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Alternative methodology to determine effective coefficient of methane diffusion in coal

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Abstract

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Keywords

Mining; Coal; Sorption; Diffusion; Methane; Diffusion coefficient

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Alternative methodology to determine effective coefficient of methane diffusion in coal

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Abstract

The article presents the results of research in to the development of an alternative method of determining the effective coefficient of methane diffusion in coal, based on a mathematical model following Fick's second law. The research was conducted based on the recorded courses of methane sorption kinetics in coal samples obtained in laboratory conditions with the precise gravimetric sorption system IGA-001. The value of the coefficient was treated as an element tuning the model in such a way that the average relative error of the *ex post* was as small as possible. Model verification indicated the correctness of its assumptions. The development of a more accurate methodology to determine the effective coefficient of methane diffusion in coal will enable the verification of the models applied in order to describe the physical and chemical mechanisms of the methane desorption natural phenomenon. It will also pave the way for further research aimed at the determination of: the gas bearing capacity of coal seams, the degree of degassing of seams which results from the mining operations being conducted; or to what extent it is possible to degas the deposits.

Keywords: mining, coal, sorption, diffusion, methane, diffusion coefficient

1. Introduction

Mining works conducted in a highly gassy deposit are accompanied by methane emissions into the mine workings, which may result in an explosion hazard. Hence, research into methane desorption from coal seams is still an important issue, both for the safety of the mine works and for environmental protection. Therefore, laboratory tests of methane sorption play an important role. They help the mechanism of the phenomenon of methane emission in nature to be recognized, the set of factors and parameters which influence its course to be determined and they provide input data for the models which determine methane emission into mine workings during mining operations. The research, which has been conducted at the Central Mining Institute in Katowice (Poland) since 2006, shows that when mining works are conducted in a gassy deposit, sorption properties of coal (methane

sorption capacity and effective coefficient of methane diffusion in coal) significantly influence the level and character of methane emission [1–11]. Sorption properties of coal influence the level of gas and rocks outburst hazard [12]. Various types of coal demonstrate different sorption properties in given deposits. To characterise hard coals and their ability to accumulate and desorb methane, the tests conducted by Central Mining Institute, Department of Gas Hazard Control in the Barbara experimental mine determine sorption capacity of coal in standard conditions and the value of the effective coefficient of diffusion. The value of the effective coefficient of diffusion characterises the kinetics of methane sorption. The methodology of determining the coefficient was developed by Timofeev in the early 1960s [13]. The order of magnitude, which the coefficient reaches, often results in significant inaccuracy exceeding the acceptable error. Hence, the research makes an

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attempt to develop a new methodology with IT techniques.

2. Currently applied methodology of determining the sorption properties of coal

Tests of the sorption properties of hard coals which primarily focus on the gas and rock outburst hazard aspect are conducted by the Central Mining Institute in the laboratory of the Department of Gas Hazard Control in the Barbara experimental mine. Hiden Isochema's Intelligent Gravimetric Analyser IGA-001, a high accuracy gravimetric and sorption system for the precise analyses of gas sorption volume, dynamics and kinetics, was applied to determine the amount of gas accumulated through sorption, i.e. mainly the capacity of coal seams. It is advanced equipment which measures changes in the weight of a sample over time, caused by the sorption processes occurring in coal and other porous materials (Fig. 1). Sorption analysers, whose working principle is based on the gravimetric method, have been used for sorption and diffusion tests for just over a dozen or so years [14]. The advantages of this method of testing hard coals are presented by, among others, [15,16] and [17].

To characterise methane accumulation and the desorption of hard coals, the tests determined the sorption capacity of coal in standard conditions and the value of the effective coefficient of diffusion [10,18,19]. Standard sorption capacity q_s was determined for a dry sample in laboratory conditions in cm^3/g of methane sorbed by coal at the point of equilibrium which was reached under standard atmospheric pressure (1 bar) and temperature (25°C , i.e. 298 K). The test results are presented the in form of methane sorption kinetics graphs. This paper will analyse a methane sorption kinetics graph obtained in the tests of a coal sample collected in incline N-1, seam 404/4 of the Pniówek coal mine in the Upper Silesian Coal Basin (Fig. 2).

