

Article

The Influence of Temperature on the Expansion of a Hard Coal-Gas System

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Abstract: This paper presents experimental results of the sorption-dilatometric kinetics of methane and carbon dioxide on a sample of hard coal from one of the coal mines in the Upper Silesian Coal Basin. The experiment included isothermal and non-isothermal-isobaric stages. For the isothermal stage, it was found that, up to a certain point (about 8 dm³/kg), the sample's expansion was linearly related to the amount of gas absorbed. Studies on dilatometric kinetics under constant pressure, with a reduction in temperature, indicate that a dominant share of the heat-expanding properties of carbonaceous material influences changes in the size of the sample in the coal-gas system. It was also found that the sample expansion, due to temperature change, was 2.25%, for the sample in both the vacuum and the non-adsorbing gas atmosphere.

Keywords: coal; methane; carbon dioxide; sorption; swelling

1. Introduction

The chemical and physical structure of hard coal makes the physico-chemical properties of the coal-gas system depend, to a large extent, on the type of gas and its pressure and temperature. In addition, the high heterogeneity of coal means that the issues of interaction involving gas molecules and pairs of different substances with carbonaceous material remain valid in relation to the technique of exploiting coal beds (gas and rock outbursts, ventilation). In recent years, they have been extended to include issues related to the sequestration of carbon dioxide [1–4] and the intensification of associated methane extraction, i.e., enhanced coal bed methane (EBCM) technology [5–7].

Studies on the kinetics of sorption and the diffusion of gases and vapours in relation to hard coals have been conducted for several years [6,8–12]. Previous work has considered coal to be a biporous, transport-sorption system, which, under the influence of high pressure from gases, results in microporous areas being compressed and macropores expanded, while the swelling of the microporous coal substance accompanying the sorption process narrows transport pores and decreases the permeability of the system [13,14]. While knowledge is increasing, there are still areas where there are no unambiguously consistent results. One of these areas is the expansion of coal caused by the accumulation of gases (mainly methane and carbon dioxide). In the literature, one can find the results of research indicating the existence of a linear relationship between the swelling of coal and the magnitude of the sorption [15,16]. There are also studies that show the course of sorption processes and the magnitude of the accompanying deformations as not existing in a linear relationship [17–21]. These discrepancies hinder attempts to generalize certain phenomena, which would be helpful, for example, in modelling the process of sequestration or extraction of methane from coal beds. In this type of technology, the transport process seems to be crucial, with the phenomenon of changes in coal rock volume being very unfavourable, owing to the possibility of closing transport and sorptive pores.

In addition, in the case of gas desorption, the endothermicity of the process changes the temperature of the system. Therefore, information resulting from the sorption-dilatometric kinetics conducted in an isothermal system is insufficient. This justifies the need to conduct research in a non-isothermal area.

The presented research results refer to previously published work [22], which described sorption-dilatometric results obtained at 298 K on a sample of coal from the Pniówek Mine in Poland. The results of isothermal measurements were extended to include non-isothermal measurement, where changes in the dimensions of the sample were followed as the temperature increased. Non-isothermal measurement; however, did not allow us to answer questions about the direct influence of pressure and temperature on the expansion of coal. The desorption process usually causes the shrinkage of the coal matrix. During the experiment, the pressure of the free gas changed (it increased as a result of the temperature increase), while the increase in the temperature of the system caused the release of gas from the sample. However, desorption was not the only factor influencing the volume changes in the sample. The influence of the expansion of the carbonaceous material itself, due to the change in temperature, was not determined.

The present article attempts to extend the experiment to include effects unexplained to date. A manostat was added to the measuring system, which eliminated the variability of pressure during the non-isothermal phase. In addition, the expansion of the coal sample itself was measured in a vacuum and in the atmosphere of a gas not subject to adsorption.

