide, hydrogen, ethylene, propylene, and acetylene) was the aim of this study. Hard coals adsorb carbon dioxide and acetylene in the highest amount, and ethylene and propylene in a slightly smaller amount. The adsorption of carbon monoxide and hydrogen is very small. The amounts of adsorbed gases depend on the metamorphism degree of hard coals as well as on their porosity. Hard coals of high porosity with good accessibility of internal surface, of low metamorphism degree and of higher oxygen content are characterized by greater adsorption capacity. Fire gases generated from the self-heating center may be the subject to adsorption on hard coals, which results in the lower gas concentration at the measuring points and as a consequence in the change of fire indicators calculated on their basis. This phenomenon applies in particular to hard coals with high sorption absorbcency and to the situation in which the self-heating center and the point of measurement of concentrations is considerably distant, which causes greater contact of the emitted gas with coal matter, and more possible intensification of the adsorption process. Fire indicators are used to assess the development of the process of self-heating of coal. Gases, the concentration of which is the part of the particular fire indicator, should have a similar adsorption capacity. Indicators constructed on the basis of a very different adsorption capacity can be a source of errors due to the observed adsorption process.

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1. Introduction

1.1. Coal self-heating process

In hard coal mines, the process of low-temperature carbon oxidation with the oxygen in the air is dealt with. The exothermicity of this process causes, in terms of accumulation of heat, the possibility of coal self-heating, and when the flashpoint is exceeded, of self-ignition, which can consequently result in the dangerous endogenous fire. However, in order to initiate this process, appropriate conditions must be met. Firstly, the coal in the deposit must have the inclination, or susceptibility, to self-ignition. The second prerequisite is the adequate oxygen supply to coal. The third condition is the ability to accumulate the heat emitted from the exothermic reaction of hard coal with oxygen from the air. In a situation of the coexistence of all these conditions, coal self-heating, respectively self-ignition and endogenous fire takes place [1–4]. Underground fire is a negative phenomenon, being a source of serious losses resulting from a temporary isolation of mining areas, and it is also a risk for the work safety of miners. Due to the inaccessibility of

fire outbreaks they are typically difficult to extinguish. In the case of mines with methane hazard the fire may additionally cause the ignition and explosion of the methane. Modern mining focuses on activities aimed at the prevention of fire in order to stop the process of coal self-heating in the early stage and to prevent the occurrence of fire [5,6].

The coal self-heating process has been the subject of many studies and scientific considerations [7–12]. A number of methods of determining coal self-ignition have been developed. The calorimetric methods [12–14], critical temperature method [15], perhydraulic method, or widely used in Poland the Olpiński method based on testing the intensity of the oxidation of carbon samples included in standard PN-G-04558: 1993 are especially known [16]. The methods of measuring the coal temperature using laser instruments, infrared, or thermographic cameras as well as the statistical methods, taking into account the impact of different coal properties on its tendency to self-ignition [17] may also be observed. Apart from the known and described methods of assessing coal tendencies to self-ignition, new solutions, especially in the field of fire risk assessments, are still searched in order to fully control the self-heating process. One of such methods is a method for early fire detection carried out on the basis of the measured gas concentrations emitted as the consequence of the coal self-heating. The oxidation processes are accompanied by a significant number of gases, and the most important from the point of view of the evaluation of the described hazard includes: carbon monoxide, hydrogen and unsaturated hydrocarbons:

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ethylene, propylene, and acetylene [18]. The concentrations of these gases increase along with the increasing temperature of self-heating coal. The emission of the gases listed above is accompanied by a decrease in oxygen content [19,20].

1.2. Fire indicators

On the basis of the measured concentration of gases emitted in the process of the self-heating of coal, fire indicators are calculated. The most commonly used indicators are constructed on the basis of the concentration, mainly of carbon monoxide, but also of oxygen, nitrogen, and carbon dioxide. Among them the most famous are:

Graham indicator [21], calculated from the dependency:

$$G = \frac{CO}{0.265 \times N_2 - O_2} \quad (1)$$

where: CO, N₂, O₂ – percentage: carbon monoxide, nitrogen, oxygen
Trickett indicator [22,23]

$$TR = \frac{CO_2 + 0.75 \times CO - 0.25 \times H_2}{0.265 \times N_2 - O_2} \quad (2)$$

where: N₂, H₂, CO, CO₂ – percentage: carbon monoxide, carbon dioxide, nitrogen, oxygen, hydrogen
carbon monoxide growth indicator

$$\Delta CO = CO - CO^x, \% \quad (3)$$

where CO – percentage of carbon monoxide at the exit measurement station; CO^x – percentage of carbon monoxide at the inlet measuring station [21],

Oxide indicator [24]

$$\frac{CO}{CO_2} \quad (4)$$

where CO, CO₂ – content of carbon monoxide, carbon dioxide
indicator WP1 [21]

$$WP1 = \frac{CO}{H_2} \quad (5)$$

where CO, H₂ – concentration of carbon monoxide and hydrogen, ppm.

