Coalification as a process determining the methane adsorption ability of coal seams

The paper presents the results of a study of methane adsorption on coal samples with various degrees of metamorphism, coming from the Polish and Czech parts of the Upper Silesian Coal Basin (USCB). The range of coalification of the samples was from bituminous with vitrinite reflectance $R_o$ equal to about 0.5% to para-anthracite coals with $R_o$ equal to over 2%. The methane adsorption capacity was determined at the temperature 303 K for each of the studied coal seams. Methane adsorption isotherms were approximated using the Langmuir model. The relationship between the Langmuir isotherm parameters ($a_m$ and $P_L$) and the degree of coalification was presented. It was shown that the degree of coalification of the coal substance affects the adsorption ability of coal with respect to methane and determines the value of the Langmuir isotherm parameters. The study was conducted in order to present the distribution of adsorption capacity of Upper Silesian coals in relation to improving work safety in active mines as well as designing technologies that use coal bed methane (CBM) from balance and off-balance resources.

Keywords: coal bed methane, degree of coalification, petrography, adsorption capacity

1. Introduction

Coal seams are considered to be unconventional sources of natural gas. The methane they contain takes the form of gas adsorbed in the coal matrix and in the free state in its fractures (coal bed methane – CBM) [1,2]. Depending on the pressure and temperature conditions, the methane adsorbed in the porous coal matrix can be described with a density close to that of a liquid [3]. The clear advantage of unconventional methane resources is that they can provide a margin of
energy security for the country [4]. The size of these resources is significant, and due to their several times lower CO₂ emissions, they are a greener alternative to the process of burning coal or oil. The disadvantage of unconventional reserves of coal bed methane is the need to use new technologies for extraction of these resources.

The presence of methane in coal seams is associated primarily with a serious threat to the safety of work of underground crews in active coal mines and our energy market is still based almost entirely on coal [5]. Due to the complicated geological and mining conditions and the unique nature of the work, mining activities in seams saturated with methane involve many accidents and disasters. Increasing natural hazards, such as methane and rock outbursts, are particularly adversely affected by steadily increasing mining depths [6]. Increasing depth of burial leads to accelerated maturation of the deposits and to increasing rank [7].

The adsorption capacity of coal is extremely important in many issues related to studies of rock mass, safety of underground work, and engineering solutions that may involve coal seams together with the methane contained in them. This parameter determines the amount of gas that can be adsorbed per unit of volume of the studied coal. The value of adsorption capacity is influenced by a number of factors, such as, among others, degree of coalification, pressure, temperature, moisture content, petrographic composition, and lithostatic loading [8-11]. Each of the aforementioned parameters affects the value of adsorption capacity to a different degree; however, the degree of coalification is decisive for the adsorption behavior of coal in relation to the gases contained in the seams [8]. Due to the fact that most of the methane present in the seam is in an adsorbed form, the adsorption capacity is a major component of methane-bearing capacity [12].

The degree of coalification of the coal substance is one of the most important parameters for determining the qualitative properties of coal [13,14]. The evolution of organic remains from peat through brown coal (lignite), sub-bituminous and bituminous coal, to anthracites and meta-anthracites is referred to as “coalification”. Coalification comprises a diagenetic stage (from peat to brown coals) and a metamorphism stage for highly coalified brown coals up to meta-anthracites. Three factors influence the increase in the degree of coalification: temperature, time, and pressure. As coalification increases, individual parameters of the sediment change, such as decreased porosity and increased anisotropy, as well as changes in the quantitative chemical composition: an increase in the quantity of carbon and a decrease in the quantity of hydrogen and oxygen (Schürmann rule) and decrease in the volatile content (Hilt’s rule) [15]. Along with these changes, a change in optical properties, such as reflectance of individual macerals, also takes place [16,17] as well as in a number of other properties, such as, among others, strength parameters and adsorption capacity [13,14,18,19].

