A BRIEF OVERVIEW OF DEVELOPMENT IN THE USE OF INDICATOR GASES FOR COAL SPONTANEOUS COMBUSTION AND PROSPECTS FOR FURTHER SOLUTION

STRUČNÝ PŘEHLED VÝVOJE VYUŽITÍ INDIKAČNÍCH PLYNŮ SAMOVZNÍCENÍ UHLÍ A VÝHLED DALŠÍHO ŘEŠENÍ

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Abstract

One of the most serious risks associated with mining using mainly underground methods is the risk of mine fires. Causes of these fires can be exogenous (external) or endogenous (internal). The authors of the article pay attention to endogenous fires in coal mines, especially to the timely indication of them. As already mentioned in many contributions, spontaneous combustion is a complicated process that passes, in the final stage, into a mine open fire. The localization of a place of such danger can be done by means of so-called indicator gases that are desorbed from the coal substance at a point of the originating seat of spontaneous combustion. The article deals with a development trend in the use of indicator gases for spontaneous combustion in coal mining. The objective of the authors was to arrange chronically and briefly knowledge of the use of indicator gases both abroad and in the Czech Republic and to provide input information about a research project being dealt with at present.

Abstrakt

Při hornické činnosti prováděné především hlubinným dobýváním je jedním z nejzávažnějších rizik vznik důlních požárů. Příčiny vzniku těchto požárů mohou být, buď exogenního (vnějšího) anebo endogenního (vnitřního) původu. Autor článku věnuje pozornost požárům endogenním v uhelných dolech a to především jejich včasné indikaci. Jak už bylo v mnoha příspěvcích zmíněno, samovznícení je složitý proces, který ve své konečné fázi přechází v otevřený důlní požár. Odhalit místo takového nebezpečí lze pomocí tzv. indikačních plynů, které desorbují z uhelné hmoty v místě vznikajícího ohniska samovznícení. Článek se zabývá trendem vývojem využití indikačních plynů samovznícení v uhelném hornictví. Cílem autora bylo chronologický a stručně setřídit poznatky o využití indikačních plynů jak v zahraničí, tak v ČR a podat vstupní informaci o současně řešeném výzkumném projektu.

Keywords: spontaneous combustion, indicator gases, coal, higher hydrocarbons

1 INTRODUCTION

Indicator gases for coal spontaneous combustion can be divided into two groups, namely majority gases (CO, CO_2, O_2, CH_4), continuously observed in the mine atmosphere, and minority gases (C_2H_6, C_2H_4, C_2H_2, C_3H_8, C_3H_6, C_4H_{10}), analysed in cases of indications of spontaneous combustion. In mines of the Czech Republic, these gases are sampled in different ways. The majority gases are sampled using a so-called “wet sampling” method to glass gas collecting tubes filled with a sealing liquid. The minority gases are sampled using a so-called “dry sampling” method by means of vacuum ejectors.

2 FOREIGN KNOWLEDGE

The first who started to investigate these problems was a British researcher J.I. Graham (1920). He focused mainly on the release of CO and CO_2. During his research he found that in the course of oxidation of coal, CO was released and oxygen concentration decreased depending on the temperature of oxidized coal. His research led to the development of a method based on the determination of two basic indexes (Graham's ratios), i.e. CO/ O_2 deficiency and CO_2/O_2 deficiency.

A Japanese author Kitagawa (1959) noticed in the course of observation of a mine fire that, in addition to CO and CO_2, ethylene and other gases were released. On the basis of this finding, an impulse was given all over the world to examine gaseous higher hydrocarbons as possible gases for the indication of a spontaneous combustion hazard. Because ethylene occurred in very low concentrations and at the given time it was difficult to measure, Kitagawa designed his own detection tubes (colorimetric tube for CO detection and linear tube for C_2H_4).