2. Research Material

The tests presented in the work were made for a selected sample of hard coal from the Pniówek Mine, taken from the 404/1 bed. Technical, elemental and petrographic analyses of the coal were performed. Selected results of the conducted analyses are presented in Tables 1 and 2.

Table 1. The results of the elemental and technical analyses of the research material.

| Sample/Bed | C ^{daf} [%] | S ^{daf} [%] | H ^{daf} [%] | N ^{daf} [%] | O ^{daf} [%] | W ^a [%] | A ^a [%] | V ^{daf} [%] |
|------------|----------------------|----------------------|----------------------|----------------------|----------------------|--------------------|--------------------|----------------------|
| Pn2(404/1) | 84.96 | 0.58 | 4.60 | 1.70 | 3.76 | 0.68 | 3.78 | 25.50 |

The elemental analysis was carried out at the Central Mining Institute in Katowice. Moisture content was determined in accordance with the PN-80/G-04511 standard and ash content in accordance with the PN-80/G-04512 standard.

Table 2. The petrographic composition of the research material.

| Group of Macerals | Vitrinite | Liptinite | Inertinite |
|-------------------|-----------|-----------|------------|
| Content [%] | 53 | 8 | 39 |

Hard coal can be very heterogenous with respect to chemical and petrographic composition, even over short distances (<1 km) [23,24]. The use of a relatively large, solid sample, with a mass of approximately 20 g, allows for better representation of the coal bed being studied, compared to literature data, where, very often, small samples, often not exceeding 1 g, are used. A cuboid with dimensions of 18 × 18 × 40 mm was cut out from the coal matrix for the purpose of sorptive-dilatometric measurements. Subsequently, strain gauges, oriented lengthwise and transversely to the bedding, were glued on the opposite walls of the sample.

3. Apparatus

An apparatus designed and built by the authors was used to carry out the measurements. The apparatus makes it possible to perform simultaneous measurements of sorption-dilatometric magnitudes for two solid samples in the form of a cube (Figure 1). Sorption capacity was measured using the manometric method, with pressure being measured using pressure transducers (P1, P2, P3). The dosing part of the apparatus, a high-quality pressure transmitter model S-10 (P3) (WIKA www.wika.com), operating in the pressure range from 0 to 100 bar, with an accuracy class of 0.25% BFSL

(best fit straight line), was used. The measurement of pressure in the ampoules (P1 and P2) (Sample Cell 1 and 2) was carried out using UniTrans pressure transmitter UT-10 (WIKA www.wika.com). The transducers make it possible to measure pressure up to 100 bar and have an adjustable measuring range on the 1:20 scale. The accuracy of the devices is 0.1% of the measuring range. The apparatus is placed in a water thermostat, which maintains a constant temperature with an accuracy of 0.1 K. A detailed description of the test methodology and calculations has been previously presented [22]. Modified measurements were obtained using a manostat attached to the measuring system, making it possible to maintain the set constant pressure in the device [25].