Despite many studies on the influence of the degree of coalification on the adsorption capacity of coal [12,20-22], no experimental studies have been carried out on samples with a wide range of coalification taken from the whole Upper Silesian Coal Basin (USCB): its Czech and Polish parts. The advantage of the paper is the base of 31 samples of hard coal obtained from seams covering almost the whole lithostratigraphic profile of the coal measures of the USCB (Fig. 1).

The purpose of the paper is to determine the influence of the degree of coalification on the adsorption ability of coal in relation to methane on the basis of tests of carried out on a wide group of samples taken from both Polish and Czech coal seams of the USCB. The research described in the article allows for a more accurate prediction of the accumulation properties of Upper Silesian coal seams of a known maturity.
2. Geological context

The analyzed coal samples come from deposits of various ages from the Polish and Czech parts of the Upper Silesian Coal Basin. The basin covers an area of about 7,250 km² and is currently the largest underground mining basin in the European Union [23]. The Polish part of the USCB is located in the Silesian-Cracow Upland and the Czech part – in the Moravian-Silesian Region. The coal deposit there was formed during the Variscan orogeny, mainly during the Asturian phase [24]. It is a mountain foredeep developed in the form of a synclinal basin filled with Carboniferous formations of the Upper Carboniferous period. The formation of coal in the USCB was possible due to specific environmental conditions prevailing in the Carboniferous period. Initially, this entire area was periodically flooded by the sea. It then transformed into plains cut by a network of meandering rivers. The marshy terrain, the hot and humid climate, and the high oxygen content of the atmosphere was conducive to lush and rapid growth of plants, which reached enormous sizes. Invertebrates and soil microorganisms could not keep up with the decomposition of the dead debris that sank into the extensive marsh areas. In marshes with an anaerobic environment, the organic matter that the dead plants were made of did not completely decompose and organic matter was subject to the coalification processes. These processes, acting cyclically, first led to the formation of numerous accumulations of peat and then, as a result of the gradual burial of the sediment, due to increasing pressure and temperature, the peat was transformed into numerous deposits of lignite, then hard coal, and sometimes anthracite [25].

3. Laboratory tests

3.1. Test material

A total of 31 coal samples were taken for testing, covering the Upper Carboniferous (Silez) strata – Namur and Westphalian (Fig. 1). The samples were taken from the Polish and Czech parts of USCB:

- the Czech part: the Ostrava Formation (“Pietrzkowice”, “Gruszów”, and “Poręba” member) and the Karviná Formation (Saddle and “Dobrava” members).
- the Polish part: seams from the Upper Silesian sandstone series (Saddle and “Ruda” members) and siltstone series (“Załęże” and “Orzesze” members), and from the Cracow sandstone series (“Łaziska” members) (Fig. 1).

3.2. Research methods

3.2.1. Basic analyses

A technical analysis of coals was carried out by determining the content of volatile matter and ash. The analysis was supplemented with measurements of the real density of the coals.
3.2.2. Microscopic analysis

Petrographic analyses of the coal samples were performed along with determination of the degree of coalification. Microscopic examinations were carried out on polished grain preparations prepared according to the recommendations contained in the PN-ISO 7404-2:2005 and PN-ISO 7404-3:2001 standards. A ZEISS AXIOPLAN polarizing microscope and a computer-controlled XYZ mechanical table were used for the petrographic analyses. The analyses were performed under reflected-white light, using oil immersion and a 500× magnification. A point quantitative analysis was carried out on 1,500 measurement points to determine the content of petrographic components of coal (main groups of macerals and mineral matter). Mean reflectance ($R_o$) was also measured for each of the coals, on impurity- and inclusion-free maceral fragments from the vitrinite (colotelinite) group. An Olympus BX50 polarizing microscope was used to measure reflectance with a 400× magnification. The polished preparations were analyzed under monochromatic light using oil immersion. The results were obtained through the “LUCIA Vitrinite” image analysis system [32].
3.2.3. Methane adsorption tests

Methane adsorption tests were carried out on the coal samples of 0.125-0.16 mm grains using the IGA-001 gravimetric adsorption analyzer made by Hiden Isochema. The dry coal material was degassed for 12 h at 353 K until a high vacuum of 1 Pa was reached. The adsorption measurements consisted in determining the adsorption capacity of coal at 303 K for the following methane pressures: 1 bar (ambient), 4 bar, and 12 bar. The results were given for standard pressure and temperature conditions (STP) and for the dry-ash free state.