Pursall and Banerjee (1961) were other authors who were concerned with the determination of higher hydrocarbons, above all ethylene and propylene in the mine atmosphere. In their researches they used chromatography with argon as carrier gas. For gas detection, a thermal conductivity detector was used; however, for low concentrations of these gases it turned out to be a failure. After experience thus the authors proposed to use a more sensitive detector for further observations. Subsequently, Pursall and Gosh (1963) used a modified argon ionization detector. Traces of ethylene began to appear before changes in values of Graham indexes were observed. They found that at a sevenfold increase in the basic $CO/\Delta O_2$ ratio, propylene began to appear. On the other hand, propylene disappeared at a threefold decrease in the basic ratio. At a ninefold increase in the $CO/\Delta O_2$ ratio, acetylene occurred. Based on these findings they concluded that the occurrence and increase in the amount of ethylene, propylene and acetylene in this order indicated a danger of heating of the coal substance. Furthermore, they arrived at the conclusion that with decreasing temperature, these hydrocarbons disappeared in the order from acetylene, propylene and ethylene.

In Poland, Muzyczuk (1966) was concerned with the problems of determination of higher saturated and unsaturated hydrocarbons. He carried out his research at the Central Mining Institute (GIG), a Polish research institute in Katowice. Thanks to his methods, the presence of aliphatic and aromatic hydrocarbons in very low concentrations could be detected. Saturated and unsaturated aliphatic hydrocarbons were generated during low-temperature oxidation. Muzyczuk also stated that unsaturated hydrocarbons, in which the prevailing component was ethylene, were formed merely during the oxidation process. In a case of aromatic hydrocarbons, these gases did not occur at temperatures less than 300°C. Muzyczuk was of the opinion that CO, the concentration of which was 100 to 1000 times higher than that of ethane or ethylene, seemed to be the most suitable indicator gas.

Chamberlain (1970) together with his research team was engaged in heating British coal samples at a constant flow of air or nitrogen. Gaseous hydrocarbons were analysed chromatographically. For the determination of aliphatic hydrocarbons (from methane to butane), a chromatograph with a FID detector was used; the detector was able to detect the presence of the observed gases even in low concentrations, e.g. the minimum detection limit for methane was about 0.5 ppm and that for butane was about 2 ppm. For the determination of hydrogen, nitrogen and oxygen, the other chromatograph with a molecular sieve, which was able to determine e.g. hydrogen even at a minimum detection limit of about 1 ppm, was used. Chamberlain came to the conclusion that at temperatures more than 40 °C, CO began to be released from the examined coal. Another released gas was hydrogen; it was released at a temperature of about 80°C with maximum release at about 200°C. Propylene was released at 137 °C with maximum release at about 230°C; but immediately after that a sharp drop in it followed. Ethylene was released at temperatures of about 155°C. From these conclusions it followed that CO seemed to be the most suitable indicator gas indicating the occurrence of spontaneous combustion. In addition to these measurements, the authors were also concerned with the problems of effects of the degree of coalification on coal oxidation. They arrived at the conclusion that the dynamics of oxidation of low-rank coals was higher (higher consumption of oxygen and greater formation of CO) than that of high-rank coals.

In the year 1973 Chamberlain and Hall (1973) continued the previous researches. This time they used a system of enrichment of indicator gases proposed by Novák and his team (1965) for the determination of indicator gases; samples being analysed were concentrated to measurable limits. Thanks to this method, Chamberlain and Hall were able to isolate 30 different hydrocarbons; 20 of them they were able to determine by experiment. They also stated that a ratio between iso-butane and n-butane could indicate the level of ongoing oxidation of the coal substance.

Chamberlain (1976) during his researches always drew attention to one serious problem, namely the fact that although CO seemed to be the most suitable indicator gas, its concentrations grew only after exceeding the critical temperature of coal spontaneous combustion moving in a temperature range of 30-70°C (according to the coal rank). For this reason, in the next tests he focused especially on gases released in the course of oxidation at temperatures below 120°C. For testing, three coal samples of different ranks were selected. It was found that at temperatures less than 10°C, none of chromatographically analysed gases with a concentration higher than the 0.01 ppm limit occurred. Measurable concentrations of chromatographically analysed gases began to appear at

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temperatures of 10-70°C and increased rapidly. However, at exceeding 70°C, the generation of the gases but CO became stable.

The problem of timely indication of spontaneous combustion was also solved by a research team of Aijperovič, as stated in Faster (1976), in the former U.S.S.R. Aijperovič together with the team found that in the course of initial oxidation, unsaturated hydrocarbons, namely ethylene and acetylene in trace concentrations were released. During their research they however tackled the problem of detection of these hydrocarbons owing to their low concentrations. They developed a special concentrator that was able to separate required components from a rather large volume of gaseous oxidation products. Subsequently, the components were desorbed in the laboratory and in concentrated form were analysed chromatographically. For the estimation of temperature of the seat of spontaneous combustion a table of values of the ethylene/acetylene ratio was determined.