The data recorded during the tests was applied to determine the value of the methane sorption capacity of the coal in standard conditions, as the amount of the methane sorbed at the point of equilibrium. Sorption capacity is expressed in cm^3/g , thus, the value expressed in $\text{mg CH}_4/\text{g}$ ought to be divided by methane density in standard conditions. For the case above, methane sorption capacity was determined as

$$q_s = 4,579 \text{ cm}^3/\text{g}$$

Methane sorption kinetics is a very important factor in the assessment of the dynamics of methane emission from a coal seam, where (or in the vicinity



Fig.1. Gravimetric analyser IGA-001 to test gas sorption.

of which) mining works are conducted (roadways, longwalls) [2–6,10,20–24]. Methane sorption kinetics is characterised by the effective coefficient of methane diffusion in coal D_e , which is determined in laboratory conditions and expressed in cm^2/s . The value of the coefficient depends on the diffusion mechanism in the pores of the solid phase and properties of the system [25–27]. To determine the value [21], the time corresponding to the half-saturation of coal is read from the sorption kinetics graph and then substituted in Timofeev's equation, from the beginning of the 1960s [13,18]:

$$D_e = \frac{0,308 \cdot R^2}{\pi^2 t_{0,5}} \quad (1)$$

where:

D_e – effective coefficient of diffusion, cm^2/s ,

R – mean coal grain radius, cm,

$t_{0,5}$ – time corresponding half-saturation of coal, s.

Mean grain radius R for laboratory conditions is calculated with the equation:

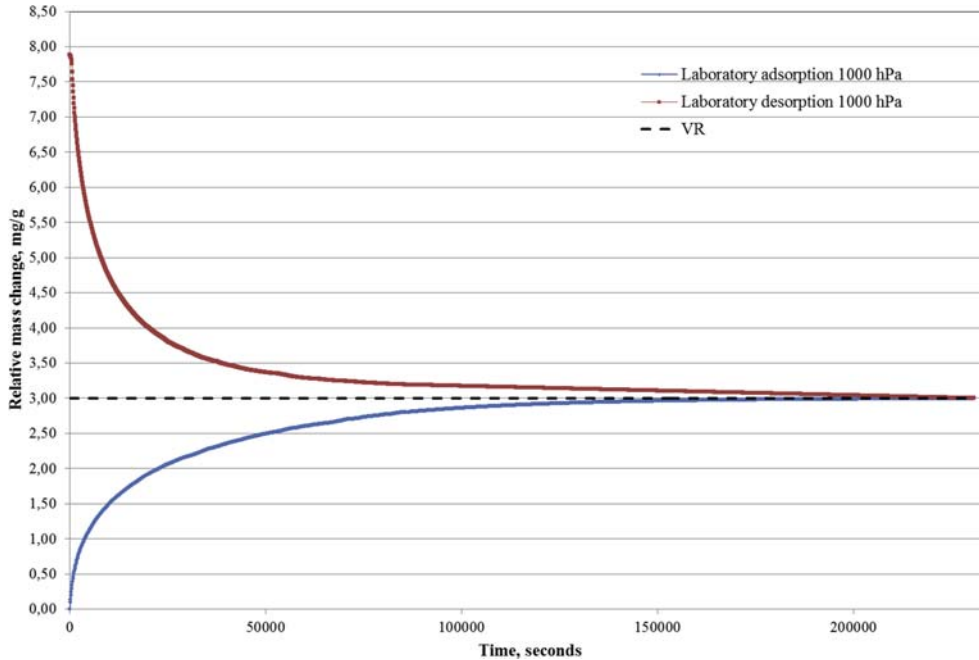


Fig.2. Methane sorption kinetics graph—coal sample of seam 404/4 in the Pniówek coal mine.

$$R = \frac{1}{2} \sqrt[3]{\frac{2d_1^2 d_2^2}{d_1 + d_2}}, \text{ cm} \quad (2)$$

where:

- R – mean radius of grain size, cm,
- d_1 – upper radius of grain size, cm,
- d_2 – lower radius of grain size, cm.

The equation is a transformation of W. Budryk's equation [21]. For the analysed case, the value of the effective coefficient of methane diffusion in coal, calculated basing on equations (1) and (2), are $R = 0.0179$ cm and $t_{0.5} = 10470$ s, is $D_e = 0.952 \cdot 10^{-9}$ cm²/s.

