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DETERMINATION AND ANALYSIS OF METHANE EMISSION FACTOR FOR OPEN-PIT LIGNITE MINES

Atmospheric methane emissions from the energy sector, particularly coal mines, are a component of total global methane emissions. Its presence causes climate changes that contribute to global warming. Methane is a potent greenhouse gas (GHG) with a global warming potential (GWP) approximately 30 times greater than that of carbon dioxide over a 100-year period. The estimation of methane emissions from both underground hard coal mines and open-pit lignite mines is performed based on guidelines that follow the methods recommended in the core publications of the International Panel on Climate Change (IPCC). The methane emission rate determination method developed by scientific institutions is allowed. This article focuses on the analysis and formulation of guidelines for determining the potential methane emissions from open-pit lignite mines, which are determined based on the emission factor and coal production. Coalbed methane content was tested using two methods to determine the methane emission factor. Results of sorption tests were also presented. The results obtained can be used for the development of new solutions or the improvement of current solutions for the determination of the methane emission rate in open-pit lignite mines.

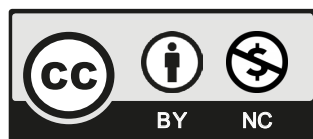
Keywords: Methane emission; open-pit mine; greenhouse gases; sorption properties; methane content

1. Introduction

Methane is a potent greenhouse gas (GHG) with a global warming potential (GWP) approximately 28-35 times greater than that of carbon dioxide over a 100-year period [1-2]. This makes it the second most important greenhouse gas, as well as a precursor to the formation of harmful tropospheric ozone (O₃), which is responsible for about a quarter of the global warming

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effects that are currently being observed [3-5]. Some studies indicate an 80 times higher global warming potential for methane than carbon dioxide over a 20-year time horizon [6]. Methane is a valuable energy source that offers the potential to reduce global greenhouse gas emissions when used [7,8]. Current efforts by the European Union and the European Parliament are aimed at reducing greenhouse gas (GHG) emissions, including methane, into the atmosphere, which have been identified as one of the main causes of climate change [9-11]. Under the European Union strategy to reduce methane emissions and the “European Parliament resolution of 21 October 2021 on an EU strategy to reduce methane emissions (2021/2006(INI))” [12], efforts are being made to analyse the above phenomenon and reduce the effects of its negative impact. Research by the International Energy Agency (IEA) shows that methane leaks from mines have a huge impact on global climate change [13].

The analysis of the phenomena describing the transport of gas in the coal structure (coal-gas system) is crucial in the context of reducing its emission to the atmosphere as well as the possibility of energy use of coalbed methane [14]. Coal mines are one of the largest sources of anthropogenic methane emissions [15-17]. Coal production releases methane that has accumulated in the coal seams and surrounding layers [18]. According to 2019 data, the coal mining industry is estimated to account for 11% of global methane emissions from human activities [19]. However, many scientists argue that current projections of methane emissions from fossil fuels may be underestimated. For example, it has been stated that methane emissions from fossil fuels could be 60 to 110% higher than current estimates [20], or another study found that global methane emissions could be 1.5 times higher than estimated in a US inventory study [21]. Although the above studies do not focus exclusively on coal mines, they show that inventories may underestimate methane emissions from fossil fuels. According to the authors of this article, some emission projections may also be overestimated in some cases, particularly in the case of open-pit lignite mines.

Methane occurring in coal-bearing formations is produced by microbiological processes, which can be divided into early and late, as well as thermogenic processes. In the early stage of coalification of plant material (peat formation and diagenesis), methane is produced by microorganisms [22-23]. The methane content of lignite coals is much lower than that of higher coalified formations. This is due to the lower productivity of microbial processes and the increasing catagenetic efficiency [24]. Therefore, methane emissions tend to be higher in underground mines than in open-pit mines because the lower-lying coal seams contain more methane [25]. Despite this, even the small amount of methane emitted during mining operations represents – in the context of a specific time interval and a specific amount of coal production – a certain amount of emissions. Until now, open-pit lignite mines have not been required to determine methane content and determine the amount of its emission into the atmosphere. Attempts have been made to determine methane emissions for lignite mines in various countries such as China, Australia, Colombia, Poland, Germany, and the USA [4]. Methane emissions from lignite deposits cannot be monitored online because of the way an open pit mine is operated. Determination of the methane emission rate for lignite mines requires studies of the methane content and sorption of a given seam or deposit, analysis of the phenomena occurring, as well as mining and geological conditions. The above-mentioned operations and research were performed as part of this article. Coalbed methane content tests were carried out using two direct methods: single-phase vacuum degassing (SPVD) and the United States Bureau of Mines (USBM). In addition, sorption tests were performed to determine the kinetics of methane sorption, i.e. the rate at which sorption

processes occur. Based on the analysis of the obtained research results and mining and geological conditions, it was possible to recommend guidelines for determining the methane emission rate for open-pit lignite mines. At present, no method in Poland specifies how to directly determine the methane content of brown coal, and there is no model available for determining the methane emission rate for open-pit lignite mines.

2. Materials and methods

2.1. Research material

The research material for this analysis was collected at two locations within a lignite deposit in Poland. At each of the selected locations, 5 samples were taken for methane content analyses by the single-phase vacuum degassing (SPVD) method, 5 samples for the United States Bureau of Mines (USBM) method, and 5 samples for sorption tests. A total of 20 methane content analyses and 10 sorption tests were conducted. Due to the confidentiality of the data presented in the article, the samples were identified as in TABLE 1 below, adopting numbering from 1 to 10 and signatures corresponding to the method, i.e. X (SPVD) and Y (USBM). Sorption tests were performed for samples identical to those taken for methane content tests, i.e. for samples 1 through 10.