Authors Chakravorty and Feng were researchers who were concerned with indicator gases for the spontaneous combustion of Canadian coals. They performed their tests on the coal from the Sparwood coal field. This coal was heated at temperatures from 20°C to 250°C. By measurements they came to the conclusion that at temperatures below 74°C, mainly CO and CO_2 were released. Only at temperature of 110°C, perceptible traces of hydrogen and ethane began to appear, but their concentrations were much lower than the concentration of CO. Higher saturated and unsaturated hydrocarbons (propane, butane and ethylene) were released only at higher temperatures. In Fig. 1 a well-known graph showing the formation of indicator gases (above all CO, hydrogen, ethane and ethylene) of the cited authors is stated. As for the dynamics of release of indicator gases, it is obvious that CO predominates. For this reason, the authors proposed to measure continuously CO using modified IR analysers directly in mining conditions.

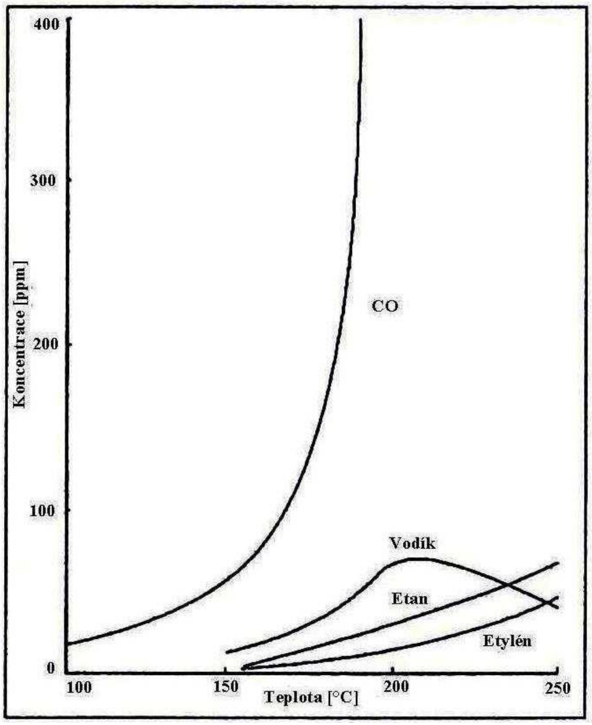

Fig. 1 Chakravorty's graph 1978

In the nineties, Cygankiewicz (1996) also began, on the basis of knowledge acquired during previous researches done at the coal research institute GIG in Katowice, to be concerned with the development of indicator gases by thermal oxidation. In samples examined at temperatures from 50 to 250°C, the formation of common indicator gases and simultaneously of C_2–C_4 saturated and unsaturated hydrocarbons was found. Based on these gases, the following six dimensionless indexes were taken as most suitable for the assessment of the process of spontaneous combustion:

- C_2H_4/C_2H_2 (ethane/acetylene),
- C_3H_6/C_2H_2 (propylene/acetylene),
- 100 * (C_2H_4/H_2),
- CO/H_2,
- 100 * G (Graham's ratio),
- $CnHm/H_2$.

From his research Cygankiewicz drew the conclusion that the value of all these dimensionless indexes increased with growing temperature. $CnHm/H_2$ and CO/H_2 indexes exhibited the highest tendency to grow; on the contrary, Graham's ratio showed the lowest. Later, this method was used successfully in practice in one of Polish mines, when the process of spontaneous combustion was revealed. According to the author, this method can be used for the assessment of the process of spontaneous combustion. The method has been offered by the research institute GIG for operational purposes up to now.