As it previously mentioned in the introduction, applying such a method of determining the effective coefficient of diffusion often results in significant inaccuracies, which exceed acceptable error (most often of up to 25%), and, as a consequence, renders it impossible to interpret properly its value to assessing methane hazard and methane and rock outburst hazard, and to carry out an accurate forecast of methane emissions into mine workings.

3. Mathematical model describing the course of methane sorption kinetics in coal samples—

alternative methodology to determine the effective coefficient of diffusion

The J.P. Seidel method, applied in the American mining industry, may be one of the methods used to illustrate the mechanism of methane desorption from coal [22,28,29]. Like other models (e.g. the uniporediffusion model – [30]), the methodology tries to reflect the complex mechanism of methane emission from spherical grain ($0 \leq r \leq R$), with effective coefficient of diffusion D_e , starting with the following equation:

$$\frac{\partial V}{\partial t} = D_e \left(\frac{2}{r} \frac{\partial V}{\partial r} + \frac{\partial^2 V}{\partial r^2} \right) \quad (3)$$

It may be observed that equation (3) follows Fick's second law for isotropic radial diffusion, determining changes in the concentration of diffusing component (in this case – methane) over time, in the specific cross-section of the diffusion flux. For the equation, the initial condition looks as follows:

$$V(r, 0) = V_0 (\text{initials gas content}) \quad (4)$$

And the boundary conditions ought to consider the variability of the amount of methane on the surface of a microporous grain over time:

$$V(R, t) = V_R(t); \left. \frac{\partial V(r, t)}{\partial r} \right|_{r=0} = 0 \quad (5)$$

The boundary value is often assumed to be constant $V_R = \text{const}$ (the target amount of gas which indicates the sorption capacity of coal in given conditions).

The solution of this issue, expressed with an infinite series after integrating the grain volume, looks as follows:

$$\bar{V}(t) = \bar{V}_\infty \left[1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} \exp\left(-n^2 \pi^2 \frac{D_e t}{R^2}\right) \right] \quad (6)$$

When $\bar{V}(t)/\bar{V}_\infty < 0.25$, [31] concluded that for the time when $\bar{V}(t)/\bar{V}_\infty < 0.5$ the equation above can be simplified to the following form:

$$\bar{V}(t) = 6\bar{V}_\infty \sqrt{\frac{D_e t}{\pi R^2}} \quad (7)$$

When $\bar{V}(t)/\bar{V}_\infty > 0.7$, equation 6 only has to consider the first sum element ($n = 1$), hence:

$$\bar{V}(t) = \bar{V}_\infty \left[1 - \frac{6}{\pi^2} \exp\left(-\pi^2 \frac{D_e t}{R^2}\right) \right] \quad (8)$$

where:

D_e – value of the effective coefficient of methane diffusion in coal, cm^2/s ,

t – time, s.

R – mean coal grain radius, cm.

Equilibrium methane content in grains \bar{V}_∞ corresponds to the infinitely long duration of this process. In the analysed case – in laboratory conditions, when the sorption curve is determined, equilibrium is reached after approximately one day. Clearly, in underground mine conditions, it would take much longer to reach equilibrium, perhaps even a few years depending on the grain radius R .

In the laboratory case, the equations are solved assuming a relative increase in the weight of methane in $\text{mg CH}_4/\text{g}$. The values of the components of the equation are as follows:

$$D_e = 0.952 \cdot 10^{-9} \text{ cm}^2/\text{s}$$

$$\bar{V}_\infty = V_r = 3.001 \text{ mg CH}_4/\text{g}, \text{ corresponding sorption capacity in standard conditions } q_s = 4.579 \text{ cm}^3/\text{g},$$

$$R = 0.0179 \text{ cm}$$

After substituting the data in equations (7) and (8) and considering the time distribution t obtained during tests, the adsorption curve was obtained. The calculations are the *ex post* forecast of the increase in the weight of the methane obtained in the laboratory.

The indirect results of the test of sorption properties conducted with gravimetric system IGA-001,

are automatically recorded in the computer using *IGASwin* software in the form of 1. DAT files, which can then be converted and opened with *Microsoft Excel*. It must also be emphasised that the data recorded by gravimetric system IGA-001, in this case included 772 measurements of an increase in the weight of the methane.