TABLE 1

Summary of samples collected for testing

Research area	Sample number		Methane content determination method
I	1	X	SPVD
		Y	USBM
	2	X	SPVD
		Y	USBM
	3	X	SPVD
		Y	USBM
	4	X	SPVD
		Y	USBM
	5	X	SPVD
		Y	USBM
II	6	X	SPVD
		Y	USBM
	7	X	SPVD
		Y	USBM
	8	X	SPVD
		Y	USBM
	9	X	SPVD
		Y	USBM
	10	X	SPVD
		Y	USBM

2.2. Mining and geological conditions

Analysing geological and mining conditions is essential for research on methane emissions from lignite open-pit mines. Poland's industrial economic lignite resources, as of December 31, 2019, amounted to 994.55 million tonnes. Much higher are the anticipated geological resources, estimated at 23.26 billion tonnes. In Poland, 91 lignite deposits have been recognized, of which 8 were in production in 2019: Adamów, Koźmin, Bełchatów – Bełchatów and Szczerców Field, Pątnów IV, Drzewce, Tomisławice, Turów and Sieniawa 2 [26].

To illustrate the geological conditions for the article, a description of the Turów lignite deposit was used hypothetically. The authors do not indicate that the samples analysed in this article are from this mine. The analysis uses, among other sources, data posted on the website of “PGE Górnictwo i Energetyka Konwencjonalna Spółka Akcyjna – Oddział Kopalnia Węgla Brunatnego Turów” [27]. The geological structure of the Turów lignite deposit is diverse and complicated, which causes difficulties in its exploitation.

The Lusatian lignite deposit is located in the Zytaw Trough, with the part of the deposit located in the southeastern, Polish part of the trough, documented under the name “Turow”. The Zytaw Trough is filled with Tertiary lignite-carbon series formations developed in the form of clays, sands and gravels with interbeds and deposits of lignite, forming several sedimentary cycles with a total thickness of up to 350 metres. These sediments are characterised by horizontal lithological variability: in the marginal zones, coarse-grained sediments dominate, and coal seams become thinner and have high ash content, while in the centre of the basin, the proportion of clay formations increases, and coal thickness increases and ash content decreases. Tertiary formations are covered by a thin layer of Quaternary sediments.

The crystalline bedrock complex consists of rocks of magmatic and metamorphic origin: granites, granite gneisses, granodiorites, accompanying vein rocks (aplites, metabasites) and subordinately gneisses and crystalline shales. They are commonly strongly kaolinised, forming extensive and thick (up to several tens of metres) weathered covers.

The subcoal complex is a set of lithologically differentiated clayey-sandy sediments, filling the depressions of the bedrock, largely levelling its strongly outlined morphology. The floor parts of this complex are characterised by the presence of inserts of redeposited granitoid and basaltoid weathering, while the ceiling parts are characterised by the presence of thin inserts of brown coals.

The I coal seam forms a uniform, with thin clay overgrowths in the bottom part, a bed of earthy coal characterised by high coalification. The overlying is highly variable related to the morphology of the subsoil and intense tectonics. Thickness ranges up to 35 m in the central part of the southern region. The average thickness is 15 m.

Lower and upper intercoal complex – similar to the subcoal complex – is a complex of clay layers, more or less sanded, sands and gravels with varying degrees of claying and thin inserts of xylitic coal. Formations of the complex lie above the first coal seam, overlying subcoal clays, or directly on weathered bedrock. The thickness of the complex varies from 3 m in the central part of the deposit to 135 m in its western part.

The II coal seam – the most widespread, with the greatest thickness and resources, is developed in the form of earthy, compact coals with thin inserts of xylitic coals, in the bottom parts abundantly interlayered with clay. The overburden of the seam is less varied. Thickness reaches up to 42 m in the central part of the southern region and thins out towards the edges of the basin. The average thickness is 20 m.

In the southern part of the Zitava basin, the upper (II) seam splits into two seams (II and III).

Coal seam III – has already been exploited to a large extent. In the northern part, it is represented by the mainly xylitic coals lying directly on the II seam. More to the south, these coals are separated from the II seam by formations of the upper intercoal clay complex and form a complex of irregular wedging layers.

Ceiling layers known as the **overcoal complex** are spread over the entire area of the Zittau basin, and the line of their extent coincides with the outline of the basin. The thickness of these layers is highly variable and varies from 0 to more than 200 m [28]. The greatest thickness was found in the zone of the central depressions of the basin, and decreases as the basin approaches its edges. In general, the described layers fill and obliterate the Zittau sinkhole to a large extent. Directly beneath the roof layers lies the upper coal seam, and in places where it wedges out, the roof layers lie on intercoal layers, from which lithologically they do not differ fundamentally. Above the roof layers lie Quaternary formations.

The Quaternary complex – is mainly Pleistocene glacial till sands and gravels, silty clays and till, sands and gravels of river terraces, and subordinate Holocene sands and silts, which together form a tight cover of several metres of Tertiary sediments.

The thickness and tightness of the overburden resting on the coal bed and the presence of faults play a crucial role in terms of methane emissions from the lignite bed in the case under analysis [29].