The Langmuir isotherm of the following form was used to quantitatively describe the methane adsorption process in the studied coals [20]:

\[ \frac{a(p,T)}{a_m} = \frac{p}{P_L + p} \]  

(1)

where: 
- \( a \) – volume of adsorbed methane at pressure \( p \), \( m^3 CH_4/M_g_{sw} \), \( p \) – equilibrium pressure of methane, bar, \( T \) – temperature, K, \( a_m \) – maximum adsorption capacity of coal at \( p \) reaching \( \infty \), \( m^3 CH_4/M_g_{sw} \) and \( P_L = \frac{1}{b} \) – half-adsorption pressure (Langmuir pressure), bar.

Although certain assumptions of the Langmuir model cannot be satisfied for coal [34], the function [1] is useful for adsorption studies of the coal-methane system. Thanks to its simple form and the agreement with experimental data demonstrated by numerous authors, the function is applicable to the description of the adsorption equilibrium state of almost all types of coal. There are two constants in the Langmuir isotherm equation: \( a_m \) and \( P_L \). These constants are referred to as Langmuir isotherm parameters. They contain information about the course (shape) of the hyperbolic isotherm, as well as the aspects of the adsorption process itself. The \( a_m \) constant determines the maximum adsorption capacity of coal at the maximum pressure of the system and assuming a finite number of adsorption sites (so-called monolayer capacity). The \( P_L \) constant is the so-called Langmuir pressure, which corresponds to the pressure at which the coal adsorption surface is filled with a half of the quantity specified by the \( a_m \) value. It is responsible for the shape of the adsorption isotherm and determines its initial course. The lower its value, the steeper the slope of the initial portion of the isotherm, which implies stronger methane adsorption at low equilibrium pressures.

4. Results and discussion

4.1. Characteristics of the coals

The results of the laboratory tests carried out, which included technical, densimetric and microscopic analysis and adsorption gravimetric tests for 31 coal samples, are presented in Table 1. As the degree of coalification increases, the volatile content shows a decreasing trend. The ash content of the samples varies from 2.58 to 18.81%, while the real density of the coals varies with coalification on a U-shaped trend basis with a minimum for the \( R_o \) value of about 1.1%.
## Characteristics of the coal samples