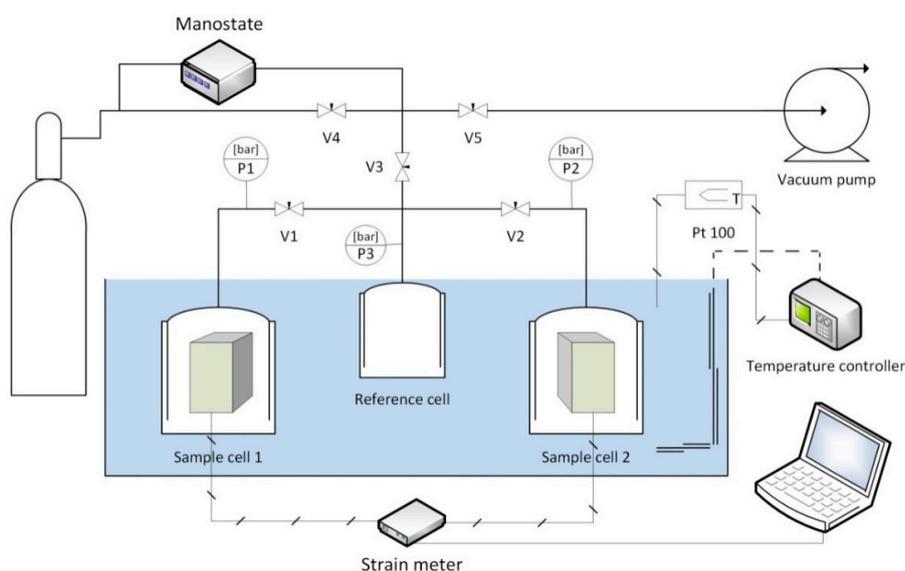


Figure 1. Schematic diagram of the apparatus.

Linear distortions of the sample were measured using a strain gauge constructed at the Strata Mechanics Research Institute of the Polish Academy of Sciences. The instrument uses electro-resistance strain gauges, while also using bridge resistance transducers. Real-time measurements are presented on an ongoing basis by means of an alphanumeric display, which can be saved in the internal memory of the device or transmitted externally via a serial link. Programming the meter includes entering the parameters of a set of connected strain gauges or transducers, setting the current time and determining the criterion for measurement data recording. A strain gauge essentially consists of a resistive element, which is glued with a special glue to the sample in a suitable configuration. The terminals for connecting the strain gauge to the cables, which run to the measuring apparatus, are most commonly soldered to the resistive element. The physical principle of this meter's operation involves correlating the measured change in the resistance of the conductor resulting from the change in its diameter, which occurs as a result of longitudinal stretching or shrinking along with the surface of the coal being subject to dilatometric phenomena [26].

4. Experiment Procedures

A coal cube, with strain gauges glued to it, was placed in a measuring ampoule, then connected to a tensometric strain gauge. To determine the sorption capacity, the dead volume of the system was determined using helium. Subsequently, the system was degassed in order to achieve a static vacuum of 10^{-2} Pa. After closing all the valves and zeroing the tensometric strain gauge, the system was ready to start the main part of the experiment. Gas (methane/carbon dioxide) was introduced into the reference cell. The recording of sorption-dilatometric kinetics was started upon the introduction of the sorbent to the measuring part by opening the V1 valve. The initial pressure, due to which the sorption-dilatometric changes took place, was approximately 11.1 bar for all measurements.

The kinetics of the linear deformation of the sample were recorded in the internal memory of the tensometric strain gauge sensor. The kinetics of sorption-dilatometric changes with respect to both sorbates were carried out at 323 K and recorded for 40 h.

After the isothermal stage, the next stage of the experiment, i.e., the non-isothermal and isobaric stage, was started. The manostat was set in such a way that it maintained pressure at the level of values established at the end of the isothermal stage. Next, the deformation measurement was started, accompanied by a decrease in the temperature of the system from 323 to 298 K, at a constant pressure value.

The final stage was to determine the changes in the dimensions of the sample accompanying the change in the temperature in the system, in which there were no sorbing gases. First, the sample was degassed, after which the changes in linear dimensions were recorded as the system temperature increased from 298 to 323 K. Following this, the system was cooled to 298 K. In the subsequent second test, helium was added to the system up to 1 bar pressure and the previously described cycle was repeated.

5. Analysis of Results

The first stage of the research consisted of sorption-dilatometric measurements taken at 323 K. The experimental results are presented in the form of sorption kinetics and volume deformations in Figure 2.

