



Effect of bituminous coal properties on carbon dioxide and methane high pressure sorption



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HIGHLIGHTS

- The effect of coal properties on CO₂ and CH₄ sorption capacity of coal.
- Parametrization of excess sorption isotherms using Langmuir and D–R Eqs.
- Different petrographic composition did not reflect in a variation of porosity.
- Macerals of inertinite group presumably contribute to CO₂ sorption capacity.
- The effect of microporous clay minerals on the sorption capacity was not distinct.

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ABSTRACT

High pressure sorption experiments with carbon dioxide and methane were carried out at a temperature of 45 °C and at pressures up to 15 MPa with three samples of methane-bearing, medium-rank coals in a moisture-equilibrated state using a manometric method. The samples were taken from selected positions of drill cores from exploration boreholes in the Bohemian part of the Upper Silesian Basin, and were characterized by a narrow range of degree of coalification and markedly different petrographic compositions, including a different mineral matter content. The total porosity of the coal samples was between 9% and 10%. A positive correlation was found between the equilibrium moisture in the coal samples and the total abundance of oxygen functional groups determined by FTIR. The excess sorption capacities ranged from 0.78 to 0.91 mmol g⁻¹ for CO₂ and from 0.45 to 0.52 mmol g⁻¹ for CH₄, and after recalculation to coal organic matter, the excess sorption capacities increased by up to 14% in the coal with the highest mineral fraction. The highest CO₂/CH₄ ratio was found in the sample that had the highest inertinite and liptinite content. The experimental isotherm data was fitted by modified Langmuir and Dubinin–Radushkevich sorption isotherms. The parameters obtained by these two methods were in good agreement for carbon dioxide. It was found that the sorption capacity of the organic matter in a coal sample with prevalence of inertinite (63.0 vol.%) was lower only by 14% for CO₂ and by 18% for CH₄ than the sorption capacity of the organic matter in a coal sample with prevalence of vitrinite (65.3 vol.%). This provided confirmation that the petrographic composition of a coal has an ambiguous effect.

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1. Introduction

Although coal seams have a lower capacity for deposition of carbon dioxide than saline aquifers and depleted oil and gas reservoirs [1], the relatively new injection technology is considered to be economically advantageous, especially when the repository is located near a power plant that produces carbon dioxide. Another positive feature of this technology is that carbon dioxide injected into a deep unmineable methane-bearing seam simultaneously increases the production of coal methane as a valuable energetic resource.

The suitability of coal seams for use as carbon dioxide repositories is based on some exceptional properties of coal. During the coalification process, coal seams retain in their interior spatial arrangement a substantial part of the porous structure of the original plant material, with a predominance of cavities with an effective size of <2 nm. The major storage mechanism is by sorption processes that take place in the porous system of the coal. The effectiveness of the sorption process depends on the properties of the coal matter, the seam environment, and the carbon dioxide under the *in situ* conditions.

One of the basic parameters for selecting a suitable repository on the basis of a simulation of the deposition process is its sorption capacity. This capacity can be determined in the laboratory by

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measuring the amount of carbon dioxide captured in a coal sample at a pressure and a temperature corresponding to supercritical conditions *in situ*, using high pressure sorption techniques. Similarly, the amount of methane bound in the coal is calculated on the basis of a high pressure measurement of the amount sorbed on it. According to principles of adsorption/desorption reversibility, this value should be roughly proportional to the amount of gas released by either spontaneous or forced degassing.

Storage of CO₂ in coal seams is a topical issue in the early 21st century. Following the introduction of experimental procedures for determining coal sorption capacity, which have been verified in interlaboratory comparisons of results obtained on defined coal samples [2–5], great attention has also been paid to the reasons for errors in determining the quantities, and to the consequences of these errors [6,7]. Research has focused on efforts to improve the accuracy with which sorption capacity is determined, and also on the effect of the properties of the coal matter on the sorption process, namely the effect of the degree of coalification, maceral composition, mineral substances, and moisture. Most of the present state of knowledge has been reviewed by White et al. [8] and by Busch and Gensterblum [9]. The complexity of coal matter and the variability of its properties cause ambiguous correlations in relation to sorption capacity, as numerous previous studies have shown. The role of the degree of coalification in relation to sorption capacity has not yet been fully elucidated, though there has been general agreement that the sorption ability of coal can be a function of the degree of coalification, with sorption capacity increasing with the degree of coalification [8]. However, several studies have failed to confirm this finding [10,11]. Similarly, many authors have confirmed a positive correlation between maceral composition and sorption capacity, which increases with an increasing total content of vitrinite and decreases with an increasing inertinite content [12–17], whereas other authors have failed to observe such a correlation [18,19]. Attention has been paid not only to the effect of coalification and maceral composition on sorption capacity, but also to the role of mineral matter in a coal. Previous research has shown that the mineral matter in a coal can be considered as an inert component with a minimum effect on microporosity and sorption capacity [8]. According to Mastalerz et al. [20], mineral matter can increase or decrease the specific surface and mesopore volume of bulk coal, depending on the magnitude of the specific surface area of the accompanying organic fraction, and it can also contribute to a reduction in micropore volume in the original coal. The presence of water in coal can adversely influence the sorption behavior of other molecules [8]. The impact of water on gas sorption is studied especially in the context of CO₂ sequestration in coal seams. Brennan et al. [21] have reported that water clusters formed at or across the opening of the pore can effectively block the entire pore space, thereby hindering the access of other sorbing molecules to the pores. For an exact simulation of *in situ* conditions, CO₂ sorption isotherms are measured mainly on moist coal.

