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Research paper

Tests of methane desorption and emission from samples of hard coal in the context of mine closures through flooding



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ARTICLE INFO	A B S T R A C T
Keywords: Methane hazard Closing mines Flooding mines Methane emission Safety Underground mining	Forecasts of methane emissions during and after flooding a closed gassy hard coal mine and the evaluation of possible methane migration to the surface in post-mining areas, after cutting off the vertical ventilation workings of hard coal mines from the surface, provide valuable information which can help to ensure public safety. This article presents research into the influence of changes in the hydrostatic pressure of a water column in a flooded mine on the volume of methane emission and migration from hard coal seams, during and after the flooding of a closed mine. The tests were conducted based on a modified research method developed by the French National Institute for Industrial Environment and Risks (INERIS), France, and the Central Mining Institute (GIG), Katowice, Poland. A test stand for gas desorption and autoclaves for emissions, under controlled pressure and temperature, were used. The tests were conducted and changes in pressure in the autoclaves over time were observed. The observations led to the conclusion that water inhibits methane desorption and emission from coal to varying extents, depending on the hydrostatic pressure exerted. Based on the conducted tests, developed a model of methane emission into flooded goafs was developed. A method of determining index k_2 was also developed, which lowers the forecast volume of methane emission into goafs depending on the value of the hydrostatic pressure of the water column and the level of submersion. Results of the tests form the basis to calculate forecasts in the developed model of methane emission into the level of methane hazard and the selection of preventive measures aimed at combating methane hazard during and after the closure of a gassy mine.

1. Introduction

Closing industrial facilities always has consequences which, to differing degrees, influence both the environment and public safety. The period after the closure of the facilities located on the surface, in most cases, is associated with reclaiming the area and managing industrial waste. The situation looks different when underground facilities, such as coal mines, are closed. Hazards associated with conducting long-term mining operations have a long-term influence on the environment after mining operations have finished. To prevent negative consequences associated with the end of mining operations, it is necessary to identify the emerging hazards, analyze them and assess the risks linked with the mine closure and the post-closure period, in order to minimise them (Duda & Krzemień, 2018; Laurence, 2011).

Methane emission into goafs from operating longwalls, and after their exploitation, influences how the level of methane hazard is shaped in the underground workings of a closed mine. The volume of methane emission into goafs depends on the volume of the destressed coal deposit, its natural methane bearing capacity within the range of the destressed zone and the amount of time which has passed from the end of mining operations (Krause, 2009). Methane emission from unmined seams, which were destressed as a result of mining works, usually lasts for 15–20 years (Krause & Pokryszka, 2013).

Methane saturation of hard coal seams, i.e. their methane bearing capacity in mining areas of hard coal mines, influences the level of methane hazard both during mining works in gassy seams and after they have been completed.

Coal seam methane bearing capacity exceeding $2.5 \text{ m}^3 \text{ CH}_4/\text{Mg}_{csw}$ is the threshold of the occurrence of dangerous methane concentrations in longwalls during and after mining operations (Krause & Łukowicz, 2000).

Methane saturation of a seam over the aforementioned value of methane bearing capacity creates the conditions for the occurrence of explosive and superexplosive methane concentrations and the migration of methane towards the surface in post-mining areas of a hard coal mine during its closure.

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The problem of gas migration to the surface in post-mining areas is one of the main aspects of risk assessment, especially in gassy hard coal mines which are closed (Krzemień, Suárez Sánchez, Riesgo Fernández, Zimmermann, & González Coto, 2016). Migration of mine gases to the surface of post-mining areas intensifies when shafts are closed and the ventilation stops. It causes the accumulation of gases in workings and goafs. In the underground structure of a closed mine, there is laminar displacement of gases under the influence of pressure gradient, temperature and natural depression. This process goes consists of two stages. In the first stage, the gas desorbs from coal. In the second stage the gas flows freely into mine workings and goafs. Further stages associated with migration to the surface depend on gas permeability (Zawisza, Macuda, & Chećko, 2005) and the method of mine closure. Gas desorbing into a mine may be emitted on the surface through

(Pokryszka et al., 2005):

- old mining works which connected a mine with the surface (shafts, adits, mine workings), which are individual points of gas emission if they are not properly sealed,
- overburden, especially in the areas where it is thin and its permeability is the biggest. This results from the fact that a thin layer of overburden is naturally permeable (high primary permeability, cracks) or it is permeable as a result of mining works,
- and, to a much lower degree, through water or mine drainage water (dissolved gas).