The total thickness of the overcoal complex and the Quaternary complex, i.e. the thickness of the overburden, determines its tightness and the amount of primary pressure in the rock mass. The analysis of the data shows that the average thickness of lignite seams is about 35 m, and the maximum is about 77 m. This affects the amount of methane reserves in the deposit, while the thickness of the overburden can range from a dozen metres to more than 200 metres. In the Turów mine, the average ratio of overburden thickness to coal deposit thickness is about 2.2:1 [30]. Taking the above into account, it can be concluded that the thickness of the overburden ranges from 77 m to 169 m (about 123 m on average).

In terms of tectonic disturbance, the structure of the sedimentary series of the deposit and coal seams is the result of multiphase tectonic movements preceding sedimentation, simultaneous with it, as well as post-sedimentation movements.

For exploitation, the most significant are the faults. Two genetic types of faults were found in the deposit:

Tensile gravity faults and accompanying compression faults with opposite slopes,

- Compaction sedimentary faults involving clipped lower links of sedimentary series on steepened slopes of highly elevated bedrock uplifts.

Gravity faults (the most common in the deposit) are associated with bedrock tectonics.

The geological documentation of the deposit (1991) identified a total of about 60 faults, mainly in the northern field area. The most significant tectonic dislocations of the deposit are:

- The Main Fault, dividing the deposit into two sedimentationally different parts, with a WE course and a drop of 15 to 90 m toward the N,
- South Fault, located in the southern part of the deposit, with a W-E course in the west and an NW-SE course in the east, with a drop of 30 to 140 m toward the N and NE.

Both faults have their foundation in the subsurface and cover all Tertiary sedimentary complexes.

The main dislocations are accompanied by subordinate pinnately developed faults with small discharges and extent. The surface parts of the deposit are additionally characterised by glacitectonic disturbances reaching a depth of 15-30 m.

Determination of the original stress state is the most crucial element of rock mass characterization. The determination of the state of stress in a rock mass unaffected by mining activities is based on the analysis of the medium treated as an elastic, continuous, homogeneous and isotropic half-space. This idealised, simplified model of the structure of the rock mass depicts a good approximation of the top layer of the earth's crust, made up of compact and solid rocks [31]. The weight of the overburden acting in the vertical direction induces a vertical stress of:

$$\sigma_z = \gamma H \quad (1)$$

Where σ_z – vertical strain (MPa), γ – the volumetric weight of overburden rocks (MN/m^3), H – the depth of the rock layer under consideration (m).

Assuming the volumetric weight of the overburden in the analysed case is equal to $\gamma = 0.02 \text{ MN/m}^3$ (mainly clay) at an average depth of $H = 123 \text{ m}$, the value of vertical stress will be $\sigma_z \approx 2.5 \text{ MPa}$.

The research areas analysed in the article and the laboratory tests performed do not directly relate to the conditions of the Turów lignite mine. The geological-mining conditions presented above are provided only for illustrative purposes to better understand their significance in the overall analysis of methane emissions.

2.3. Methods of determining the methane emission factor in open-pit coal mines

The amount of methane (CH_4) emitted into the atmosphere during coal exploitation depends on several factors. Some of the most important are the degree of coalification, the depth of the coal seams and the method of extraction. Coal mining in underground mines releases a higher amount of methane due to the greater saturation of deeper seams.

In general, the release of methane is influenced by the pressure distribution in the rock mass. In the primary rock mass, everything is in equilibrium (in the case under review, about 2.5 MPa). During mining and when removing the overburden, the pressure decreases to about 0.1 MPa and the original equilibrium of the rock mass is violated. At lower pressure, the sorption capacity of the coal decreases and part of the methane becomes free methane contributing to its emission.

Methane emissions in underground mining come from both ventilation and degassing (drainage) systems. These emissions usually originate from specific locations and can therefore be considered point sources. In the case of open-pit mines, methane emissions are dispersed in different parts of the mine and can be considered area sources. These emissions can result from crushing and the extraction of coal along with the overburden [32].

Methane emission measurements for open-pit mining are becoming increasingly available. However, they are difficult to perform and there are currently no routine, commonly used methods. Access to in situ gas data in the reservoir is also limited. As recommended by the International Panel on Climate Change (IPCC) [32], for open-pit mines, methane emissions are determined by applying the global emission factors or individual emission factors determined for a given basin or deposit. Two variants were identified according to which methane emissions can be determined for open-pit mines: Tier 1 and Tier 2. The choice of variant is determined by a decision tree, shown as a figure (Fig. 1).

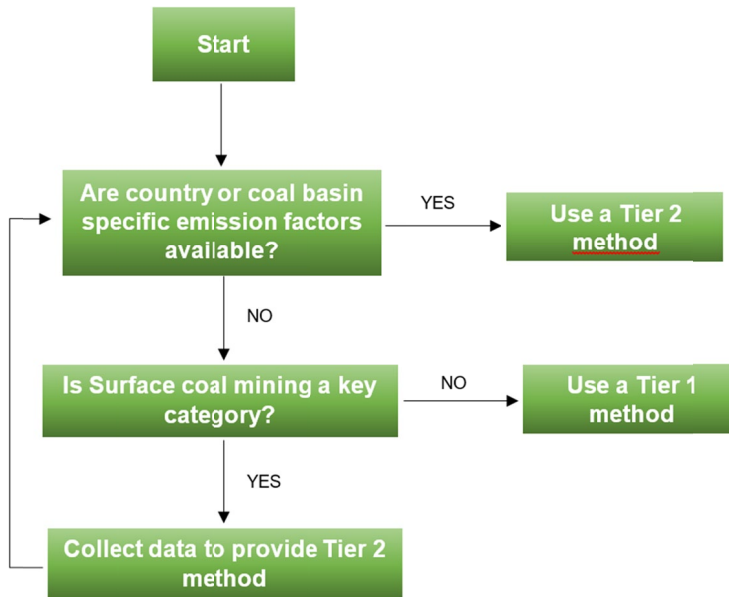


Fig. 1. Decision tree for open-pit lignite mining for determination of methane emission

