

# POSSIBILITY OF USING TRACER GASES TO DETERMINE THE COAL MASS IN THE OUTBREAK OF SPONTANEOUS COMBUSTION AND RELATED AFFECTING FACTORS

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## Abstract

Spontaneous combustion of coal mass is a very actual problem in underground coal mines. Professional research workplaces have investigated the problem since the first half of the twentieth century. Spontaneous combustion of coal in the form of endogenous fires is a reason of extraordinary events whose consequences are serious both in terms of economic losses, and in the field of security, because in the worst cases they are accompanied by the loss of human lives. Tracer gases are associated manifestations of each spontaneous combustion process, but their utilization to determine the extent of the outbreak of endogenous fire is burdened by numerous factors. The article addresses these affecting factors as well as the possibility of using the tracer gases to determine the mass of spontaneous combustion outbreak.

## Abstrakt

Samovznícení uhelné hmoty je stále aktuálním problémem hlubinných uhelných dolů. Odborná výzkumná pracoviště se tímto problémem zabývají již od první poloviny minulého století. Samovznícení uhlí je v podobě endogenních požárů příčinou mimořádných událostí, jejichž důsledky jsou závažné jak v oblasti ekonomických ztrát, tak v oblasti bezpečnosti, neboť jsou v nejhorších případech doprovázeny ztrátami na lidských životech. Indikační plyny jsou doprovodným projevem každého samovzěcovacího procesu, ale jejich využití pro zjištění rozsahu ohniska endogenního požáru je zatíženo četnými faktory. Předložený článek se zabývá těmito ovlivňujícími faktory a rovněž možnostmi využití indikačních plynů pro stanovení hmotnosti ohniska samovznícení.

## 1 INTRODUCTION

Coal mass spontaneous combustion is a problem whose intensive solution is being searched for many decades. First scientific findings on spontaneous combustion of coal originate from the first half of the twentieth century from Great Britain and are a work of the authors Winmill, Graham, et al. The objective findings concerned especially oxidation of coal mass, kinetics of spontaneous combustion and tracer gases, e.g. till now known and respected so far Graham's numbers [1]. A significant shift in the early indication of spontaneous combustion was made by Japanese and European research in the field of monitoring higher light gaseous hydrocarbons, e.g. [2-8]. In Czechoslovakia and later in the Czech Republic this research was conducted in the seventies of the last century at the Mining Institute of the Academy of Sciences of the Czech Republic [9], Science-Research Coal Institute in Ostrava-Radvanice [10], in the eighties at the Central Mines Rescue Station in Ostrava [11] and from the late nineties the research was concentrated at the Institute of Mining Engineering and Safety of the Faculty of Mining and Geology at the VSB-Technical University of Ostrava [12, 13]. The mentioned research in the Czech Republic has brought important findings in the field of spontaneous combustion outbreak temperature estimation and a basis for the adoption and derogation of binding decisions of the State

Mining Administration [14], aimed at minimizing the risk of the occurrence of spontaneous combustion in OKR mines. The last cited research [12, 13] was implemented in the Czech Republic within research and development activities of the Czech Mining Authority and was aimed at improving the methods for early indication of spontaneous combustion. The research started from actual findings and dealt with the kinetics of spontaneous combustion of coal in the OKR, repeatability of thermal oxidation method, factors affecting the accuracy of the estimate of the spontaneous combustion temperature, application of computer technology for estimating the temperature of spontaneous combustion, etc.

## 2 METHOD OF THERMAL OXIDATION

The principle of thermal oxidation consists in determining concentrations and compositions of gaseous products released during thermal oxidation of coal by atmospheric oxygen in the temperature interval from 40 to 200°C [15], in some cases up to 380°C [16] under laboratory conditions. The concentrations of tracer gases are verified by a chromatographic method every 20°C. The so-called tracer gases of spontaneous combustion of coal mass, which are represented in the mine atmosphere, are gases - carbon monoxide, carbon dioxide, hydrogen, methane, ethane, propane, butane, ethylene, propylene and acetylene.

In order to verify the repeatability and reproducibility of the results obtained by the method of thermal oxidation in the framework of the research CBU R&D project, no. 29 [17], repeated series of testing analyses with identical coal samples were carried out at three research workplaces using the same methodology proposed by VVUÚ, a.s. Ostrava. The research was conducted at the following workplaces: VSB-TU Ostrava, VVUU a.s., Ostrava - Radvanice and Institute of Geonics AS CR Ostrava.