3 KNOWLEDGE OF UTILIZATION OF INDICATOR GASES IN THE CZECH REPUBLIC

At the beginning of the seventies, Lanková et al. (1975) as the first ones began carrying out oxidation tests on coal. This team began to determine the concentrations of methane, ethane, propane, n-butane, ethylene and propylene by means of a chromatograph at temperatures of 150, 200 and 240°C. Conclusions confirmed the above-mentioned information, i.e. the higher the temperature, the greater formation of hydrocarbons; the higher the hydrocarbon, the smaller formation of it; at the same number of carbons, a saturated hydrocarbon is formed in a higher degree. To distinguish hydrocarbons evolved by oxidation from hydrocarbons released by heat action, the tests in which nitrogen was used as a circulating gas were carried out. The result was the conclusion that ethylene and propylene were formed in low concentrations when the coal substance was exposed to the mere action of heat without the presence of oxygen.

Other authors concerned with these issues were Harašta, Čermák (1979). On the basis of research aimed at preparing a proposal for the method of determination of higher hydrocarbons to find indications of spontaneous combustion, to assess the extent of a space affected by spontaneous combustion in the early stage and temperature of spontaneous combustion in the early stage, they arrived, at the research institute VVUÚ in Ostrava-Radvanice, at the finding that higher hydrocarbons and their ratios (methane, ethane, ethylene, acetylene, propane, propylene and butane) were, under normal conditions of mining without indications of spontaneous combustion, with common ventilation systems and under stable pressure, constant and their ratios between each other did not change. By continuous decreasing the concentration ratios of methane/ethane, ethane/propane and propane/butane, it was possible to expect the increased oxidation of coal with accumulation of heat. The presence of higher unsaturated hydrocarbons, mainly ethylene and propylene, warned of spontaneous combustion in the early stage. The occurrence of acetylene in measurable concentrations indicated an increase in the temperature of coal and a situation when the fire almost originated.

In this period, researches based on the above-mentioned knowledge were also carried out at the Main Mining Rescue Station (henceforth referred to as MMRS) in Ostrava. The output of these researches was "A Temporary Method for the Evaluation of Chromatographic Determination of Hydrocarbons", Apfelthaler (1983), and subsequently "The Evaluation of Spontaneous Combustion Process of Hard Coals of Saddle Seams in the Ostrava-Karviná Coalfield Using Gas Chromatography – Method of MMRS in Ostrava", Hajník (1987). The researches concerned were based especially on the evaluation of numerous samples of the mine atmosphere. The mentioned method prescribes the way of dry sampling of the atmosphere. On the basis of detected concentrations of indicator gases, individual phases of spontaneous combustion process are assigned to particular gases. According to this method, in the early phase of spontaneous combustion, CO and gases of alkane series (ethane, propane, butane), which under normal conditions with the exception of ethane do not occur in the mine atmosphere, begin to appear. From the occurrence of other hydrocarbons, temperature at the seat of spontaneous combustion can be deduced as well; if propane and butane in concentrations of 0.01-1 ppm appear, temperatures at the seat of spontaneous combustion can be expected to be in the range of 30-60°C. Temperatures above 60°C indicate the presence of ethylene in concentrations of 0.01-1 ppm and a sharp increase in CO. By subsequent increase in temperature to 90-110°C, hydrogen appears in concentrations of 10-1 000 ppm and simultaneously the concentrations of hydrocarbons determined earlier grow to the values of 1-100 ppm. If the limit of 230°C is exceeded, the presence of acetylene is detected, when the fire originates. On the basis of these findings, a column graph, determining amounts of formed individual gases in litres in the given phases of spontaneous combustion was constructed (Fig. 2).