<table>
<thead>
<tr>
<th>Sample</th>
<th>(R_o) [%]</th>
<th>(V_{daf}) [%]</th>
<th>(ρ_{He}) g/cm³</th>
<th>(A_a) [%]</th>
<th>(V_{t mmf}) [%]</th>
<th>(I_{mmf}) [%]</th>
<th>(L_{mmf}) [%]</th>
<th>(\alpha_m\ CH_4)</th>
<th>(P_L\ CH_4)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coal 1</td>
<td>0,54</td>
<td>39,63</td>
<td>1,394</td>
<td>8,41</td>
<td>59,34</td>
<td>31,66</td>
<td>9,00</td>
<td>19,815</td>
<td>7,87</td>
</tr>
<tr>
<td>Coal 2</td>
<td>0,70</td>
<td>39,32</td>
<td>1,477</td>
<td>12,00</td>
<td>54,57</td>
<td>34,20</td>
<td>11,23</td>
<td>18,803</td>
<td>6,90</td>
</tr>
<tr>
<td>Coal 3</td>
<td>0,78</td>
<td>40,97</td>
<td>1,429</td>
<td>14,58</td>
<td>85,08</td>
<td>4,76</td>
<td>10,15</td>
<td>15,789</td>
<td>7,94</td>
</tr>
<tr>
<td>Coal 4</td>
<td>0,79</td>
<td>36,61</td>
<td>1,361</td>
<td>4,99</td>
<td>68,82</td>
<td>17,81</td>
<td>13,36</td>
<td>15,001</td>
<td>8,66</td>
</tr>
<tr>
<td>Coal 5</td>
<td>0,85</td>
<td>36,45</td>
<td>1,395</td>
<td>10,54</td>
<td>76,71</td>
<td>16,44</td>
<td>6,84</td>
<td>15,900</td>
<td>7,46</td>
</tr>
<tr>
<td>Coal 6</td>
<td>0,86</td>
<td>36,59</td>
<td>1,407</td>
<td>5,65</td>
<td>80,00</td>
<td>16,00</td>
<td>4,00</td>
<td>14,063</td>
<td>6,71</td>
</tr>
<tr>
<td>Coal 7</td>
<td>0,91</td>
<td>36,41</td>
<td>1,393</td>
<td>14,03</td>
<td>71,00</td>
<td>20,00</td>
<td>9,00</td>
<td>17,019</td>
<td>6,99</td>
</tr>
<tr>
<td>Coal 8</td>
<td>0,98</td>
<td>24,78</td>
<td>1,380</td>
<td>6,00</td>
<td>72,73</td>
<td>21,27</td>
<td>6,00</td>
<td>18,419</td>
<td>4,77</td>
</tr>
<tr>
<td>Coal 9</td>
<td>1,02</td>
<td>28,45</td>
<td>1,390</td>
<td>10,19</td>
<td>76,39</td>
<td>18,42</td>
<td>5,19</td>
<td>17,440</td>
<td>5,46</td>
</tr>
<tr>
<td>Coal 10</td>
<td>1,04</td>
<td>20,58</td>
<td>1,389</td>
<td>11,57</td>
<td>75,56</td>
<td>21,36</td>
<td>3,08</td>
<td>17,330</td>
<td>5,70</td>
</tr>
<tr>
<td>Coal 11</td>
<td>1,05</td>
<td>27,84</td>
<td>1,435</td>
<td>2,58</td>
<td>72,49</td>
<td>23,35</td>
<td>4,15</td>
<td>15,160</td>
<td>5,04</td>
</tr>
<tr>
<td>Coal 12</td>
<td>1,07</td>
<td>20,2</td>
<td>1,324</td>
<td>5,56</td>
<td>87,55</td>
<td>9,78</td>
<td>2,67</td>
<td>15,483</td>
<td>5,71</td>
</tr>
<tr>
<td>Coal 13</td>
<td>1,09</td>
<td>19,89</td>
<td>1,356</td>
<td>6,73</td>
<td>62,90</td>
<td>30,55</td>
<td>6,55</td>
<td>16,620</td>
<td>5,31</td>
</tr>
<tr>
<td>Coal 14</td>
<td>1,11</td>
<td>23,25</td>
<td>1,373</td>
<td>6,31</td>
<td>76,12</td>
<td>18,50</td>
<td>5,38</td>
<td>15,371</td>
<td>5,85</td>
</tr>
<tr>
<td>Coal 15</td>
<td>1,15</td>
<td>21,44</td>
<td>1,326</td>
<td>8,76</td>
<td>43,39</td>