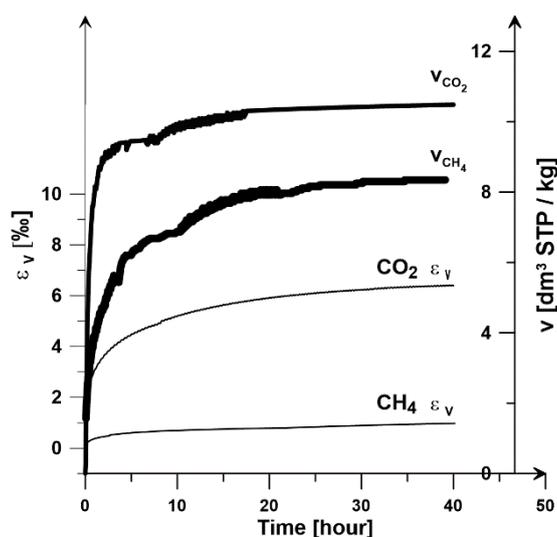


Figure 2. Sorption-dilatometric kinetics of the tested sample with respect to CO₂ and CH₄.

The course of the kinetics of sorption of both gases is typical [17,27–29], while the sorption capacity of the coal sample in relation to carbon dioxide is higher than for methane. In the case of dilatometric kinetics, the volume changes associated with the absorption of carbon dioxide are almost five times higher than those induced by methane. To analyse the effect of the quantity of sorbed gas on the swelling of coal, a graph of the volume change in the sample, as a function of the amount of gas absorbed, was created (Figure 3).

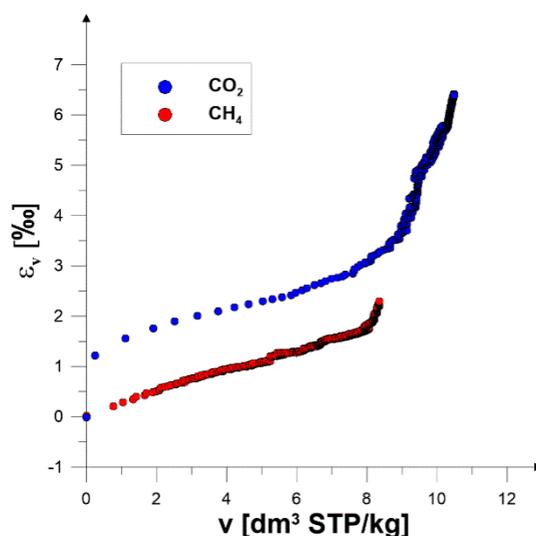


Figure 3. The dependence of the size of the volume deformation as a function of the sorption capacity of the tested sample.

Uptake of gas by coal is the complex effect of several mechanisms. In the coal-gas system, we mainly deal with the surface phenomenon (adsorption) and the volumetric dissolution of gas in the coal matrix (absorption) [30]. Given the difficulty of determining the contribution of individual phenomena, the uptake of gas by coal is best described by the term ‘sorption’. The dual mechanism of the uptake of gas in coal causes its swelling, thus the analysis of the impact of the amount of sorbed CO₂ or CH₄ on coal expansion should indicate the dominant contribution of any of the indicated mechanisms [31]. Data from the available literature indicate a possible linear relationship between sorption and swelling [16,32]; however, there are also studies in which this linearity has not been demonstrated [33,34]. The results presented in the paper (Figure 3) highlight the bimodal nature of the relationship. In the initial filling range (up to about 7–8 dm³/kg), the volumetric expansion increases linearly for both gases. The practically uniform inclination of both relationships allows us to assume that the adsorptive gas absorption mechanism dominates in this area, while expansion is related to the stretching of micro- and submicropores by the adsorbed molecules. Another range (>8 dm³/kg) is also characterized by a linear relationship, where there is a significant increase in sample volume expansion with a relatively small increase in the amount of adsorbed gas. The swelling effect can be attributed to the dominant role of gas absorption in the coal matrix, which is significant for CO₂ and low for methane [31,35]. As such, the explanation for such discrepancies in the literature findings is as follows. The ambiguous effect of gas sorption on swelling is related to many factors. The degree of coalification, as well as the porous coal structure and the associated permeability, is important [36]. The second factor is temperature, given that the kinetics of adsorption and absorption phenomena will be strictly dependent on it. In the system under study, it can be seen that the difference in adsorption and absorption kinetics is significant and translates into the course of the relationship shown in Figure 3.