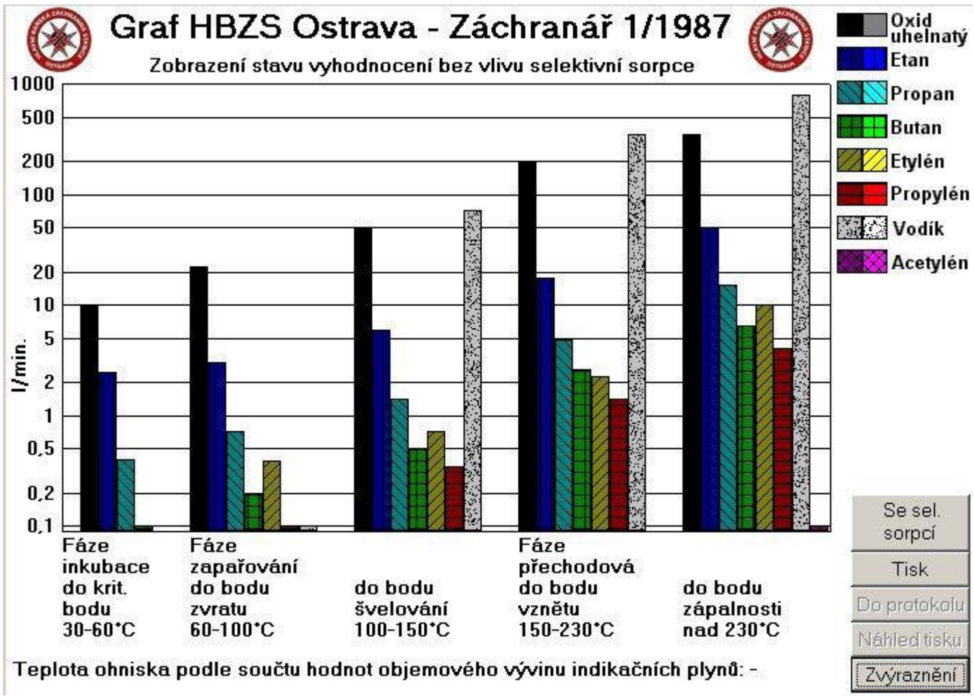

Fig. 2 Print screen from the program "Evaluation of Indicator Gases for Spontaneous Combustion of Coal Substance", version 2.3 (Stecker, 2005)

On the basis of chromatographic measurements of the amounts of formed hydrocarbons, Prof. Taraba (2003) developed a so-called gas thermometer, on which intervals for measurable evolved indicator gases were illustrated (Fig. 3). Thick lines represent the most frequently determined ranges of temperatures of indicator gases and thin lines represent the maximum spreads of temperature values.

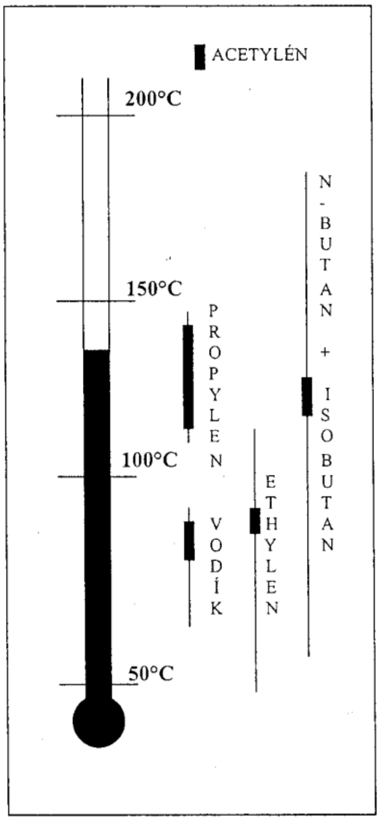

Fig. 3 Gas thermometer (Taraba, 2003)

Researches were done at VŠB – Technical University of Ostrava as well. One of researchers was Jindřich Šancer (2004), who in the course of his study for the doctoral degree participated in research dealing with the problems of indicator gases for spontaneous combustion of the coal substance. He stated acquired knowledge in his Ph.D. thesis. Above all, he was concerned with the formation of products of thermal oxidation of coal samples from the Ostrava-Karviná Coalfield (henceforth referred to as OKC) depending upon temperature (Fig. 4) and with their relations to the physical and chemical properties of coal and to the sampling locality. Based on knowledge obtained from previous researches, he tried to verify these properties in practice using mathematical statistical methods and data analysis.

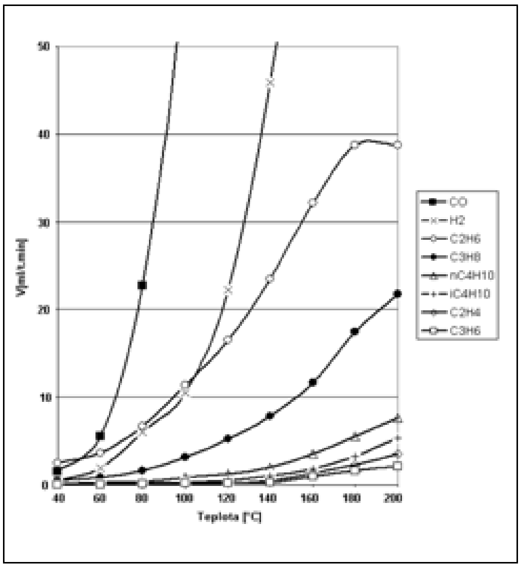

Fig. 4 Average volumes of formed products of thermal oxidation of OKC coal samples (Šancer, 2004)