Tier 1: Global Average Method – Surface Mines – Methane

$$\text{CH}_4 \text{ Emissions} = \text{CH}_4 \text{ Emission Factor} \times \text{Surface Coal Production} \times \text{Conversion Factor} \quad (2)$$

Where Methane (CH₄) Emissions (Gg/year); CH₄ Emission Factor (m³/tonne); Surface Coal Production (tonne/year); Conversion Factor (Gg/m³).

Emission Factors:

- Low: CH₄ Emission Factor = 0.3 m³/tonne
- Average: CH₄ Emission Factor = 1.2 m³/tonne
- High: CH₄ Emission Factor = 2.0 m³/tonne

Conversion Factor: $0.67 \times 10^{-6} \text{ Gg/m}^3$ – The value of CH₄ density converts the volume of CH₄ to the mass of CH₄. The density is taken at 20°C and 1 atmosphere pressure.

For the Tier 1 variant, it is good practice to use a low emission factor for mines with an average overburden thickness above the coal seam of 25 m, a medium emission factor for overburden thickness between 25 and 50 m, and a high emission factor for mines with overburden thickness of more than 50 m. In the absence of overburden thickness data, an average emission factor of 1.2 m³/tonne should be used.

Tier 2: Method based on data disaggregated to country specific or coal basin level

The method for variant 2 (Tier 2) uses the same equation (2) as for variant 1 (Tier 1) but with data specific to a particular area or the entire deposit. For countries using this option, it is necessary to determine individual emission factors by analysing the gas content of the coal layers for a given open-pit mine. Sampling and analysis of the gas content should be carried out

according to a specific procedure. Thus, this option allows the determination of methane emission factors required to determine total annual emissions based on proprietary methods developed by scientific institutions. Based on the above provisions, some countries have attempted to calculate methane emissions based on individual data for a given deposit (Colombia, Australia, USA, Mexico, China) [4,33-35]. In Colombia, attempts were made to determine methane emissions through the IPCC guidelines according to Tier 1 and their method, based on measurements of actual CBM (coal bed methane) content in the analysed deposit, resulting in Tier 2 [4]. The method was based on the determination of naturally occurring methane in coal based on core samples of lignite, which are placed in special hermetically sealed containers. The measurement process involved three different steps, given that the total gas content associated with coal was the sum of three gas components.

Another method described in the literature is based on the correlation between the volume of gas contained in the coal and the internal pressure of the seam [9]. Based on this theory, the gas pressure increases with the depth of the seam, as does the volume of methane contained in the coal. The equation that can predict the amount of gas that coal can contain at a certain pressure or depth is the adsorption isotherm. The Langmuir sorption isotherm is considered the best model for determining the sorption capacity of gas relative to coal [36-38]. The Langmuir sorption isotherm can be used to determine the gas content of coal, and can be described by the following equation:

$$EF = V_L(d \times L)/(P_L + d \times L) \quad (3)$$

Where V_L – The Langmuir volume coal sample; P_L – The Langmuir pressure of that sample; L – The Langmuir constant; d – mining depth (metres)

2.4. Determination of coalbed methane content

2.4.1. Single phase vacuum degassing (SPVD)

The basic direct method for determining methane content, which is currently used in the conditions of the Polish mining industry, is the single-phase vacuum degassing method. It is based on the requirements of the PN-G-44200:2013-10 standard [39] and the Regulation of the Minister of Energy of November 23, 2016, on detailed requirements for the operation of underground mining plants [40]. The study of direct methods dates back to the 1970s [41]. The basis of this method is borehole samples taken from a drilling depth of 3.5-4.0 m into specially sealed airtight containers with steel balls (Fig. 2). To adapt the method to the conditions of an open-pit lignite mine, core and lump samples were used for the tests described in the article.

After being transported to the laboratory, in the first step, the coal sample is ground using steel balls inside the container on a special shaker. Subsequently, the gas released from the coal structure is pulled off under vacuum and analysed on a gas chromatograph to determine the percentage of gas components. The coal sample remaining in the container is subjected to physicochemical analysis for hygroscopic moisture content, transient moisture content, ash content, and volatile matter content. These parameters are necessary to calculate the final result of methane content in terms of a dry, ash-free basis (daf). The final result of the determination is subjected to compensation for gas losses, which are generated during sampling for testing (before placing the coal in an airtight container). The Central Mining Institute – National Research Institute, based



Fig. 2. Hermetically sealed containers for coal sampling

on many years of empirical research, established a loss factor of 1.33, which was also used for this study [42,43]:

$$M_o = 1.33 \times M_L \quad (4)$$

Where M_o – methane content with gas loss coefficient ($\text{m}^3\text{CH}_4/\text{t}_{\text{daf}}$); M_L – methane content obtained in the laboratory (without gas losses) ($\text{m}^3\text{CH}_4/\text{t}_{\text{daf}}$).