The basis of most of the indicators is the concentration of carbon monoxide, and changes of its content may be affected by various factors, not related to the processes of coal oxidation. As shown by mining practice, not always are the used indicators sufficient to correctly assess fire hazard. The Graham indicator takes too high values proving a fire risk at high mine air thinness [25]. In addition, the inertization processes, frequently used in mines, also affect the value of this indicator and can lead to erroneous data.

In recent years, thanks to the development of gas chromatography and therefore the possibility of the determination of low concentrations of hydrocarbons: ethylene, propylene and acetylene in order to increase the control of the fire hazard state in mine excavations, fire indicators on the basis of which one can estimate the maximum temperature of coal heating have been developed. The main ones are shown below:

$$\frac{C_2H_4}{C_2H_2} \quad (6)$$

$$WP2 = \frac{C_2H_4 + C_3H_6}{H_2} \quad (7)$$

$$WP3 = \frac{C_2H_4 + C_3H_6}{C_2H_2} \quad (8)$$

where C₂H₄, C₂H₂, C₃H₆, and H₂ are the gas concentrations measured in mine air, ppm.

Before proceeding to the assessment of fire hazard in the given seam on the basis of the indicators listed above, it is necessary to carry out laboratory tests of coal from this seam. Coal samples are placed in a special chamber, air is brought to it and the concentrations of emitted gases at increasing temperatures are measured. On the basis of the content of the gases are determined the reference fire indicators, which are compared with those calculated on the basis of current gas concentrations measured in the mining areas during the monitoring of the fire hazard. This allows assessing the temperature of coal heating [21].

The most important parameter used in the assessment of fire hazard is therefore the concentrations of individual gases measured in mine air. Coal is a porous material with an extensive structure of pores capable of absorbing vapors and gases. It may be therefore assumed that gases emitted from the center of self-heating, migrating through excavations, are subject to the sorption phenomenon which can cause a reduction in the concentrations of these gases measured at the measurement points, and thus an invalid assessment of self-heating. The sorption phenomenon does not occur in thermal coal oxidation processes carried out in laboratory conditions. The emitted gases undergo the chromatographic analysis immediately.

The purpose of the proposed work is to use the results of sorption test of gases emitted as a result of coal self-heating in the assessment of this process. For this purpose, the sorption absorbency of coals in relation to gases significant for fire hazard assessment has been determined: CO, H₂, C₂H₂, C₂H₄, C₃H₆, and CO₂ to determine the extent to which the gases emitted from the center of self-heating are absorbed and how it can affect the fire indicators determined on the basis of the concentrations. The obtained outcome may contribute to a better interpretation of the results of the current chromatographic analyses of mine air and to improve the methods for early detection of fires.

2. Materials and methods

Six samples of hard coal taken from active Polish coal mines located in the Upper Silesian Coal Basin in the southern part of Poland have been selected for sorption tests. The test samples have been collected in accordance with the standard PN-G-04502: 2014-11 “Hard coal and lignite collection and preparation of samples for laboratory tests. Basic methods.” The collected samples have been crushed in a jaw crusher, ground and with the help of a sieve manufactured by the company Fritsch, the grain class 0.5–0.7 mm has been separated. The samples prepared in such a way have been stored in nitrogen atmosphere until the sorption measurements.

Chemical and technological characteristics as well as petrographic analysis of the tested samples have been carried out on the basis of the guidelines of Polish standards. The results of these analyses have been summarized in Table 1.

Porosity as well as the volume of pores with the method of mercury porosimetry have been determined for the tested coal samples. The tests have been performed using the Pascal 440CE Instruments device in the following range of pressure 0.1–400 MPa. The test results have been provided in Table 2.

The surface area calculated with the Dubinin–Radushkevich model on the basis of carbon dioxide sorption isotherms designated in the temperature of 298 K as well as the surface area calculated with the BET model on the basis of nitrogen sorption isotherms designated in the temperature of 77.5 K have also been determined. The sorption isotherms of carbon dioxide and nitrogen have been determined with help of the volumetric method using an apparatus ASAP 2010 made by Micromeritics. The obtained results have been shown in Table 2. Table 2 also includes the volume of micropores calculated according to

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