Further research concerning these issues was carried out by a team of researchers led by Prof. Adamus (2002, 2004) in the framework of scientific research projects VaV ČBÚ No. 3/1999 (1999-2002) and No. 29/03 (2003-2005). One of subtasks of this research was timely indication of the early stage of spontaneous combustion of the coal substance. Here, findings from previous two researches were processed and a database making it possible to generate gas patterns of indicator gases for thermal oxidation of 63 verified coal samples was created. One of gas patterns of thermal oxidation from catalogues mentioned below is given in Fig. 5. In addition to the assessment of the released amounts of gases, ratios between particular oxidation products could be assessed here as well. As suitable dimensionless indexes of spontaneous combustion of coal, C_2H_6/C_3H_8, C_2H_6/C_2H_4, C_2H_4/C_3H_6 and CO_2/C_2H_4 indexes were determined besides the already used CO_2/CO ratio. Two of outputs of the concerned research were catalogues of coal seams prone to spontaneous combustion in the Czech Republic, Adamus (1999-2002), and a computer program for work with gas patterns of indicator gases for thermal oxidation of verified coal samples, Stecker (2005). At the Faculty of Mining and Geology of VŠB – Technical University of Ostrava, a system for the creation of gas etalons for partial developed faces in the OKC was designed and verified. The procedure for etalon preparation was based on the verification of gas patterns of indicator gases for spontaneous combustion using the method of thermal oxidation of three coal samples from a coal block prepared for mining. The concerned gas etalon was subsequently designed for use in practice in case of spontaneous combustion at the given face, e.g. "Indicator Gas Etalon for Coal Substance Spontaneous Combustion Valid for the ČSM-Sever Mine, Seam 32, Block 2, Face No. 320 203, Faculty of Mining and Geology of VŠB-Technical University of Ostrava, Ostrava 8 June 2005".