2.4.2. United States Bureau of Mines (USBM)

The second method used to determine methane content was also a direct method, based on United States Bureau of Mines (USBM) standards, which has been analysed by many researchers in the literature, proposing various possibilities for its application as well as modifications [43-49]. This method is based on the Bertard method developed in France in the 1970s [44]. USBM desorption tests are performed based on the American Society for Testing and Materials (ASTM) standard ASTM D7569/D7569M – 10 [50] and the United States Department of The Interior's documentation "The Direct Method of Determining Methane Content of Coalbeds for Ventilation Design" RI 7767 [45]. The USBM method is used to identify and document methane deposits occurring in coal seams. The method is based on the analysis of the free desorption of gas from the coal core and the measurement of its volume. The use of the USBM method makes it possible to determine individual gas components (Eq. (5)) such as:

- Desorbing gas Q_D – the component of total gas that is freely emitted in a hermetically sealed container, determined by volume measurement over time.
- Lost gas Q_L – the component of total gas that is released in an uncontrolled manner before the sample is sealed in an airtight container.
- Residual gas Q_R – the component of total gas that remains in the coal sample after the desorption tests and is sorbed to the coal structure.

$$Q_T = Q_L + Q_D + Q_R \quad (5)$$

Where Q_T – total gas; Q_L – lost gas; Q_D – desorbing gas; Q_R – residual gas.

The use of the USBM method makes it possible to determine individual gas losses for each of the analysed coal samples as well as to identify potentially desorbable methane resources, which is particularly important when determining its emissions.

The USBM tests are based on coal core samples taken into special airtight containers (Fig. 3A). The methane content tests using this method at the Central Mining Institute – National Research Institute are performed using a proprietary test stand (Fig. 3B).

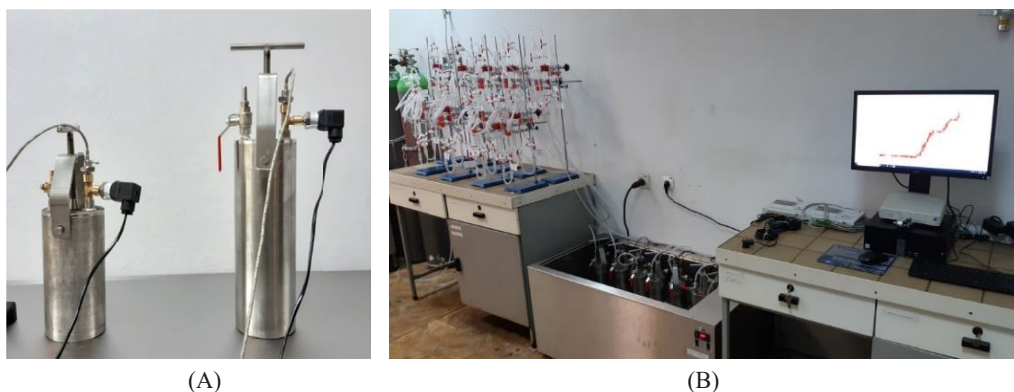


Fig. 3. Central Mining Institute – National Research Institute test stand for methane content determination by the USBM direct method: (A) hermetically sealed containers with pressure and temperature sensors; (B) the original measurement system

During the measurement of the volume of desorbing gas from the coal core, its composition is periodically tested by gas chromatography. The reading of the volume of desorbing gas is recorded until its complete disappearance from the sample. To determine the residual gas, a piece of the core after free desorption is placed in an airtight container with steel balls and subjected to grinding. The volume of gas released is then measured and its composition analysed by gas chromatography. The lost gas is determined graphically based on the desorption graph, which is derived from the obtained data. The final methane content value is determined as the sum of the individual gas components.

2.5. Determination of sorption properties

The sorption tests for this article were conducted using the gravimetric method. A gravimetric sorption system IGA-001 (Intelligent Gravimetric Analyzer) was applied (Fig. 4), which is used for precise analysis of the size, dynamics and kinetics of gas sorption in porous materials. The performed tests allowed us to determine the kinetics of methane sorption on the analysed coal samples and determine the parameters describing the rate of sorption processes in its structure.

Sorption tests were carried out using samples analogous to those taken for the methane content analysis. Representative analytical samples of grain class 0.2-0.25 mm were prepared. Coal samples of approximately 150 mg were used for the study. The analysis was carried out under stable and continuously controlled temperature conditions of 25°C, and the sorption kinetics were analysed at a methane saturation pressure of 0.1 MPa. Based on the obtained methane sorption



Fig. 4. IGA-001 gravimetric sorption system for methane sorption kinetics investigation

curves, the effective diffusion coefficient (D_e) was determined, which characterises the course of sorption kinetics (its dynamics), as well as the half sorption time ($t_{1/2}$), which determines the time when the amount of sorbed gas is half of the amount of total sample saturation. Coal with a higher value of the effective diffusion coefficient can release the same amount of gas in a shorter time, assuming the same grain size of the pulverised coal mass.

3. Results and discussion

Based on laboratory tests conducted for samples taken at two locations of one of Poland's lignite deposits, it was possible to determine correlations and provide guidelines for determining the methane emission factor. The first part of the work involved testing the natural methane content of coal (methane content). The research was carried out using two direct methods: single-phase vacuum degassing (SPVD) and the United States Bureau of Mines (USBM). A comparison of the results obtained for both methods is summarised in the form of TABLE 2.

