International Journal of Coal Geology 178 (2017) 110–121

Contents lists available at ScienceDirect

International Journal of Coal Geology

journal homepage: www.elsevier.com/locate/coal

Natural content of gases: Carbon monoxide, carbon dioxide, hydrogen and unsaturated hydrocarbons of ethylene, propylene and acetylene in selected bituminous coal seams

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ARTICLE INFO

Keywords:
Coal
Gas
Desorption
Adsorption
Coal self-heating

ABSTRACT

Chromatographic analysis of gas desorbed from coal reveals the presence of carbon monoxide, carbon dioxide, hydrogen, ethylene, propylene and acetylene, i.e. gaseous components liberated into the mine atmosphere with the increasing temperature of self-heating coal. Elevated levels of carbon monoxide, dioxide and ethylene were registered as well. Investigations showed that these gases are contained in the coal structure and their liberation to the mine air can be triggered by other factors apart from coal self-heating, for example coal fragmentation or disturbing of the virgin coalbeds by mining operations. The amounts of thus desorbed gases can alter their actual concentrations in the mine air, which serve as indicators of the self-heating propensity of coal. No straightforward relationship was established between the concentrations of individual gaseous components in the mixed-gas desorbed from coals sample and the rank and other physico-chemical properties of coals.

Sorption and desorption tests by the volumetric method demonstrated that the volume of sorbed carbon monoxide and dioxide, hydrogen, ethylene, propylene and acetylene is closely related to the coal rank and its porous structure. Low-rank coals with well-developed pore systems tend to absorb/adsorb greater amounts of gases than low-rank coals with a less developed pore structure.

Tests established the intensity of gas desorption from coals, showing that nearly 80–90% of carbon monoxide, dioxide and hydrogen accumulated in coals would be released. In the case of adsorbed ethylene, propylene and acetylene that proportion is the lowest (50%–70%).

Recalling the Ideal Adsorbed Solution (IAS) theory, simulations were performed to predict the mixed-gas adsorption isotherms for binary gas mixtures containing non-saturated hydrocarbons-gas components that are indicators of the self-heating propensity of coal. It appears that acetylene adsorption in the systems $C_2H_2{-}C_2H_4$ and $C_2H_2{-}C_3H_6$ is a favoured process whilst preferential adsorption in the system $C_3H_6{-}C_2H_4$ is that of propylene.

1. Introduction

Coal self-heating events, quite common in collieries, are the consequence of accumulation of heat generated in the course of coal reactions with air. Uncontrolled processes give rise to endogenous fires, posing a threat to miners' safety and generating financial losses (Kaji et al., 1985; Cygankiewicz, 2000; Rosema et al., 2001; Dai, 2007). Reliable assessment of the coal self-heating rate allows appropriate measures to be put in place in order to control the process and to minimise the risk of an endogenous fire. It is recalled that gases released to the mine atmosphere as a result of coal oxidation, such as carbon monoxide, hydrogen and unsaturated hydrocarbons: ethylene, propylene, acetylene, can serve as index gases i.e. indicators of the coal self-heating rate (Wacławik et al., 2000; Lu et al., 2004; Singh et al., 2007; Xiao et al., 2008; Adamus et al., 2011). Their concentrations tend to increase with temperature of the self-heating coal, which can be utilised to determine the temperature of coal and the scale of development of the self-heating process. Among those gases, carbon monoxide content in mine air is the most widely used indicator of the coal self-heating rate. In accordance with the relevant mining regulations in Poland, continuing carbon monoxide concentrations above 26 ppm in registered in the airflow are considered to be the indicators of an underground fire (Regulation by the Ministry of Economy 2002).

In some cases the carbon monoxide concentrations in mine air will be elevated though no fire hazard occurs, which can be confirmed by specialised techniques (infrared cameras, pyrometers). Besides, it is a

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http://dx.doi.org/10.1016/j.coal.2017.05.005
Received 20 September 2016; Received in revised form 8 May 2017; Accepted 8 May 2017

Table 1
Characteristics of coal samples.

Properties	Symbol	Unit	Samples from the following mines					
			Staszic seam 407	Brzeszcze seam 510	Marcel seam 505	Bielszowice seam 405	Borynia seam 407	Marcel-1 seam 712
Carbon	C^{daf}	%	83,19	84,70	84,07	86,21	86,07	86,68
Sulphur total	S^{daf}	%	0,64	0,38	0,32	0,48	0,71	0,24
Hydrogen	H^{daf}	%	5,01	4,62	4,44	4,94	5,10	5,19
Nitrogen	N^{daf}	%	1,39	1,40	1,40	1,56	1,49	1,50
Oxygen (calculated)	O^{daf}	%	10,14	8,98	9,99	7,14	6,79	6,57
Moisture	W^{a}	%	3,73	1,61	1,19	1,57	1,23	1,77
Ash	A^{a}	%	8,45	5,82	7,69	9,20	27,45	4,44
Volatile matter	V^{daf}	%	32,38	32,48	32,94	31,52	24,87	34,96
Reflexivity of vitrynite	R_0	%	0,81	0,85	0,89	0,92	1,12	0,90
Wirtynite	V	%	61	62	67	63	67	68
Egzynite	L	%	10	7	2	6	1	11
Inertynite	I	%	29	31	31	31	32	21
Mineral substance	MM	%	2	1	4	3	16	1