<td>53,28</td>
<td>3,33</td>
<td>15,148</td>
<td>5,21</td>
</tr>
<tr>
<td>Coal 16</td>
<td>1,16</td>
<td>17,4</td>
<td>1,321</td>
<td>4,38</td>
<td>55,33</td>
<td>42,31</td>
<td>2,36</td>
<td>17,077</td>
<td>5,22</td>
</tr>
<tr>
<td>Coal 17</td>
<td>1,19</td>
<td>18,73</td>
<td>1,326</td>
<td>4,28</td>
<td>77,07</td>
<td>22,11</td>
<td>0,81</td>
<td>14,432</td>
<td>6,04</td>
</tr>
<tr>
<td>Coal 18</td>
<td>1,25</td>
<td>13,82</td>
<td>1,380</td>
<td>6,48</td>
<td>50,99</td>
<td>41,45</td>
<td>7,56</td>
<td>14,627</td>
<td>4,75</td>
</tr>
<tr>
<td>Coal 19</td>
<td>1,42</td>
<td>18,55</td>
<td>1,394</td>
<td>18,81</td>
<td>78,25</td>
<td>17,40</td>
<td>0</td>
<td>15,863</td>
<td>5,07</td>
</tr>
<tr>
<td>Coal 20</td>
<td>1,65</td>
<td>15,49</td>
<td>1,513</td>
<td>5,58</td>
<td>77,50</td>
<td>20,55</td>
<td>0</td>
<td>16,652</td>
<td>5,56</td>
</tr>
<tr>
<td>Coal 21</td>
<td>1,67</td>
<td>12,92</td>
<td>1,427</td>
<td>3,03</td>
<td>77,80</td>
<td>21,60</td>
<td>0</td>
<td>16,950</td>
<td>4,86</td>
</tr>
<tr>
<td>Coal 22</td>
<td>1,73</td>
<td>12,76</td>
<td>1,433</td>
<td>3,50</td>
<td>76,60</td>
<td>22,50</td>
<td>0</td>
<td>19,040</td>
<td>4,83</td>
</tr>
<tr>
<td>Coal 23</td>
<td>1,85</td>
<td>14,26</td>
<td>1,408</td>
<td>17,14</td>
<td>82,43</td>
<td>17,57</td>
<td>0</td>
<td>22,166</td>
<td>4,98</td>
</tr>
<tr>
<td>Coal 24</td>
<td>1,87</td>
<td>13,91</td>
<td>1,378</td>
<td>8,83</td>
<td>85,51</td>
<td>14,49</td>
<td>0</td>
<td>19,753</td>
<td>4,79</td>
</tr>
<tr>
<td>Coal 25</td>
<td>1,87</td>
<td>10,59</td>
<td>1,455</td>
<td>3,98</td>
<td>78,20</td>
<td>21,15</td>
<td>0</td>
<td>19,018</td>
<td>4,81</td>
</tr>
<tr>
<td>Coal 26</td>
<td>1,89</td>
<td>11,17</td>
<td>1,467</td>
<td>5,10</td>
<td>83,60</td>
<td>15,70</td>
<td>0</td>
<td>19,810</td>
<td>4,61</td>
</tr>
<tr>
<td>Coal 27</td>
<td>1,89</td>
<td>12,34</td>
<td>1,437</td>
<td>7,73</td>
<td>76,85</td>
<td>21,65</td>
<td>0</td>
<td>20,035</td>
<td>4,79</td>
</tr>
<tr>
<td>Coal 28</td>
<td>1,91</td>
<td>12,07</td>
<td>1,422</td>
<td>10,01</td>
<td>75,05</td>
<td>21,65</td>
<td>0</td>
<td>19,966</td>
<td>5,03</td>
</tr>
<tr>
<td>Coal 29</td>
<td>1,94</td>
<td>13,62</td>
<td>1,403</td>
<td>7,54</td>
<td>91,29</td>
<td>8,71</td>
<td>0</td>
<td>20,781</td>
<td>4,78</td>
</tr>
<tr>
<td>Coal 30</td>
<td>2,01</td>
<td>13,51</td>
<td>1,497</td>
<td>9,02</td>
<td>88,18</td>
<td>11,82</td>
<td>0</td>
<td>20,624</td>
<td>4,57</td>
</tr>
<tr>
<td>Coal 31</td>
<td>2,03</td>
<td>12,8</td>
<td>1,387</td>
<td>6,87</td>
<td>78,60</td>
<td>21,40</td>
<td>0</td>
<td>22,269</td>
<td>4,78</td>
</tr>
</tbody>
</table>