Based on the obtained results, it can be concluded that the results for study area I were lower than those for area II. The methane content for area I obtained by the single-phase vacuum degassing method ranged from $0.003 \text{ m}^3\text{CH}_4/\text{Mg}_{\text{daf}}$ for sample No. 2X to $0.028 \text{ m}^3\text{CH}_4/\text{Mg}_{\text{daf}}$ for sample No. 3X. For the USBM method, the results ranged from $0.004 \text{ m}^3\text{CH}_4/\text{Mg}_{\text{daf}}$ for sample No. 2Y to $0.052 \text{ m}^3\text{CH}_4/\text{Mg}_{\text{daf}}$ for sample No. 3Y, which indicates higher values than for the SPVD method. For area II, the determined natural methane content of coal was higher. For the single-phase vacuum degassing method, values range from $0.028 \text{ m}^3\text{CH}_4/\text{Mg}_{\text{daf}}$ for sample No. 7X to $0.176 \text{ m}^3\text{CH}_4/\text{Mg}_{\text{daf}}$ for sample No. 8X. However, for the USBM method, values range from $0.025 \text{ m}^3\text{CH}_4/\text{Mg}_{\text{daf}}$ for sample No. 7Y to $0.306 \text{ m}^3\text{CH}_4/\text{Mg}_{\text{daf}}$ for sample No. 8Y.

Based on the results of the methane content testing, it can be concluded that their values were relatively low. Only in one case for the USBM method for sample No. 8Y did the value exceed $0.3 \text{ m}^3\text{CH}_4/\text{Mg}_{\text{daf}}$. The above results show that the use of universal global methane emission

TABLE 2

Results of the investigation of methane content in the analysed lignite samples

No.	Research area	Methane content by SPVD method M_o [m ³ CH ₄ /Mg _{daf}]	No.	Methane content by USBM method		
				Sum of lost and desorbing gas [m ³ CH ₄ /Mg _{daf}]	Residual gas [m ³ CH ₄ /Mg _{daf}]	Total gas M_o [m ³ CH ₄ /Mg _{daf}]
1	X	I	Y	0.010	0.003	0.013
2	X		Y	0.003	0.001	0.004
3	X		Y	0.037	0.015	0.052
4	X		Y	0.032	0.008	0.040
5	X		Y	0.007	0.002	0.009
6	X	II	Y	0.036	0.011	0.047
7	X		Y	0.019	0.006	0.025
8	X		Y	0.216	0.090	0.306
9	X		Y	0.141	0.072	0.213
10	X		Y	0.053	0.013	0.066

factors for lignite deposits, which can vary significantly in methane content depending on the origin, mining and geological conditions and depth, can affect overestimation or underestimation in the determination of emissions.

The rate of sorption processes and the dynamics of the release of methane from the coal structure have a significant impact on the subject of its emission. TABLE 3 shows the results of methane sorption kinetics studies on the analysed lignite samples.

TABLE 3

Results of the investigation of methane sorption kinetics on lignite samples

No.	Research area	Parameters of sorption kinetics	
		D_e [cm ² /s]	$t_{1/2}$ [s]
1	I	1.70×10^{-8}	228 (~4 min)
2		1.18×10^{-8}	329 (~5 min)
3		1.52×10^{-8}	255 (~4 min)
4		1.31×10^{-8}	298 (~5 min)
5		1.18×10^{-8}	329 (~5 min)
6	II	1.61×10^{-8}	241 (~4 min)
7		1.17×10^{-8}	333 (~4 min)
8		1.03×10^{-8}	377 (~6 min)
9		1.47×10^{-8}	264 (~4 min)
10		1.25×10^{-8}	311 (~5 min)

Based on the results obtained from the study of sorption kinetics, relevant parameters such as the effective diffusion coefficient (D_e) and the half sorption time ($t_{1/2}$) were determined, which describe the dynamics of the occurrence of sorption processes in the coal structure. The obtained values of the effective diffusion coefficient range from 1.03×10^{-8} to 1.70×10^{-8} cm²/s. To illustrate the scale of the magnitude of this parameter, for hard coal, values exceeding 0.15×10^{-8} cm²/s (the

accepted limit) have been determined that structural changes are possible, indicative of possible tectonic disturbances, such as faults [51,52]. The values obtained for the lignite samples analysed are an order of magnitude higher than this limit. It is not possible to compare their scale with hard coal. While it is not feasible to directly compare the scale of lignite with that of hard coal, one significant conclusion can be drawn: the structure of lignite differs from that of hard coal, resulting in a distinct ratio of pore size distribution. As stated by other researchers, the pore structure of lignite coals is highly developed and extensive, and is likely to be dominated by mesopores with pore sizes of 2-50 nm and macropores with pore sizes >50 nm, while the proportion of micropores in the structure is only about 3% [24,53]. Confirmation of the above thesis is also provided by the value of the half sorption time, which defines the time at which the amount of sorbed gas is half the amount of total saturation of the sample. The obtained values of $t_{1/2}$ for the analysed lignites ranged from 228 to 377 s, which is about 4 to 5 minutes. This is a very short time compared to hard coals, which typically require 2 to 6 hours to half-saturate the sample with methane (assuming the same sample grain size) [51]. Based on the above observations, it can be concluded that lignite is a porous material that is quickly able to release the methane sorbed in its structure. Therefore, when determining its emissions, it should be taken into account that after just a few minutes, lignite can fully release most of the gas. This is particularly important for determining methane emissions in open-pit mines during post-mining activities, i.e. transportation and storage. It can be suspected that the vast majority of the desorbing gas, i.e. the gas that can be released freely from the coal structure, will be emitted in the first minutes of the mining process. Thus, emissions in the post-mining process for lignite in the case of an open-pit mine, may prove to be marginal, or negligible.