The results of measurements of the dilatometric kinetics, measured together with decreasing temperature from 323 to 298 K, without changing the pressure prevailing in the system (isobaric conditions), are shown in Figures 4 and 5.

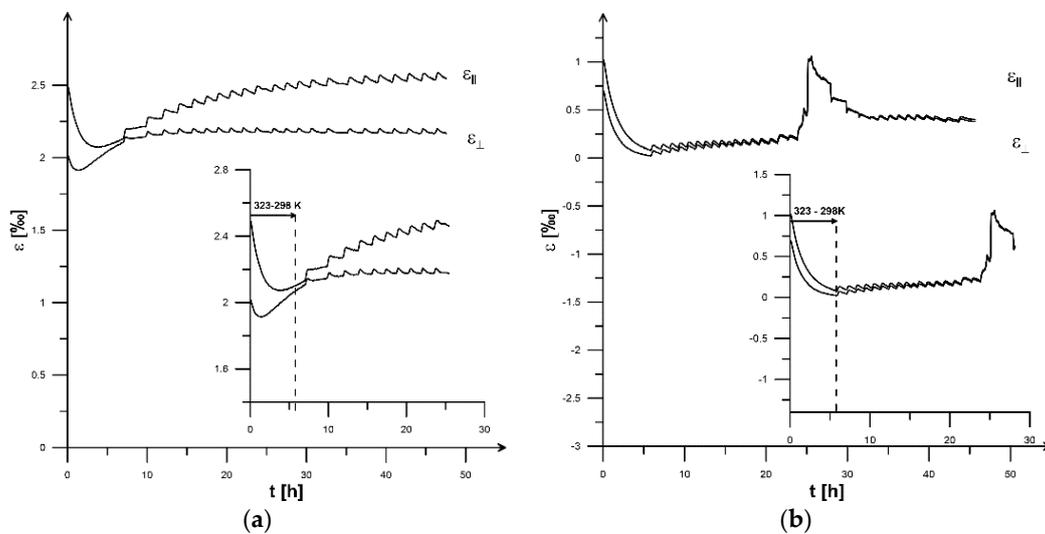


Figure 4. Kinetics (in the direction parallel ε_{\perp} and perpendicular ε_{\parallel} to the bedding plane) of the coal sample linear strain during temperature decreases for (a) CO_2 and (b) CH_4 .

As sorption is an exothermic process, lowering the temperature of the system will favour the intensification of this phenomenon. The effect of an increase in sorption capacity does not, however, directly affect the course of changes in the linear dimensions of the coal sample (Figure 4). An increase in sorption capacity should be accompanied by swelling of the sample. With carbon dioxide, the studied system initially begins to shrink dramatically (Figure 4a), while relaxation begins just before reaching 298 K. In the case of methane, shrinkage of the system is also observed, the changes are more dynamic, and the dimensions after 50 h are slightly smaller than before the non-isothermal stage begins. In the course of the kinetics of dilatation in the methane-coal system, an artefact is observed, the occurrence of which is attributed to unexpected electrical signals, which should, in no way, be related to the physical behaviour of the sample.