**Fig. 1** Identical coal sample packed in bags [18]

One repeatability test series of thermal oxidation was represented by an identical coal sample taken pointwise in a coal seam (20 kg of lump coal). After preparation of fraction, homogenization and vacuum packaging of one sample (Fig. 1) the sample portions were transported by five to the above three research workplaces. This way two series were tested (two samples per 20 kg of coal), which means 2x (3x5), i.e. 30 measurements of thermal oxidation. The third series (the third coal sample of 10 kg) was tested only by a quintuple analysis only at the workplace of the VSB-TU Ostrava. In total, within the repeatability 35 gas images of tracer gases of thermal oxidation have been obtained.

The results of thermal oxidation are expressed by a graphic gas image drawn up from volume developments of partial tracer gases related to 1 ton of a coal sample ( $\text{ml.t}^{-1}.\text{min}^{-1}$ ) according to the formula (1):

$$q_v = \frac{q_{vo} * c}{m} \quad (1)$$

Where:

$q_v$  - volume development of tracer gas of the thermal oxidation method [ $\text{ml} \cdot \text{t}^{-1} \cdot \text{min}^{-1}$ ],

$q_{vo}$  - volume flow rate of oxidizing medium of the thermal oxidation method [ $\text{ml} \cdot \text{min}^{-1}$ ],

$c$  - concentration of tracer gas [ppm],

$m$  - weight of coal [kg].

Measurements of volume developments of tracer gases of thermal oxidation were evaluated by using two methods. Within the first method a range of variation was used. The measured results of the developments of partial tracer gases were averaged. For the calculated averages the range of variation from the mean value was determined percentually. After the evaluation it was possible to say simply that the average range of variation of deviations from the mean value was for carbon dioxide in the range of  $+ / - 10\%$ , while for gaseous hydrocarbons  $+ / - 40\%$ . Within the other method a coefficient of variation was selected. The same input data was used. The coefficient of variation is defined as the ratio of the standard deviation and the absolute value of the mean according to the relation (2):

$$V_k = \frac{s}{\bar{X}} * 100 \quad (2)$$

Where:

$V_k$  - coefficient of variation [%],

$s$  - standard deviation [ $\text{ml} \cdot \text{t}^{-1} \cdot \text{min}^{-1}$ ],

$\bar{X}$  - arithmetic mean [ $\text{ml} \cdot \text{t}^{-1} \cdot \text{min}^{-1}$ ].

The coefficient of variation was calculated for each gas and each temperature and by the calculation the average resulting intervals of the experimental error for the carbon dioxides  $\langle 0.2690 \text{ to } 0.4151 \rangle$  and gaseous hydrocarbons  $\langle 0.2750 \text{ to } 0.3818 \rangle$  were determined. The resulting calculated experimental error, expressed percentually, is for carbon dioxide equal to the interval  $\langle 26.90 \text{ to } 41.51\% \rangle$  and for gaseous hydrocarbons  $\langle 27.50 \text{ to } 38.18\% \rangle$ .

Differences in values resulting from the previous and new data processing are probably caused by using other statistical methods of calculation. More evident are the differences for carbon dioxides, where after the new data processing the experimental error is higher, while the upper limit of the interval for gaseous hydrocarbons nearly corresponds with the earlier calculation. Another affecting factor of the calculation can be the omission of improper data from the input database of developments in litres.

The experimental error of the thermal oxidation method was calculated to be obvious what is the error the subsequent estimation of the extent of endogenous fire using tracer gases is burdened with and the calculation clearly shows that the desorption property of coal mass is very wide and is one of the most important affecting factors.

### 3 UTILIZATION OF TRACER GASES TO ESTIMATE THE WEIGHT OF COAL IN THE OUTBREAK OF SPONTANEOUS COMBUSTION

Estimating the amount of coal in the outbreak of spontaneous combustion resulted from the volume development of tracer gases of the thermal oxidation method, i.e. the formula no. (1). By the adjustment of the formula a calculation formula was obtained for estimating the weight of coal in the outbreak of spontaneous combustion (3):

$$m = \frac{q_{vo} * c}{q_v} \quad (3)$$

The calculation of coal weight in the outbreak of spontaneous combustion is conditioned by taking a sample of the air mass from passing air current as it is necessary to know for the calculation the volume air flow rate at the measurement point. From the measured volume flow rate of passing air current at the sampling point and found out tracer gas concentrations the developments in litres of partial gaseous components are determined.