\(R_o\) – vitrinite reflectance, \(V_{daf}\) – volatile matter, \(A_a\) – ash yield, \(ρ_{He}\) – real density, \(V_{t mmf}\) – vitrinite content, \(I_{mmf}\) – inertinite content, \(L_{mmf}\) – liptinite content

### 4.2. Petrography

According to the international classification of coal in seams [33], the analyzed samples include medium-coalified bituminous coals (medium-rank D, C, B and A – para, ortho, meta, per) and highly coalified anthracitic coals (high rank C – para) (Fig. 2). According to classification ASTM [15] the analyzed samples belong from subbituminous coal through high volatile bituminous, medium volatile bituminous, low volatile bituminous to semiantracite coals.

In the analyzed coals, as the degree of coalification, determined by measuring the average of vitrinite reflectance \((R_o)\), increases, the volatile content \((V_{daf})\) decreases (Fig. 3) \((R^2\) fitting at
It has been observed that in coals with a low and medium degrees of coalification ($R_o$ from about 0.5 to about 1.3%), the volatile content decreases much more rapidly as $R_o$ increases. In the diagram (Fig. 3) this is indicated by the red straight line, which descends at an acute angle.

Fig. 3. Dependence of the degree of coalification ($R_o$) on the volatile content ($V_{daf}$). Blue line – the trend line for all samples ($R^2 = 0.86$); red line – the relationship for samples with $R_o$ from 0.8% to 1.3%; green line – the relationship for samples with coalification above 1.3%.

The graph clearly shows that both parameters are closely linked to the coal sludge coalification. It has been observed that in coals with a low and medium degrees of coalification ($R_o$ from about 0.5 to about 1.3%), the volatile content decreases much more rapidly as $R_o$ increases. In the diagram (Fig. 3) this is indicated by the red straight line, which descends at an acute angle from
the coals with about 40% to about 18% \( V_{\text{daf}} \). In other, older coals \( R_o \) from about 1.3% to more than 2%, the volatile content decreases somewhat more slowly (at a smaller angle) in relation to the degree of coalification. This is illustrated by the green line.

Quantitative spot analyses showed that the variation of individual maceral groups is not directly dependent on the age of the seam, but as the coalification increases, a constant domination of vitrinite over inertinite is evident in the “pietryzkowickie” and “gruszkowskie” seams. Typically, regardless of the age of the sample, vitrinite is the dominant maceral, but locally there are samples where there is an increase in inertinite content. In the USCB, this type of lithological variation is not an anomaly. This has been discussed by e.g. [35], who noted an increased inertinite content in the Zofiówka monocline. This is confirmed by the research conducted for that publication. The coals from the Zofiówka region, which belong to the “Ruda” and “Saddle” member contain as much as over 50% by volume (mmf) of macerals of the inertinite group (Tab. 1). In other locations and strata, the inertinite content is only slightly elevated in single samples, but vitrinite usually dominates. Macerals of the liptinite group are the least numerous group making up the analyzed coal samples. Usually their content does not exceed a few per-
cent. Only in relatively young coals of the “Doubrava” and “Poręba” member and in the Czech coals from the “saddle” member, whose degree of coalification ($R_o$) does not exceed 1%, single samples with liptinite content of ten to twenty percent were found (Tab. 1). In the case of coals with $R_o > 1.3$, microscopic analyses do not show any presence of liptinite (Tab. 1), because optical features make this maceral similar to vitrinite (due to the so-called vitrinization of liptinite); therefore, only vitrinite and inertinite are determined in these samples.