The calculations will be conducted in two ways:

- > variant I – substituting the values of the effective coefficient of diffusion calculated according to Timofeev's equation, i.e. $D_e = 0.952 \cdot 10^{-9} \text{ cm}^2/\text{s}$,
- > variant II – the value of coefficient D_e is treated as an element tuning the model, substituting various values within the range which are similar to the value assumed in variant I and observing which value of D_e results in the lowest value of mean relative forecast error *ex post* Ψ ; in order to simplify the process, the *Microsoft Excel Solver* add-in can be applied.

The calculations made according to variant II are also assumptions for the methodology of determining the effective coefficient of methane diffusion in coal which is an alternative to the methodology which uses the Timofeev equation.

Figs. 3 and 4 present the methane sorption (adsorption) kinetics curve for the analysed coal sample from coal seam 404/4 in the Pniówek coal mine, obtained with both the laboratory method and the computational method, after substituting data in equations (7) and (8) according to variant I and comparing the course of methane adsorption for the coal sample, obtained with the laboratory and analytical methods according to J. P. Seidle's model in the given variant.

The comparison shows that the value of the coefficient of determination $R^2 = 0.9956$, corresponding to Pearson's correlation coefficient $r = 0.9978$, indicates nearly full correlation between the results recorded during the laboratory tests and the results obtained with the analytical method. The value of mean relative error is $\Psi = 1.64\%$.

Figs. 5 and 6 below present the curve of methane sorption kinetics (adsorption) for the tested coal sample from seam 404/4 of the Pniówek coal mine, obtained with the laboratory method and the computational method, after substituting data in equations (7) and (8) according to variant II; and the comparison between methane adsorption courses for a coal sample, obtained with the laboratory and analytical methods according to J.P. Seidle's model in the variant.

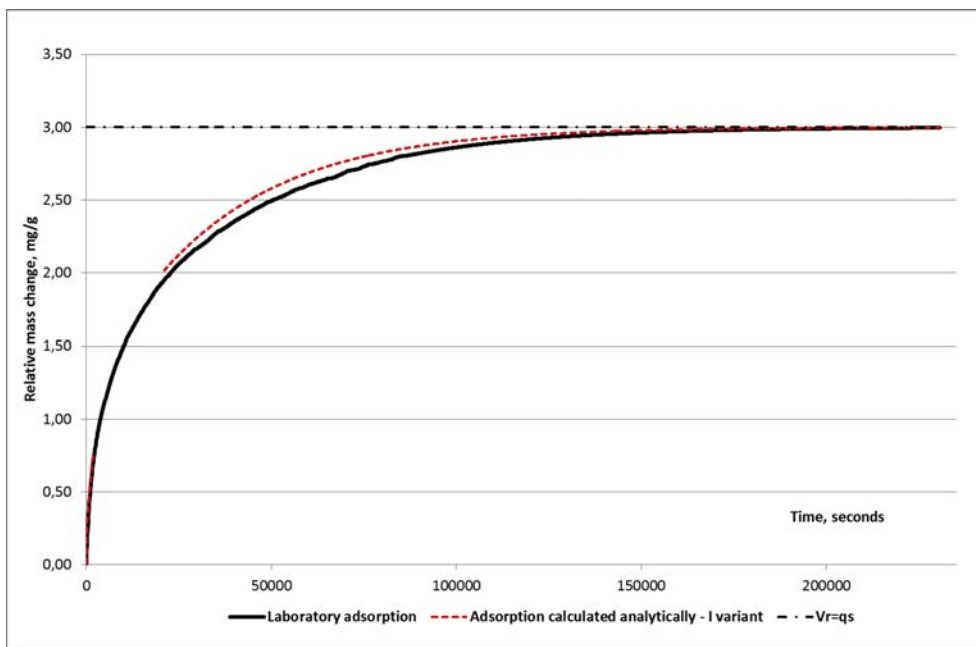


Fig.3. Graph of methane sorption kinetics for the coal sample (seam 404/4 Pniówek coal mine), obtained with laboratory and analytical methods, variant I.

The smallest mean relative error *ex post* was obtained at the value of effective coefficient of diffusion $D_e = 0.835 \cdot 10^{-9} \text{ cm}^2/\text{s}$.

The above comparisons show that the value of coefficient $R^2 = 0.9996$, which corresponds to coefficient $r = 0.9998$, also demonstrates nearly full correlation between the results recorded during laboratory tests and the results obtained with the analytical method. In this instance the value of

mean relative error became even lower, $\Psi = 0.27\%$.