well-established fact in coal mining that carbon monoxide occurs naturally in coalbeds. In holes drilled in coalbeds the registered levels of carbon monoxide may exceed 1% (Wacławik et al., 2000), which suggests that gases released from coal seams for yet unknown reasons can be liberated to mine air by way of desorption. Elevated carbon monoxide concentrations in mine air are often registered during the coal mining operations involving coal fragmentation and crushing, which enhances the gas release. In certain cases, however, elevated hydrogen concentrations were registered in mine air, though these were not the consequence of coal self-heating. In lesser degree will the concentrations of unsaturated hydrocarbons be increased in the absence of coal self-heating.

Literature on the subject abounds in reports on desorption of carbon dioxide from coals (Ceglarska-Stefańska, 1998), whilst little published literature is available on desorption of the remaining gases from coals. In his work (Davidi et al., 1995) established that ethylene desorption from coal occurs only during the oxidation of coal. Another study investigating carbon monoxide desorption on coals in the conditions of reduced gas pressure above the coal sample (Cygankiewicz et al., 2007) emphasises the irreversibility of carbon monoxide sorption. Published literature on desorption of hydrogen contains the work by (Street et al., 1975) investigating hydrogen liberation during the crushing of coal prior to its combustion in coal-fired power plants and suggesting that hydrogen release may be due to desorption occurring when coal blocks were broken. According to several researchers (Grossman et al., 1994; Marzec and Czajkowska, 2005; Marzec, 2006), hydrogen emission from coals is mostly the result of bituminous coal oxidation.

In consideration of the fact that concentrations of carbon monoxide, carbon dioxide, hydrogen and unsaturated hydrocarbons are commonly used in assessment of the state of development of coal self-heat, tests on desorption of these gases on coal seem fully merited.

Most pertinent issues to be considered are as follows:

- Are these gases naturally bound to the coal structure and whether their desorption to the mine air should be possible when no self-heating of coal occurs?
- Is the actual process of desorption of those gases on coal related to the type and physical and chemical properties of coals?
- Which gas shows a stronger propensity for desorption?

Test results can be utilised to improve the existing methods of fire risk assessment in mines.

2. Experimental

2.1. Samples

Testing was done on six coal samples from Polish collieries (Borynia seam No 409, Staszic seam No 407, Brzeszcze seam No 510, Marcel seam No 505, Bielszowice seam No 405 and Marcel-1 seam No 712) revealing elevated carbon monoxide or hydrogen concentrations in the absence of the coal self-heating. Selected coal samples were obtained directly from the boreholes drilled in the coalbeds in the mine. Coal samples were collected from the two boreholes in one coal seam, distant by no more than 1 m, drilled to the depth 1.5–2 m directly before sampling and with no air blow. When the borehole depth reached 1.5 m, the drilling was interrupted and the borehole was cleaned. An airtight steel container with steel balls was then provided under the borehole outlet and the drilling was recommenced to collect samples (i.e. drill borings). Thus obtained drill borings were blown off with an inert gas - helium, until the oxygen content fell below 2% at the outlet of the container. Coal samples in the containers were shaken for 2 h in the laboratory conditions to get them crushed. Afterwards, gas was collected from coal samples for chromatography analysis to determine the contents of carbon monoxide, carbon dioxide, ethylene, propylene and acetylene, also of oxygen, methane, ethane and propane. To determine the hydrogen content, a coal sample was placed inside a separate container and blown off with nitrogen since the hydrogen content is most difficult to determine in the presence of large amounts of helium (similar peak retention times of the helium and hydrogen registered in gas chromatography).

Coal samples from the boreholes were also collected for chemical, technical and petrographic analysis of coals, in accordance with the procedure set forth in relevant Polish standards. Results are summarised in Table 1. Surface areas of analysed samples were calculated according to the Dubinin – Radushkevich model basing on the carbon dioxide sorption isotherms obtained at 298 K, and according to the BET model based on nitrogen sorption isotherms obtained at 77.5 K. Sorption isotherms of carbon dioxide and nitrogen were determined by the volumetric method with the use of the Micromeritics ASAP 2010 apparatus. The results are summarised in Table 2 alongside the micropore volumes calculated recalling the Dubinin-Radushkevich model, basing on sorption isotherms of carbon dioxide.

2.2. Chromatography of gas desorbed from coal

Gas desorbed from the coals was subjected to gas chromatography analysis, involving the separation of gas mixture components between the stationary phase (column packing) and a mobile phase (carrier gas), followed by their elution from the chromatography column by the