The present study has aimed to investigate the effect of coal properties on the carbon dioxide and methane sorption capacity of coal. Three samples of bituminous coal from the Bohemian part of the Upper Silesian Basin were selected for the purposes of our study. These three samples had similar degrees of coalification, as reflected by the narrow range of light reflectance from 1.02% to 1.10%, but they had significantly different maceral compositions.

2. Experimental

2.1. Samples and sample characterization

Three samples of methane-bearing medium-rank coal from selected positions of drill cores from exploration boreholes in the

Bohemian part of the Upper Silesian Basin were selected for the study. The main criterion in selecting the samples was a narrow range of the degree of coalification, as characterized by light reflectance, and different petrographic composition. Sample No. 1 was collected in the Václavovice exploration area from the bottom of the Hrušov Member in the Ostrava formation by the VA – 1H Datyně bore from a depth of 664.2–664.6 m. Sample No. 2 was collected from the DP-1 Dětmárovice – Petřkovice bore from a depth of 1138.0–1139.0 m from the vicinity of seam No. 33 (605), which is the last seam of the Suchá Member in the Karviná formation. Sample No. 3 comes from the TR – 2 Frenštát bore of the Trojano-vice mine from a depth of 1109.2–1109.6 m in the middle part of seam No. 40-Prokop (504) from the Saddle Member of the Karviná formation.

In all experiments, a fraction with grain size in the range of 0.25–0.05 mm prepared from each coal sample was used. The coals were crushed with minimum fines to less than 0.25 mm.

Proximate analysis was carried out, and ash yield (A^d), water (W^a), volatile matter (V^{daf}), and total sulphur (S_t^d) contents were determined according to national standards. Ultimate analysis was carried out using a Thermo-Finnigan CHNSO microanalyzer 2000. The basic qualitative parameters of the studied coals are given in Table 1.

The equilibrium moisture content was provided according to the procedures for the ASTM D1412 Standard Test Method for Equilibrium Moisture of Coal at 96–97 percent Relative Humidity and 30 °C. The wetted samples were immediately transferred into a sorption cell. Proper aliquots from the samples were used for determining the equilibrium water content (W_{eq}).

2.1.1. Petrographic analysis

From the fraction with grain size 0.25–0.05 mm, two grain polished sections were prepared for determining the mean light reflectance of vitrinite R_o and the maceral composition. The reflectance of vitrinite in coal grain polished sections was measured using a Nikon Eclipse ME 650 microscope (immersion objective, 50× magnification, oil immersion with refractive index $n = 1.515$) with a LUCIA VITRINITE image analysis system for measuring vitrinite reflectance. The maceral composition of the vitrinite, liptinite and inertinite groups, and the composition of the mineral matter, were assayed using a Opton-Zeiss UMSP 30 Petro microscope (reflected light, wavelength $\lambda = 546$ nm, immersion objective, 40× magnification, oil immersion with refractive index $n = 1.518$), and an ELTINOR point-counter for precise determination of the studied petrographic components according to ISO 7404 and Taylor et al. [22].

2.1.2. Textural analysis

The parameters characterizing the microporous texture of the studied coal samples, i.e., the total micropore volume V_{micro} , the modulus of the micropore radii r_{mode} , and the micropore distribution were determined on the basis of the sorption isotherm of CO₂ measured at a temperature of 25 °C up to a relative pressure of 0.015, using a Hiden IGA 002 gravimetric sorption analyzer. Before the

Table 1
Proximate and ultimate analyses.

Sample	Parameter (wt.%)								
	W^a	W_{eq}	A^d	V^{daf}	C^{daf}	H^{daf}	N^{daf}	O^{daf}	S_t^d
1	1.20	2.47	8.50	27.47	85.03	5.25	2.31	7.42	≤0.1
2	1.25	2.54	12.24	26.92	83.50	4.41	2.58	8.90	≤0.1
3	1.30	2.78	1.16	26.61	85.83	5.02	2.60	6.55	≤0.1

W – water, W_{eq} – equilibrium moisture, A – ash, V – volatile matter, a – analytic, d – dry basis, daf – dry and ash-free basis, t – total.

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