Taking into consideration all of the facts, it can be concluded that the equations following the assumptions of J. P. Seidle's methane sorption model, based on Fick's second law, accurately describe the phenomenon in both variants of the calculations. In variant II, the value of mean relative error *ex post* is approximately 84% lower than in variant I, which

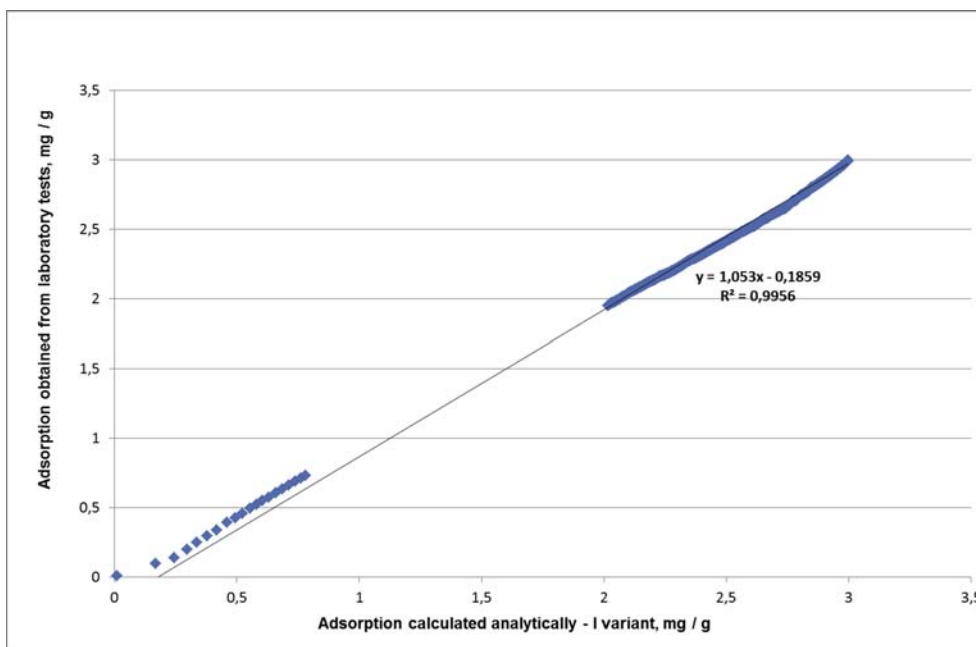


Fig.4. Comparison of methane adsorption courses, obtained with laboratory and analytical methods, variant I.

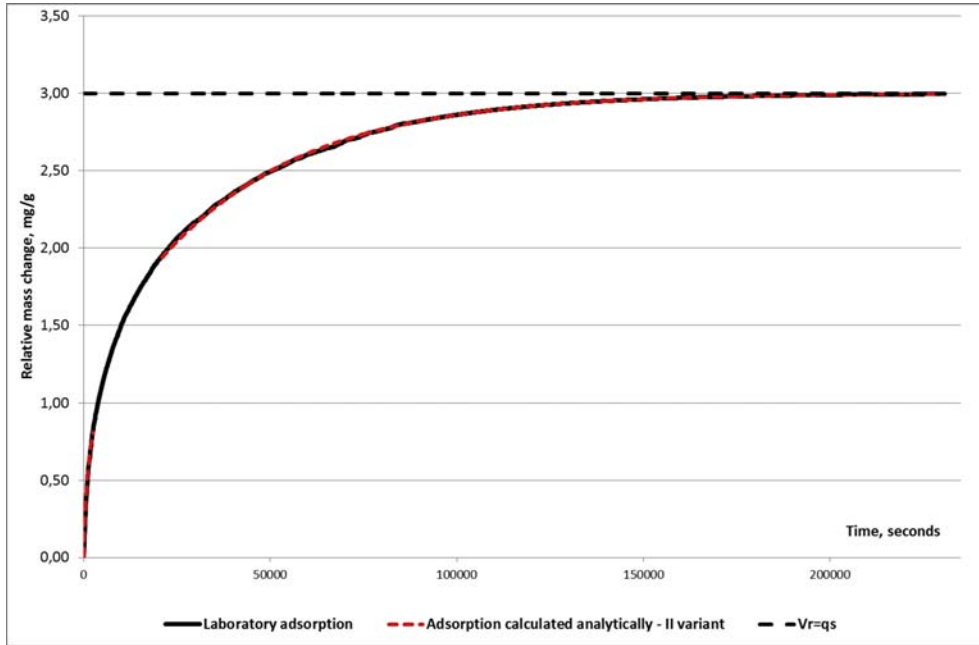


Fig.5. Graph of methane sorption kinetics for the coal sample (seam 404/4 Pniówek coal mine), obtained with laboratory and analytical methods, variant II.