Taking into consideration the number of environmental events and incidents which took place in the Czech Republic (Dvořáček & Slivka, 2004), the United Kingdom (Broughton, 2014) and Germany (Heitfeld, Rosner, Mühlenkamp, & Sahl, 2004), it is highly desirable and advisable to take steps aimed at limiting environmental hazard and ensuring public safety. The literature describes numerous accidents and incidents associated with the emission or collection of mine gases on the surface of post-mining areas (Díaz Aguado & González Nicieza, 2007; Jackson, 2000; Kral, Paletnik, & Novotny, 1998; Novotny, Platenik, Takla, & Kral, 2001; Pokryszka & Tauziède, 2000; Robinson, 2000).

The issues presented in this article were analysed within the framework of an international project, financed by the Research Fund for Coal and Steel, named MERIDA (Krzemień et al., 2016, 2017) and realized by the Central Mining Institute and its foreign partners from Spain, France, Germany, the United Kingdom and the Czech Republic.

The article consists of an introduction which presents the issues associated with methane emission from unmined seams, which are destressed as a result of the mining work conducted, and its migration to the surface of post-mining areas. Then, it describes the research area which was analysed and tested and the research methodology applied in the laboratory. The article goes on to present the model of methane emission into goafs which were not flooded, developed by Krause and Łukowicz (2000), and a new model developed within the framework of the MERIDA project, concerning the forecasts of methane emissions into partially or completely flooded goafs, which is presented for the first time in the article. The final section presents the results of laboratory tests which formed the basis for the determination of the characteristics of index k_2 , lowering the forecast volume of methane emission into goafs depending on the value of the hydrostatic pressure of the water column. At the end, the final results are discussed in relation to hitherto approach to the phenomenon in the literature.

The model of methane emission into flooded goafs presented in the article is applicable and enables forecasts of the volume of methane desorbing into goafs when they are partially or completely flooded. Therefore, providing valuable information during mine closure and when public safety work on the surface is planned.

2. Materials and methods

2.1. Research area

The Rydułtowy hard coal mine is the oldest operating mining enterprise in Upper Silesia, dating back to 1792. In 2004, the Rydułtowy coal mine was connected via a ventilation network = with the Anna coal mine, as exploitation was conducted within the borders of the mining areas of both mines. After merging the mines, mining operations were conducted in seams 703/1, 706 (707/1-2), 713/1-2 and 712/1-2 of Ruch Rydułtowy (Rydułtowy coal mine) as well as 707/1-2 (706) and 713/1–2 of Ruch Anna (Anna coal mine). The closure of Ruch Anna started in 2006 and gradually progressed by liquidating the ventilation infrastructure. Securing work also included drainage which connected the mining operations of both mines with a selected mine working in seam 713/1-2. Mine water from the natural inflow of Ruch Anna flowed to the dewatering system of Ruch Rydułtowy. In the 2nd half of 2017, shafts Chrobry I and Ryszard II were backfilled and, at present, there is no ventilation connection between the underground mine workings of the former Anna mine and the mine workings of the Rydułtowy mine. This resulted in changes in the migration of mine gases, particularly of methane, and may pose a threat to the surface infrastructure (Duda & Krzemień, 2018).

The research project included tests conducted in the Rydułtowy-Anna coal mine where the Ruch Anna mining area was closed. The level of methane saturation of the coal deposit in the mining area of Ruch Anna is slightly lower than in the deposit of Ruch Rydułtowy. The existing hydraulic connection between the closed mine Anna and the operating mine Rydułtowy, creates the conditions for the partial flooding of the mining area of the closed Ruch Anna. The total height of the water column in the closed hard coal mine will be 78 m, thus contributing to limiting methane migration from the degassing seams which were destressed during prior mining operations in Ruch Anna.

The laboratory tests conducted on coal samples collected from the seams which were mined in Ruch Anna enabled the determination of the kinetics and tendencies of changes in methane emission under the influence of the hydrostatic pressure of a water column between 0 and 7.8 bar. The chosen range corresponds to the rising water level in the flooded goafs of Ruch Anna. The tests conducted enabled the evaluation of methane emission into Ruch Anna after it was partially flooded. The article presents results of the conducted laboratory tests on the volume of methane emission from coals under water which are subjected to the influence of hydrostatic pressure of between 0 and 7.8 bar.

2.2. Research methodology

The tests presented in the article were carried out to determine the influence of changes in the hydrostatic pressure of the water column on the intensity of methane emission from coal samples collected in seams 703/1 and 705/2–3. Results of the tests form the basis to calculate, with the developed model, forecasts of methane emission into the goafs of a closed mine.