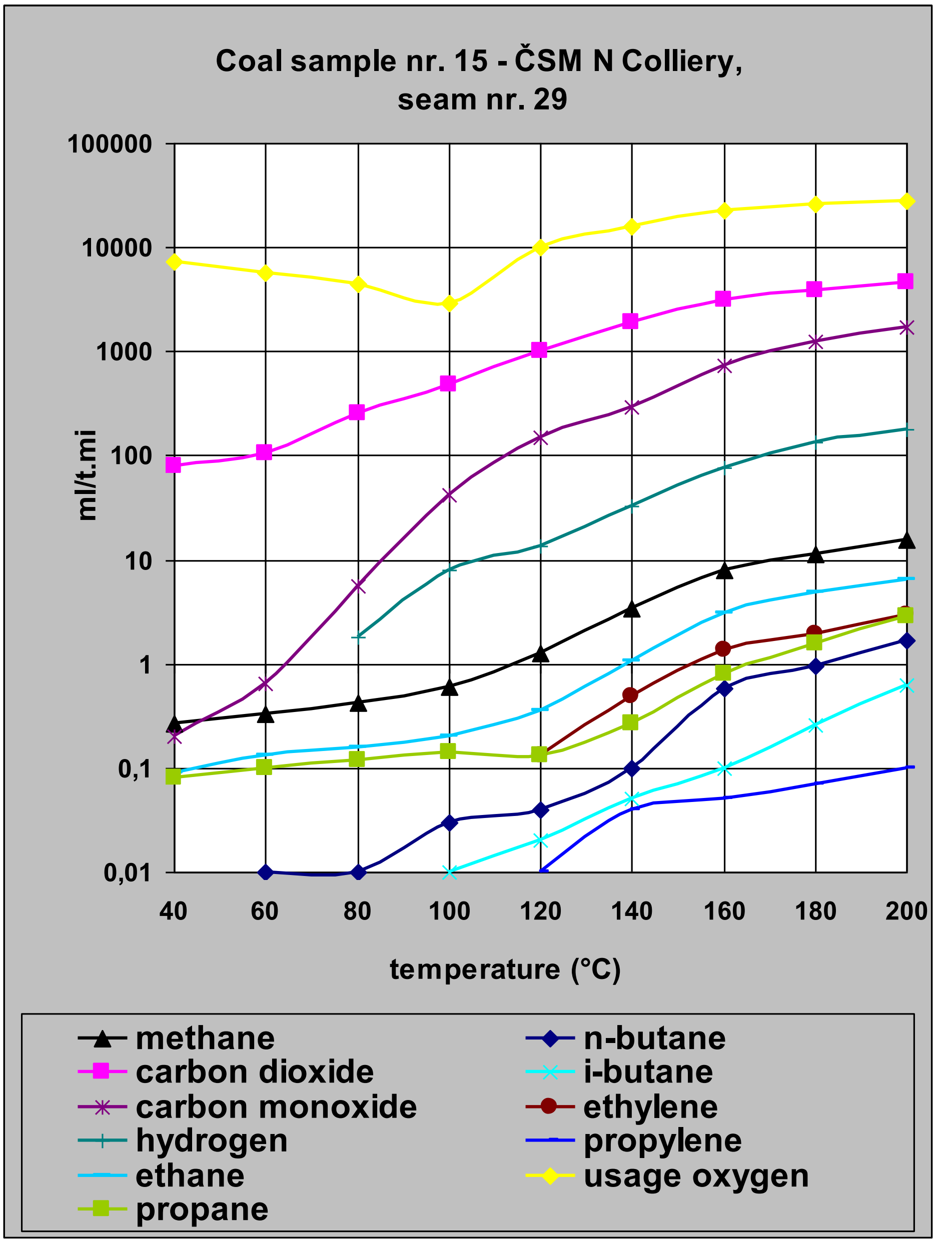

Fig. 5 Evolution of products of thermal oxidation of an OKC coal sample (Adamus, 1999-2002)

4 RESEARCH INTO INDICATOR GASES FOR SPONTANEOUS COMBUSTION OF BROWN COAL IN THE CZECH REPUBLIC

In the early seventies of the 20th century, Hrubý, Charamza and Kusý (Lanková, 1975) from the former Czechoslovak Socialist Republic were concerned with the problems of evolution of gaseous hydrocarbons from hard coals. These authors focused especially on the problems of distinguishing so-called primary methane, physically bound to the coal substance, from secondary methane. During tests they observed that at slight heating the coal (50-55°C), besides methane, iso-butane appeared; the other hydrocarbons, mainly ethene and n-butane were not identified. At temperatures by about 15°C higher (critical temperature of spontaneous combustion), ethane and ethene occurred. Other hydrocarbons began to appear at temperatures of 80-100°C.

The authors Hrubý, Trýzna, Kusý and Hautke (1990) examined, on the basis of research “Determination of a Boundary between the Oxidation Reduction Process and the Initial Stage of Spontaneous Combustion Process. Determination of Indicators of Processes of Coal Spontaneous Combustion”, gaseous indicators of the process of spontaneous combustion in a caved zone of a longwall face. The research was carried out as laboratory research into coal oxidation and as measurement of air masses from the caved zone of the longwall face. In the conclusion, both these activities were evaluated and compared. After the evaluation of the result, the authors arrived at the conclusion that at the beginning of so-called “flushing” of the coal seam, in addition to the release of CO and CO_2, the desorption of methane (from 2 to 100 ppm) also took place. At heating the coal to temperatures of about 50-55°C, when water was released, even iso-butane in concentrations of 0.05-0.1 ppm began to appear besides CO, CO_2 and methane. After the vaporization of a large amount of water, temperature grew and methane content was small. Ethane and ethene began to appear in trace concentrations of about 0.1 ppm. At temperatures of 90-120°C, the proportion of methane increased and C_3 hydrocarbons began to appear. Up to the temperature of 180°C, an increase in the content of iso-butane was not too progressive. At temperatures above 180°C, a steep increase in the concentrations of individual components occurred.

Other findings concerning the indicator gases for the thermal oxidation of brown coal were obtained in the framework of preparation of catalogues of Czech Republic’s coal seams prone to spontaneous combustion, Adamus (1999-2002).