shows that this method of the determination of the effective coefficient of methane diffusion in coal D_e is more accurate.

4. Results and discussion

1. During mining works in a highly gassy deposit, sorption properties of coal strongly influence the volume and kinetics of methane emission, as shown by the sorption kinetics graphs based on laboratory tests conducted with the gravimetric

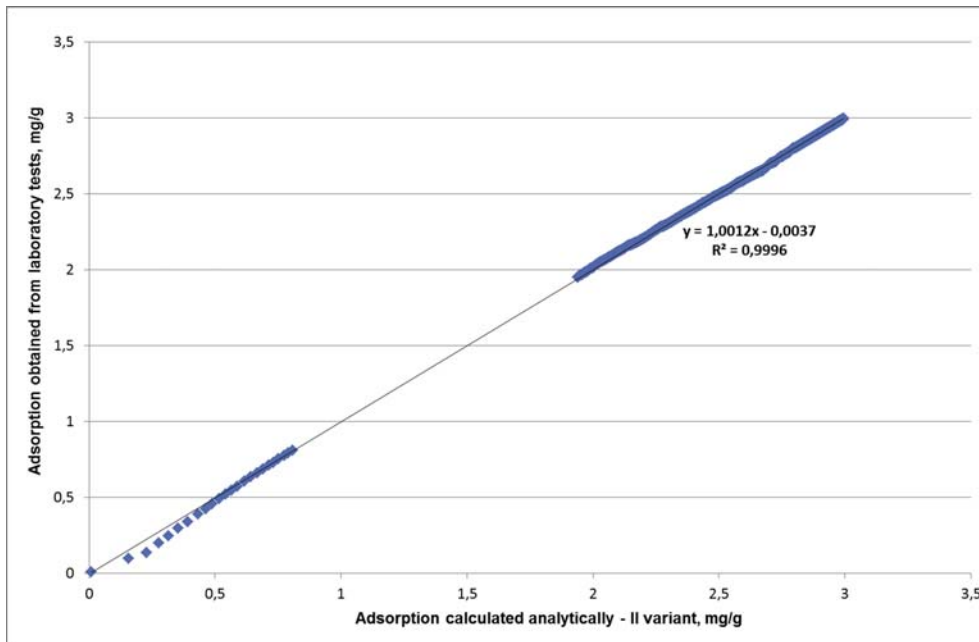


Fig.6. Comparison of methane adsorption courses, obtained with laboratory and analytical methods, variant II.

and sorption system. To characterise the ability of hard coals to accumulate and release methane, the tests determine the sorption capacity of coal in standard conditions and the value of the effective coefficient of diffusion D_{er} .

2. The 2-variant attempt to describe the sorption kinetics graphs with equations which following J.P. Seidle's assumptions of methane sorption kinetics model, based on Fick's second law for the isotropic radial diffusion, shows that the assumptions made for the mathematical model are correct and the model itself describes properly the phenomenon in both variants of calculations.
3. The value of coefficient D_{er} , determined as an element which tunes the mathematical model of sorption (variant II of calculations), makes it possible to model the sorption kinetics graph determined in laboratory conditions, better than the value calculated following the Timofeev equation (the value of mean relative error *ex post* is approximately 84% lower).
4. Development of more accurate methodology to determine the effective coefficient of methane diffusion in coal also paves the way for further research aimed at the determination of: the gas bearing capacity of coal seams, the degree of the degassing of coal seams as a result of mining operations, or to what extent it is possible to degas deposits.
5. Results of this work will make it possible to obtain more accurate assessments of the possible dynamics of methane emissions during mining operations under methane hazard and gas and rock outburst hazard in hard coal mines.

Conflict of interest

None declared.

Ethical statement

Authors state that the research was conducted according to ethical standards.

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