The relevant developments in litres are assigned to the corresponding phases of column diagram of the Central Mines Rescue Station Ostrava, i.e. according to the methodology by [11]. Thus the approximate temperature of the outbreak of spontaneous combustion is determined. According to the temperature the sample is assigned its laboratory development [ $\text{ml. t}^{-1}.\text{min}^{-1}$ ]. The result, i.e. the estimated amount of coal in the outbreak of spontaneous combustion, is achieved by the proportion of the volume flow rate generated in the laboratory in the volume flow rate of tracer gas determined on in-situ conditions according to the formula (3). From the gases falling into the identical temperature phase the arithmetic average of volume developments is calculated. The resulting average value is used to calculate the amount of coal in the outbreak of spontaneous combustion in a given temperature phase. Due to the large number of affecting factors, which are listed in the following part of the article, it is only an estimate of the extent of the outbreak of spontaneous combustion.

#### **4 FACTORS AFFECTING THE USE OF TRACER GASES TO ESTIMATE THE EXTENT OF THE OUTBREAK OF SPONTANEOUS COMBUSTION**

As mentioned above, the estimate of the extent of the outbreak of spontaneous combustion is burdened by a number of affecting factors. These include especially a broad desorption feature of coal mass, which has already been described above, but also the factors affecting the laboratory procedures and factors occurring in mining operations. These are listed in the following part of the paper.

##### **4.1 Factors affecting laboratory procedures**

###### **Deviations of chromatography**

The research verification results of the accuracy of chromatography showed that measurement results may be affected, inter alia, by technical means, e.g. a reducer used [13]. Based on the research the following recommendations for the chromatographic analysis have occurred:

- for chromatograph calibration it is appropriate to use a certified calibration mixture,
- to perform chromatograph calibration always immediately prior to the analysis and to carry out the calibration more frequently in the case of a series of analyses,
- to reduce FID detectors noise by using the synthetic air, i.e. the air prepared from pure oxygen and nitrogen.

Currently, there are available three gas chromatographic laboratories for the needs of the OKR, namely at the Lazy Mine in Orlova, Darkov 2 Mine in Karvina and Central Mines Rescue Station in Ostrava Radvanice.

###### **Effect of sample container**

Within the evaluation of affecting factors a series of measurements of the sample composition was carried out depending on the type of sample container [18]. The analysis results showed that the best type of sample container with a relatively stable composition is a sample-glass, on the contrary the sample container with silicone tubing, for which the steepest decrease in concentrations was observed in all cases, proved itself the least. The sample container with rubber tubing is comparable with the all-glass sample container thanks to its proved features.

###### **Effect of coal grain size**

The grain size of coal was examined as another possible factor affecting the composition of gaseous products [18]. The measurements implied that the grain size of coal mass is a significant parameter affecting the final values of the volume developments of gaseous components released during coal oxidation. The measurements showed that in tests with a coarser grain size all gaseous components show substantially less volume developments of desorbed gases than in the tests with a finer fraction.

##### **4.2 Affecting factors occurring in mining operations**

As is clear from the above, the development of gases under laboratory conditions is dependent on many factors. However, also the mining practice is influenced by many factors, which significantly distort the estimated temperature of spontaneous combustion. Given that there are various operating conditions, each case of spontaneous combustion of coal mass is unique. For the most accurate estimation of the temperature in the outbreak of spontaneous combustion it is necessary to realize the differences between the laboratory and operating conditions.

## Temperature inhomogeneity of the outbreak of spontaneous heating

Temperature is one of the most important factors affecting the release of tracer gases from coal mass. Some methods of assessing the cases of spontaneous combustion of coal is trying to use this knowledge in practice. One of the ways to obtain an estimate of the temperature in the outbreak is to compare the gas image obtained by a laboratory test of thermal oxidation with the sample of air mass taken in a mining environment. However, this estimate is influenced by many factors.