The experiments were conducted using a stand for desorption and an autoclave for gas emission tests under controlled pressure and temperature. The technical parameters and the conditions of the process as well as the technical parameters of the tested coal samples are presented in Table 1. The parameters of hydrostatic pressure mapped a change in the conditions in flooded coal seams of Ruch Anna.

The preparation stage, before conducting the tests, involved the selection of an appropriate high level of methane saturation for the samples so that the tests were conducted in conditions reflecting *in situ* ones. In order to do this, a number of tests were run to select the proper

Table 1

Technical parameters and conditions of gas desorption and emission in laboratory conditions.

Technical parameters and test conditions				
Temperature	40 °C			
Methane pressure	5 bar			
Time of saturating sample with methane	approx. 7 days (until equilibrium)			
Set hydrostatic pressure	0.0; 0.4; 0.8; 2.4; 4.8; 7.2; 7.8 bar			
Height of water column over sample	1 cm			
Weight of sample	100 g			
Time of degassing sample in vacuum	24 h			
Grain size of sample	0.5–1.0 mm			
Methane bearing capacity of saturated sample	5.93 m ³ CH ₄ /Mg _{csw}			

pressure for saturating a coal sample with methane and the proper saturation time, and to determine the losses of gas after opening the autoclave, until a sample is under water. The actual laboratory tests were conducted in two stages: the samples preparation; observation of methane desorption from a submerged coal sample depending on the pressure exerted on the water layer. To avoid errors associated with the differences in the structure and sorption properties of coal, the same sample was used for each variant of the tests.

The tests were conducted according to the following methodology:

- Coal samples of 0.5–1 mm grain size were prepared and degassed in a vacuum for at least 24 h.
- Coal samples were placed in the autoclave and saturated with methane under constant pressure of 5 bar and temperature of 40 °C.
- After saturating a coal sample and reaching the state of equilibrium (taking approximately 7 days). The autoclave was opened and 100 ml of water was added, forming a 1 cm layer of water over the sample.
- The autoclave was closed and sealed approximately 1 min after the sample was covered with water. The determined value of the methane bearing capacity of a sample prepared in such a way was 5.93 m³ CH₄/Mg_{csw}, which was recreated for each of the further test variants.
- Compressed air was pumped into the autoclave, which was transferred onto a sample in the form of hydrostatic pressure of the values: 0.0; 0.4; 0.8; 2.4; 4.8; 7.2; 7.8 bar.
- Observations of methane desorption were made through measuring an increase in pressure in the autoclave over time, until the system stabilised completely (between 14 and 30 days).

Fig. 1 presents a schematic view of the course of a test, considering both aforementioned stages.

2.2.1. Model of methane emission into unflooded goafs

The tests concerning methane emission from destressed deposit into goafs of operating and closed longwalls, conducted by the Central Mining Institute (GIG), enabled the development of a model to forecast the volume of methane desorbing into goafs (Krause & Łukowicz, 2000). After the end of mining operations, methane desorbs from destressed seams of secondary methane bearing capacity into goafs for approximately 15 years (Krause, 2008; Krause & Łukowicz, 2000). The model assumptions refer to three stages with different courses of methane emission:

- 1. Mining stage: absolute methane bearing capacity increases to the maximal value of methane emission during mining operations for given gas conditions of an exploited seamand its surroundings.
- 2. Longwall closure stage: this includes the time necessary to prepare and close a longwall (approximately 3 months) when absolute methane bearing capacity drops to 20–40% in comparison with the mean absolute methane bearing capacity of the mining stage.



Fig. 1. Schematic diagram considering the sample preparation stage of the course of a test of methane emission and desorption from coal samples submerged under water, under the influence of hydrostatic pressure.

Stage after longwall closure: includes time when methane emission into goafs declines (approximately 15 years).