The high methane sorption kinetics found as a result of the study also provide very valuable information regarding the potential gas losses generated at the sampling stage for testing. During core sampling, it is necessary to determine the losses generated from the start of drilling until the sample is placed in an airtight container. For the single-phase vacuum degassing method, a compensation factor of 1.33 was used, which is suitable for hard coal. As observed, due to the high methane sorption kinetics for lignite, this coefficient should be much higher, as the losses in the first minutes will be higher. This may explain the lower methane content values obtained by the SPVD method compared to the USBM method. In the USBM method, gas losses are determined individually for each sample analysed. This approach allows for more reliable test results. Therefore, the results of methane content determination obtained by the United States Bureau of Mines method will be considered for further analysis. An additional advantage of the USBM method is the determination of individual gas components such as desorbing gas, lost gas, and residual gas. This is particularly important for the analysis of methane emissions from an open-pit mine, since only the desorbing and lost gas components can freely emit into the atmosphere. Therefore, it is possible to determine the total potential amount of methane that can be emitted to the atmosphere (including crushing and combustion), or the potential amount of methane that can be emitted through free desorption during the mining process. A similar approach is presented in the work of Mariño-Martínez [4], where to determine the proper methane emissions from a specific open-pit mine in Colombia, 70% of the total methane content was considered, while 30% was the residual gas sorbed in the coal structure, which is unable to emit freely in the mining process.

Based on the conducted analysis, it was concluded that the methane emission factor presented in Eq. (2), as a key factor for determining methane emissions to the atmosphere, should be described by the value of methane content determined by the USBM method expressed in m^3

of methane per tonne of mined coal. When determining the emissions during the mining process, it is necessary to consider only the value of the desorbed and lost gas component ($Q_D + Q_L$). This does not include the residual gas component (Q_R), which is sorption-bound to the coal structure and cannot be freely emitted at this stage. To determine the methane emission factor, it is necessary to properly plan the collection of representative samples and average the methane content value, which will constitute the emission factor for a given area or deposit. In the case of this article, 5 samples were taken for each of the two areas analysed for methane content. For a hypothetical lignite deposit in an open-pit mine with an annual production of 15 million tonnes, a simulation of methane emissions was carried out. An equal distribution of extraction to study areas I and II (7.5 million tonnes/year each) was assumed and emissions were determined using accepted IPCC recommendations [32]. An analysis was carried out for variant I (Tier I) based on established global emission factors and variant II (Tier II) based on obtained actual data and factors for a specific deposit or a specific open-pit mine. The analysis was carried out individually for study areas I and II. The results of the simulation are presented in the form of TABLES 4 and 5.

For variant I (Tier I), potential emissions were simulated for the entire analysed hypothetical deposit with an annual production of 15 million tonnes using the IPCC recommended emission factors of 0.3, 1.2 and 2.0 m³/tonne. Simulations were performed for both study areas, i.e. area I and area II. Due to the same coefficients and the amount of coal produced, the obtained results were also the same. For study areas I and II, using a coefficient of 0.3 m³/tonne, the total annual emission value was 1508 tonnes of CH₄, using a coefficient of 1.2 m³/tonne, the total annual emission was 6030 tonnes of CH₄, and using a coefficient of 2.0 m³/tonne, the total annual emission was 10050 tonnes of CH₄.

TABLE 4

Simulation results of methane emission for the analysed test areas I and II for the hypothetical lignite deposit based on IPCC regulations with global factors (Tier I)

Tier I IPCC regulations with global factors				
Research area	Annual coal production [tonne]	Conversion factor [tonne/m ³]	CH ₄ emission factor [m ³ /tonne]	Total forecast emission [tonne CH ₄]
I	7500000	0.00067	0.3	1508
			1.2	6030
			2.0	10050
II	7500000		0.3	1508
			1.2	6030
			2.0	10050

The simulation results obtained using the actual emission coefficients determined from the United States Bureau of Mines methane content studies are summarised in TABLE 5. The coefficients were determined separately for study areas I and II. Simulation results for two variants are presented:

- Projected methane emissions based on the factor calculated for emissions in the mining process, for which the potential release of desorbing and lost methane ($Q_D + Q_L$) is possible, and the residual gas (Q_R) remains sorption-bound in the internal structure of coal.

- The total projected methane emissions that can be released from the entire coal structure, from the mining process, through crushing and grinding, and finally to combustion in a power plant.

TABLE 5

Simulation results of methane emission for the analysed test areas I and II for the hypothetical lignite deposit based on individual coal mine data (Tier II)

Tier II Based on individual coal mine data						
Research area	Annual coal production [tonne]	Conversion factor [tonne/m ³]	CH ₄ emission factor ($Q_D + Q_L$) [m ³ /tonne]	Forecast emission based on $Q_D + Q_L$ [tonne CH ₄]	CH ₄ emission factor (Total gas) [m ³ /tonne]	Total forecast emission [tonne CH ₄]
I	7500000	0.00067	0.018	90	0.024	121
II	7500000		0.093	467	0.131	658

Based on the obtained results, it can be concluded that for the emission factor determined on the basis of the amount of desorbing and lost gas, which for study area I was 0.018 m³/tonne, an emission volume of 90 tonnes of CH₄ per year was obtained, while for study area II the factor was 0.093 m³/tonne, and the determined annual CH₄ emission volume was 467 tonnes. In the case of the determined factor for the total content of methane that can be released in all processes related to its extraction and processing, for the study area I it was 0.024 m³/tonne and the calculated annual emission of methane was 121 tonnes. For the study area II, the determined factor was 0.131 m³/tonne and the annual emission was 658 tonnes of CH₄.