Under laboratory conditions the test of thermal oxidation is made with a relatively small amount of coal, c 150 g, so we can say that the temperature throughout the portion of the sample is constant [17]. Therefore, it is assumed that the gas images of thermal oxidation represent a thermally homogeneous state of coal mass in each rated temperature, which is one of the most significant differences between the laboratory and operating conditions. In the mining environment tracer gases are released from a thermally non-homogeneous outbreak and usually represent a mixture of gaseous components released from the entire volume or area of spontaneous heating with different temperature representation, implying that the gases collected in the mine air show the average temperature of the outbreak which they are released from.

## Inhomogeneity of the spontaneous heating outbreak fraction

As mentioned above, a coal mass fraction has an effect on the release of tracer gases. For coarser fractions the oxidation reaction is less intense and less gases is released from coal than for finer fractions. In mining operations a homogeneous fraction of coal mass cannot certainly be assumed and given that the vast majority of cases of spontaneous combustion take place in gob areas, it is not possible to determine the degree of coal mass crushing. For this reason when comparing the laboratory results with the operational values the estimate of the temperature of outbreaks of spontaneous combustion can be distorted.

## Oxygen concentration

The research [17] found that the oxygen concentration affects the oxidation reactions and the subsequent development of tracer gases. The lack of oxygen reduces the intensity of the oxidation reactions and thus the development of tracer gases, on the contrary for increasing oxygen concentrations the increase of unsaturated hydrocarbons is higher than of saturated hydrocarbons. The oxygen concentration affects also the rate of the temperature increase in the outbreak of spontaneous combustion. In operational practice, the oxygen concentration is significantly influenced by operational and technological conditions. Generally speaking, the oxygen concentration decreases with increasing distance into the depth of the gob, and this decrease is dependent on the directional incline of the face, gas capacity, gob permeability, gob sealing, artificial inertisation, etc. Given that in cases of spontaneous combustion of coal left in the gobs it is not possible to localize accurately the outbreak, the oxygen concentration estimate is hardly to determine.

## Reverse adsorption of tracer gas

The accuracy of the estimate of the spontaneous combustion outbreak temperature is also affected by a reverse adsorption [17] of the gaseous products of oxidation on the surrounding rocks. Hydrocarbons, which are released in the spontaneous combustion process, are subject to a subsequent interaction with the coal mass or surrounding rocks. In laboratory-acquired gas images of thermal coal mass loading, which can be used to estimate the temperature of the outbreak, the adsorption is not taken into account. It is not possible to include into the laboratory research additional parameters that influence the selective sorption such as the quantity of surrounding rocks, through which gaseous products are filtered, time of filtering through the surrounding rocks, the exact physical properties of the rocks (temperature, degree of breaking-up, etc.).

As is evident from the above, the use of tracer gases is burdened by numerous factors. Some of the factors are not susceptible to influence and it should be underlined that current methods used for assessing the development of spontaneous combustion using tracer gases are not exact. This is only an estimate.

## 5 CONCLUSION

Using the tracer gases of spontaneous combustion is influenced by a number of important factors, both in the laboratory and under mine conditions. The most important factor is the broad desorption feature of coal mass. A number of described laboratory-technical and technical-operational factors are connected with this basic factor. All of these affecting factors are included in the calculation of the outbreak extent. For this reason, this is only an estimate, not an exact calculation. An improvement of the calculation and its wider use in practice remain therefore an open question. Given that spontaneous combustion is still a current issue in underground coal mines, we need to continue to address this problem.

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## RESUMÉ

V ostravsko-karvinském revíru patří výskyt samovznícení uhelné hmoty k závažným problémům ovlivňujícím dobývání a to jak z hlediska ekonomického, tak zejména z hlediska ohrožení pracovníků. Nebezpečí výskytu záparů na dolech je spojeno zejména s výskytem oxidu uhelnatého a možnou iniciací metanovzdušné směsi. Z tohoto důvodu jsou v OKR aplikována závazná opatření, jejichž základem je sledování tzv. indikačních plynů samovznícení. V současné době jsou pro potřeby OKR v provozu tři plynové chromatografické laboratoře (důl Lazy v Orlové, důl Darkov 2 v Karviné a HBZS, a. s., Ostrava-Radvanice), dále řada chemických plynových laboratoří a monitorovací sítě oxidu uhelnatého.

Proces samovznícení uhelné hmoty je procesem složitým a jeho podrobné poznání je cestou k možné minimalizaci výskytu. Proto by měla být tato problematika nadále řešena.