The model of methane emission into goafs after the end of mining operations and a longwall is closed enables the determination of the total value of methane emission after mine closure for a period of 15 years after the completion of mining operations (Krause & Łukowicz, 2000):

$$\dot{V}_M = \sum_{i=1}^n \dot{V}_{G_i} \tag{1}$$

where:

 \dot{V}_{M} – forecast volume of methane desorbed from destressed seams into goafs from *n* longwalls in consecutive years after the completion of mining operations [m³ CH₄/min],

 \dot{V}_{G_i} – forecast volume of methane emission into goafs of *i*-th longwall in consecutive years after the completion of mining operations [m³ CH₄/min],

n- number of longwalls mined in consecutive years over the 15 years after the completion of mining operations.

$$\dot{V}_{G_i} = 0.2 \, \dot{V}_A \left(1 - \frac{u}{15} \right)$$
 (2)

where:

 \dot{V}_A – mean value of absolute methane bearing capacity of *i*-th longwall during the mining period [m³ CH₄/min],



Fig. 2. Model of methane emission from goafs after the completion of mining operations (Krause & Łukowicz, 2000).

u – number of years since the completion of mining operations in i-th longwall.

flooded goafs is calculated with the dependence:

2.2.2. Model of methane emission into flooded goafs

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The model of forecast methane emission into flooded goafs was developed within the framework of the international project financed by the Research Fund for Coal and Steel, named MERIDA (management of environmental risks during and after mine closure).

The tests conducted on methane emission from coal under the hydrostatic pressure of a water column enabled the identification of the influence of variable values of pressure on methane emission. The forecast volume of methane emission into completely or partially

$$\dot{V}_{GW} = \dot{V}_{G_l} k_1 k_2 + \dot{V}_{G_l} (1 - k_1)$$
(3)

where:

 \dot{V}_{G_i} – forecast methane bearing capacity of goafs of *i*-th longwall, k_1 – index of the surface area of flooded goafs of *i*-th longwall, k_2 – index of methane emission into goafs of destressed seams within the destressed zone depending on the pressure of the water column.

Index k_1 of the surface area of flooded goafs of the exploited longwall assumes the values of 0-1 and is calculated as follows:

$$k_1 = \frac{P_w}{P_G} \tag{4}$$

The surface area of goafs P_G is determined based on the product of the length of the exploited longwall and its width:

$$P_G = L w [m^2] \tag{5}$$



Fig. 3. Area of flooded sections of goafs.

where:

L – longwall width [m], w - length of exploited longwall [m].

The surface area of flooded goafs $P_{\rm m}$ is calculated for two mining systems. For the transverse mining system:

$$P_w = L w_1[m^2] \tag{6}$$

 w_1 – length of exploited and flooded longwall [m].

For the longitudinal mining system:

 $P_w = w (L - l) [m^2]$

where

l – unflooded longwall width [m].

The value of index k_2 is calculated based on the dependence determined based on the tests conducted for the MERIDA project.

3. Results and discussion

In the literature there is information concerning water-inhibited methane emission and emission from hard coal seams (Krause & Pokryszka, 2013; Pokryszka et al., 2005). Coals can be treated as methane reservoirs, which are typically characterized by dual porosity systems, with gas in the cleats and absorbed gas in the matrix (Cienfuegos & Loredo, 2010). So far, the existing mechanisms and the quantitative aspect of the phenomena have not been recognised and described in detail. The aim of the conducted tests, described in this article, was two-fold: to prove the thesis that water inhibits methane emission and to develop and determine a dependence for index k_2 , which lowers the forecast volume of methane emission into goafs, which is connected to the value of the hydrostatic pressure of the water column and the flooding depth in coal mines. The in situ conditions (temperature, hydrostatic pressure during mine flooding and methane

bearing capacity value) in the vicinity of seam 713/1-2 were modelled in a laboratory.

The tests were conducted for pressure of 0.0; 0.4; 0.8; 2.4; 4.8; 7.2; 7.8 bar, observing the phenomenon of methane emission from coal samples submerged in water in the autoclave. The samples were collected in seams 703/1 and 705/2-3. Results of the analyses conducted for all the variants are presented in Fig. 4.

The first conclusion which can be drawn after analysing the results is that the presence of water and hydrostatic pressure exerted on a coal sample limits methane emission. The higher the pressure, the more distinct the phenomenon. Different times needed for stabilising pressure in an autoclave, depending on the pressure set over the water laver, was also observed. To compare all the variants of tests, the mean time of terminating the experiment was assumed to be 25000 min (approximately 17 days). In each case, this meant the time when the pressure in an autoclave stabilised. The maximal value for the pressure increase axis was assumed to be 4.5 bar.

Based on the obtained test results, it was concluded that coal samples collected from seams 703/1 and 705/2-3 show similar properties which are associated with methane desorption and emission, hence the internal structure and sorption properties of both samples are similar. Due to this observed tendency, further discussion will be based on a general phenomenon, unrelated to the place where the samples were collected.