TABLE 6

Simulation of methane emissions in the mining process for the entire analysed hypothetical deposit based on IPCC recommendations and determined emission factors, per 1000 tonnes of lignite produced

	Tier I IPCC regulations with global factors	Tier II Based on individual coal mine data
Forecast CH ₄ emission for analysed coal deposit [tonne CH ₄]	3015 (for emission factor 0.3)	563 (for averaged factor 0.056 for the whole deposit)
	12060 (for emission factor 1.2)	
	20100 (for emission factor 2.0)	
Forecast CH ₄ emission for 1000 tonnes of coal for analysed coal deposit [CH ₄ tonne/1000 tonne of coal]	0.201 (for emission factor 0.3)	0.038 (for averaged factor 0.056 for the whole deposit)
	0.804 (for emission factor 1.2)	
	1.340 (for emission factor 2.0)	

Based on the Proposal for a Regulation of the European Parliament and of the Council on the Reduction of Methane Emissions in the Energy Sector and Amending Regulation (EU)

2019/942, among others, operators of open-pit mines will be obliged to submit annual methane emissions, and as stipulated in Chapter 4, Section I, Article 20 – also to establish emission factors in accordance with applicable scientific standards [54]. The above proposal stated that the allowable methane emissions would be determined in tonnes of methane per 1000 tonnes of coal mined. A compromise of 5 tonnes of CH₄ per 1000 tonnes of coal mined is assumed for the above allowable emission level. Based on the presented guidelines, the obtained test results were recalculated for the entire analysed hypothetical deposit, which consists of test areas I and II. For the two variants analysed, Tier I and Tier II, the emission values obtained in the mining process as a result of the simulation were converted per 1000 tonnes of lignite for the entire deposit. The results are shown in TABLE 6.

Based on the obtained results, it can be concluded that for the analysed hypothetical deposit, the volume of emissions does not exceed the permissible level of 5 tonnes of CH₄ per 1000 tonnes of mined coal. This confirms the thesis that lignite coals are less saturated with methane than hard coals. For variant I (Tier I), determined using global factors according to IPCC recommendations, the following values were obtained successively: for a factor of 0.3-0.201 tonnes of CH₄ per 1000 tonnes of mined coal, for a factor of 1.2-0.804 tonnes of CH₄ per 1000 tonnes of mined coal, and for a factor of 2.0-1.340 tonnes of CH₄ per 1000 tonnes of mined coal. In the case of variant II (Tier II), which is based on actual data for the deposit, the result is presented in the form of an average for the entire deposit, as a component of study areas I and II analysed. As a result of averaging the emission factor, its value of 0.056 m³/tonne was obtained. Based on this, the methane emission value for the entire deposit was determined at 0.038 tonnes of CH₄ per 1000 tonnes of mined coal.

As a result of studying the methane content of the analysed hypothetical lignite deposit and the determined emission factors, it can be concluded that the emission values for the determined factor based on real data were lower than for the global factors recommended by the IPCC under variant I (Tier I). However, the results obtained cannot be used as a rule. Each deposit may be different. In this case, a low methane saturated lignite deposit was analysed. Based on the results, it can be concluded that the use of common global methane emission factors to determine annual methane emissions to the atmosphere may lead to some underestimation or overestimation.

The results of the research described in the article are aimed at presenting recommendations and a course of treatment leading to the determination of the methane emission factor for open-pit lignite mines. The values obtained refer to a hypothetical lignite deposit, and therefore should not be taken into account when considering the level of methane emissions for lignite mines in Poland.

4. Conclusions

Within the framework of this article, an analysis related to the determination of methane emissions from open-pit lignite mines was carried out. A simulation related to the application of global, universal emission factors recommended by the IPCC [32], and the factor determined individually for the analysed deposit based on the conducted research was carried out. A methane content study was conducted using two direct methods: the single phase vacuum degassing (SPVD) method and the United States Bureau of Mines (USBM) method. In addition, a methane sorption kinetics study was conducted to describe the dynamics of the occurrence of this process in the coal structure.

The results show that the methane content of coal deposits in some countries could be lower than in major lignite producing countries. For this reason, emission calculations should not be based on international tables and global emission factors, but on direct measurements of the gas content. The results of the conducted research and analysis indicate the validity of determining the content of methane of natural origin in coal for open-pit lignite mines using the United States Bureau of Mines method for coal core samples. This method allows accurate and individual determination of gas losses at the sampling stage. The method also allows the determination of individual gas components such as desorbing gas, lost gas and residual gas.

Based on the simulation carried out using global emission factors for the entire analysed hypothetical deposit, applying variant I (Tier I) according to IPCC recommendations, for a factor of 0.3 m³/tonne a value of 3015 tonnes of CH₄ annual emissions was obtained, for a factor of 1.2 m³/tonne annual emissions were 12060 tonnes of CH₄, and for a factor of 2.0 m³/tonne annual emissions were 20100 tonnes of CH₄. Using variant II (Tier II) of the IPCC recommendations, an individually determined emission factor of 0.056 m³/tonne was applied. This resulted in a total annual emission of 563 tonnes of CH₄. Based on this, it can be concluded that the use of globally established factors, when each coal/lignite deposit in different countries is different due to the diversity of geological and mining conditions, may lead to an overestimation or underestimation of the relevant methane emissions.

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