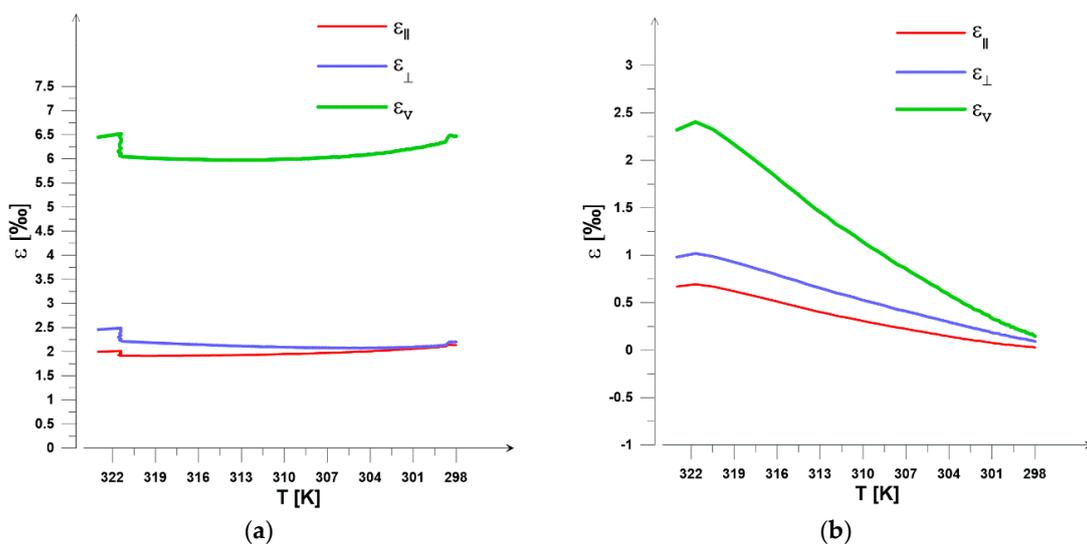


Figure 5. Volumetric (ε_v) and linear strains during temperature decreases for (a) CO_2 and (b) CH_4 .

The non-isothermal course of deformation changes, as a function of temperature, is shown in Figure 5. In the case of a system containing carbon dioxide, changes in temperature do not cause any changes in the dimensions of the sample; rather, in the case of methane, monotonic shrinkage is observed. Thus, it can be concluded that sorption processes do not play a key role in the studied

system, given that an increase in the magnitude of sorption does not cause swelling of the sample. Instead, it indicates the effect of the physical properties of the carbonaceous material. In the case of carbon dioxide, changes in the linear dimensions measured in the perpendicular and parallel directions are closely aligned. With isothermal measurements, the changes in the perpendicular direction are greater. This lack of similarity can be explained by the anisotropy of porosity changes as a result of coal shrinkage, which changes with temperature [37]. To determine the thermal expansion of the sample itself, measurement of the linear changes in the sample was carried out in a vacuum-degassed system (Figure 6a). To determine whether the vacuum does not cause physical shrinkage of the sample, which would limit expansion under the influence of temperature, an analogous measurement was performed in a system to which helium with a pressure of 1 bar was added (Figure 6b).