At the initial stage of the tests, a rapid increase in pressure was observed in the autoclave for each of the variants. Methane desorption was inhibited to different degrees, depending on the pressure exerted by the layer of water. In some cases it stabilised after approximately 2500 min, and in others after as long as approximately 22000 min.

The presented results of the tests show that water is able to significantly limit methane emission from coal, and the inhibition effect increases together with the hydrostatic pressure affecting a coal sample. The tests also confirmed Krause and Pokryszka's thesis (2013) concerning the inhibition of methane emission from coal by water, where the index of methane emission from coal.

W was expressed with the following dependence:

$$W = \frac{V_w}{V_{\text{des}}} \tag{8}$$



(7)

Fig. 4. Dependence of total increase in pressure in autoclaves on time, for coal samples collected from seams 703/1 and 705/2-3, for various variants of set hydrostatic pressure.



Fig. 5. Dependence of increase in pressure in the autoclave on time, for coal sample under hydrostatic pressure of 15 bar.

where:

 V_w – volume of methane desorbed from a given coal sample,

 V_{des} – volume of desorbed methane when the pressure of gas drops from the given initial pressure to atmospheric pressure.

To draw conclusions concerning the inhibition of methane emission from coal, the values of pressure increase in the autoclave was focused on, unlike Krause and Pokryszka (2013) who considered methane emission in relation to volume. The joint conclusion is confirmation that water inhibits methane emission from coal.

It was proven that the maximal value of hydrostatic pressure which may be set to inhibit methane emission from a coal sample is limited. To prove this, a test was conducted in which the value of pressure exerted on the water layer was 15 bar. The results of the test are presented in Fig. 5. The graph shows that the pressure in the autoclave constantly increased, even after reaching the prior assumed time of 25000 min, after which, in each of the variants, the system reached equilibrium and the pressure in the autoclave stabilised. Based on the conducted tests, it may be concluded that too high pressure triggered a "piston effect", which gradually displaced methane which was sorbed within the coal structure.

Dimensionless index k_2 which lowers the forecast volume of methane emission into goafs, depending on the value of the hydrostatic pressure of a water column, determined based on the conducted tests, assumes the values of 1–0. The value of 1 means free desorption and uninhibited emission, while 0 means total inhibition of methane emission from coal. Based on the tests, index k_2 is calculated with the dependence:

$$k_2 = \frac{\Delta p_e}{\Delta p_t} \tag{9}$$

 k_2 – dimensionless index which lowers the forecast volume of methane desorption into goafs depending on the value of the hydrostatic pressure of a water column,

 $\varDelta p_e$ – total increase in pressure in the autoclave during methane desorption from a coal sample submerged in water, under the influence of hydrostatic pressure of a specific value [bar],

 Δp_t – total increase in pressure in the autoclave during free desorption of methane from a coal sample without set hydrostatic pressure [bar].

Table 2 Values of index k_2 depending on set hydrostatic pressure.

Set hydrostatic pressure [bar]	Total increase in pressure in autoclave [bar]	Value of index k 2
0	4.069	1.00
0.4	3.486	0.86
0.8	2.358	0.58
2.4	1.555	0.38
4.8	0.898	0.22
7.2	0.642	0.16
7.8	0.584	0.14

Table 2 presents the values of index k_2 calculated using formula (9) for the results obtained in the tests.

Table 2. The tests presented in the article enabled the updating of information on the assessed volume of methane emission into the closed Ruch Anna after it was partially flooded.

4. Conclusions

It may be concluded that the pressure exerted on a coal sample in the form of hydrostatic pressure determines how much methane desorption and emission from coal are actually inhibited by water.

The hydrostatic pressure affecting a coal sample has its threshold until which it inhibits methane desorption and above which water triggers a "piston effect" gradually displacing the methane sorbed in coal.

The developed index k_2 , which lowers the forecast volume of methane emission into goafs depending on the value of the hydrostatic pressure exerted on a coal sample, may be applied to verify the forecasts of methane emission into flooded goafs.

When goafs are flooded, the pressure of the water column constitutes a barrier which limits the forecasted inflow of methane into the goafs.

Based on the model of methane emission into goafs, when they are partially or completely flooded, it is possible to verify the volume of methane emission into goafs, providing valuable information during mine closure works and the planning of public safety on the surface.

Conflicts of interest

None declared.

Ethical statement

The research was conducted according to ethical standards.

Appendix A. Supplementary data

Supplementary data to this article can be found online at https://doi.org/10.1016/j.jsm.2019.03.005.

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