5 CONCLUSION

Czech Republic’s research findings in question are partially used within the meaning of Sections 187, 189 and 194 of Decree of the Czech Mining Authority No. 22/1989 Sb., on safety and health protection at work and operation safety in mining activity and in underground mining of non-reserved minerals, which was in the year 1990 amended for the OKC by the Decision of the District Mining Authority in Ostrava No. 10/1990, the currently valid Decision of the District Mining Authority in Ostrava No. S 0300/2008. At present, Article 27 of the Decision lays down e.g. the classification of coal seams into categories based on the proneness of the coal substance to spontaneous combustion; the localization of places that have to be checked regularly for possible spontaneous combustion of the coal substance; periods and places for CO concentration measurement; determination of frequency of taking air mass samples from the current of air for gas chromatographic analysis (determination of H_2, C_2H_6, C_2H_4, C_2H_2, C_3H_8, C_3H_6, n-C_4H_{10}, iso-C_4H_{10} concentrations); obligations in the case of repeated analysis of gaseous unsaturated hydrocarbons in the sense of closing an endangered area.

In spite of the fact that current technologies enable already improved prediction of spontaneous combustion in coal mines, this problem, especially in the area of determination of temperature condition of the seat of spontaneous combustion, is not solved sufficiently yet. The existing above-mentioned methods enable the estimation of temperature at the seat of spontaneous combustion but they do not enable its exact determination. For these reasons, it is suitable to continue to do the given research. One of activities that deal with the given problems is a project of the Technology Agency of the Czech Republic No. TA01020351, which is just in progress, “Research into Possibilities of Predicting the Occurrence of Initial Stage of Spontaneous Combustion and Subsequent Spontaneous Combustion of Brown Coal Fuels” with duration from 01/ 2011 to 12/2014, the recipient of which is the Brown Coal Research Institute, JSC in Most and a project participant is the Faculty of Mining and Geology of VŠB – Technical University of Ostrava. The project focuses on the use of thermometry and gasmetry in the framework of prediction of spontaneous combustion in brown coal dumping grounds.

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RESUMÉ

Předmětné poznatky výzkumu ČR jsou částečně uplatněny ve znění §187, §189 a §194 vyhlášky ČBÚ č. 22/1989 Sb., o bezpečnosti a ochraně zdraví při práci a bezpečnosti provozu při hornické činnosti a při dobývání nevyhrazených nerostů v podzemí, která byla v roce 1990 pro OKR doplněna rozhodnutím OBÚ č. 10/1990, nyní platné rozhodnutí OBÚ v Ostravě č. S 0300/2008. V článku č. 27 rozhodnutí je v současnosti stanoveno např. členění slojí do kategorií podle náchylností uhelné hmoty k samovznícení; stanovení míst, která musí být pravidelně kontrolována z důvodu možného vzniku samovznícení uhelné hmoty; lhůty a místa pro měření koncentrace CO; stanovení četností odběrů vzorků vzdušin ve větrním proudu pro plynovou chromatografickou analýzu (zjištění koncentrací H_2, C_2H_6, C_2H_4, C_2H_2, C_3H_8, C_3H_6, n-C_4H_{10}, iso-C_4H_{10}), povinnosti při opakované analýze nenasycených plynných uhlovodíků ve smyslu uzavření ohrožené oblasti.

Přesto, že současné technologie umožňují již zdokonalenou predikci samovznícení v uhelných dolech, není tato problematika dosud uspokojivě dořešená, především v oblasti určení teplotního stavu ohniska samovznícení. Dosavadní, výše zmíněné postupy umožňují odhad teploty ohniska samovznícení, nikoliv její exaktní určení. Z těchto důvodu je vhodné pokračovat v daném výzkumu. Jednou z aktivit, která se dané problematice věnuje je v současné době řešený projekt Technologické agentury České republiky č. TA01020351 „Výzkum možností predikce vzniku záparů a následného samovznícení hnědouhelných paliv" s dobou řešení 01/2011 - 12/2014, jehož řešitelem je Výzkumný ústav hnědého uhlí v Mostu a spoluřešitelem Hornicko-geologická fakulta Vysoké školy báňské v Ostravě. Projekt je zaměřen na využití termometrie a plynometrie v rámci predikce samovznícení na deponiích hnědých uhlí.

Článek byl napsán s podporou projektu Technologické agentury České republiky č. TA01020351.