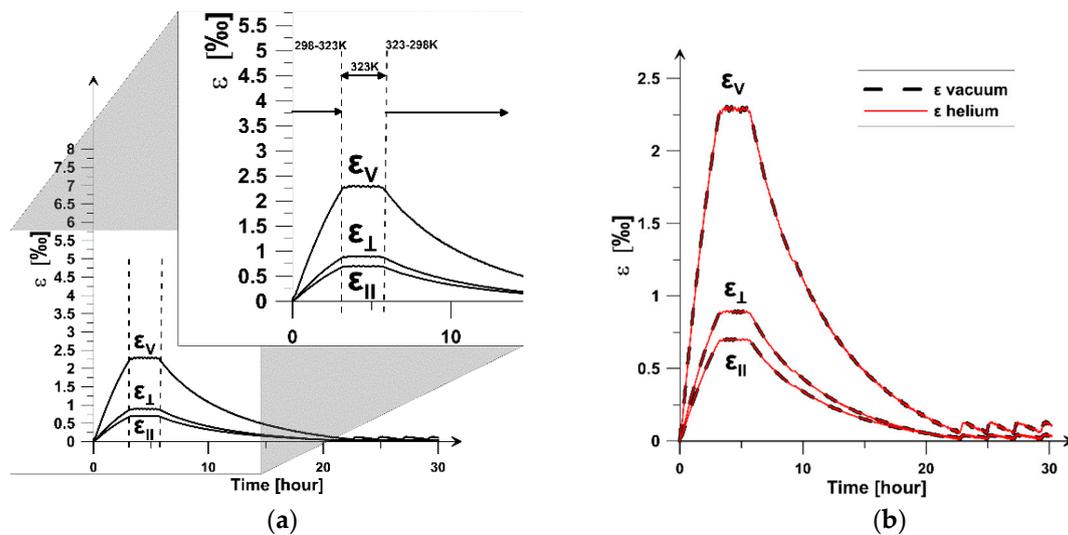


Figure 6. Thermal expansion of the coal sample (a) in the vacuum, and (b) in the vacuum and in the helium atmosphere.

It was found that the changes in linear and volumetric dimensions are identical, regardless of whether the system is in the vacuum or the coal cube is relaxed in the presence of a non-absorbing gas. Anisotropy of thermal expansion is also observed. The sample expands more in the direction perpendicular to the layering. Expansion anisotropy can be derived from the anisotropy of coal porosity and permeability [37]. The measured volume change in the vacuum, in the presence of helium, is high, which confirms that, in the case of non-isothermal processes in the coal mine gas system, the expansion of coal is the result of the coal's physical properties. Sorptive properties are also affected, where the impact of the latter is much greater in the case of carbon dioxide.

It should also be emphasized that volume changes in coal, caused by sorption-desorption of mine gas, will strongly affect the change in coal permeability. Low permeability is a big problem in the exploitation of coal bed methane (ECBM). In practice, vertical or/and horizontal wells are made to increase the permeability of the bed [38–40]. The temperature also affects the change in permeability [41]; however, the experimental results of the methane and CO₂ sorption kinetics do not allow for a reliable estimation of the permeability of the tested coal sample.

The assessment of the impact of temperature on expansion in the coal-gas system should, therefore, include several components, which are not easy to interpret. The results presented in previous work [22], where measurement of temperature changes was performed and the free gas pressure was not constant, were slightly different. The dimensional changes resulting from the thermal expansion of coal itself were not considered, while the pressure, which changed along with the temperature, caused interpretation problems. Determining the pressure in the experiment presented in the current study eliminated one variable and added to our knowledge of the coal-gas system.

6. Summary

The experiment of sorption-dilatometer kinetics determined at a constant temperature confirmed the previously published results. The expansion of the coal sample, due to the sorption of methane, is lower than that of carbon dioxide. The interdependence of sorption capacity and volumetric expansion, measured at 323 K for both gases, is linear to a certain point, after which a change to a higher inclination was observed. This proves the dominant role of adsorption kinetics over the kinetics of gas absorption by coal.

The measurements made in the system, subjected to a temperature change, while maintaining the constant value of the free gas pressure, provide interesting information about the nature of coal, which is of practical importance. The measurements showed that, together with a decrease in the temperature of a system containing a gas-saturated sample, the sample shrinks, after which the system relaxes. The analysis of the temperature range change showed that, in the coal-CO₂ system, a reduction in temperature did not change the volume dimensions of the sample. It can, therefore, be concluded that thermal expansion is entirely compensated by the expansion caused by gas absorption. Such behaviour will be beneficial in the process of carbon dioxide sequestration in coal beds, because possible changes in the temperature of the bed will not cause volume changes in the coal, which may lead to unsealing.

In the case of the coal-CH₄ system, lowering the temperature results in large shrinkage of the sample, whose relationship is practically linear. In this situation, the change is mainly a derivative of the thermal conductivity properties of the carbonaceous material. The contribution of the expansion caused by methane sorption is negligible. This is confirmed by the experiment carried out for a degassed system, for which the value of reduction in volumetric expansion with temperature change is close to the value measured in the coal-methane system.

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