Coal, depending on the degree of coalification, varies in color in the microscopic image. The fundamental difference is most easily observed in the grey shades of the individual macerals depending on the degree of coalification. In samples with a relatively low degree of coalification (0.53%), such as those shown in Fig. 5 (photos 1 and 2) vitrinite is dark grey, liptinite is almost black, and inertinite is grey. The more coalified the samples are, the more the macerals change their shades of grey to slightly lighter ones, as shown in Fig. 5 (photos 3 and 4), and in coals with coalification $R_o$ over 1.3-1.4%, liptinite is not observed and vitrinite is almost completely...
white, as shown in Fig. 5 (photos 5-8). Regardless of the degree of coalification, vitrinite is the dominant maceral in all of the coals analyzed. Only in single samples does the content of macerals from the inertinite group increase. Another petrographic parameter that differentiates coals of different degrees of coalification is the content of mineral matter. There are usually many more minerals in young coals than in mature sediments.

4.3. Adsorption test results

The methane adsorption isotherms determined at 303 K, for coal samples grouped according to their coalification degree intervals, are shown in Fig. 5. The arrangement of the isotherms shows that the adsorption capacity of coal with respect to methane changes in coal groups with different degree of metamorphism. It can be seen from Fig. 5a that the lowest methane adsorption capacity is found in coals with a low degree of coalification, which are characterized by reflectance in the range from 0.5 to 0.9%. An increase in the CH₄ adsorption capacity can be observed along with an increase in the degree of coalification of the samples taken from the investigated seams, and, at the same time, with a transition to groups of more metamorphised coals. In the case of medium-metamorphised coals, with reflectance in the range of 0.9% to 1.7%, a partial overlap of the methane adsorption isotherms shown in Fig. 5b with those for low-metamorphised coals shown in Fig. 5a can be observed. As a result, some of the medium-metamorphised coal samples show a similar methane adsorption capacity to that of the lower-metamorphised coals. The highest adsorption capacity of coal within the range of the analyzed equilibrium pressures of methane was obtained for highly metamorphised coal samples with reflectance from 1.7% to 2.03% (Fig. 5c).

Based on the methane adsorption isotherms plotted in Fig. 5, the Langmuir equation parameters $a_m$ and $P_L$ were determined and summarized in Tab. 1. The values of the maximum adsorption capacity $a_m$ of coals obtained from the approximation of isotherms with Langmuir model range from 14.06 to 22.27 cm³CH₄/g. The Langmuir pressure values $P_L$ are in the range from 4.6 to 8.7 bar. The relationships showing the effect of the degree of coalification on the values of the $a_m$ and $P_L$ parameters are shown in Fig. 6 and Fig. 7, respectively. As it may be seen from the graphs, both parameters change together with the degree of coalification of the coal substance.

Fig. 6 shows that in the reflectance range of 0.5-1.9% there is a symmetric U-shaped trend of changes in the maximum adsorption capacity with the degree of coalification, the minimum of which is reached for coals with reflectance around $R_o = 1.2\%$. The values of $a_m$ for coals with reflectance in the range of $R_o = 0.5$ to 0.9% (the lowest metamorphised coals) varied widely, but showed a decreasing trend with progressive coalification of the samples. For coals with reflectance of $R_o > 1.4\%$ (the upper range of medium-metamorphised coals and highly metamorphised coals), the results are completely different and the maximum adsorption capacity of coal in relation to methane increases almost linearly with coalification. Highly coalified coals with reflectance of $R_o > 1.7\%$ are characterized by the highest values of $a_m$ of all the examined seams.

The $P_L$ parameter of the Langmuir equation (Fig. 7), which determines the initial course of the isotherm, shows the tendency to decrease its value in the investigated range of coalification. For the low-metamorphised coals and some of the medium-metamorphised coals, the half-adsorption pressure decreases strongly with an increasing degree of coalification, after which, from the value of $R_o$ equal to about 1.2%, the decrease in the half-adsorption pressure $P_L$ is still evident, but less rapid. The most coalified coals with reflectance of $R_o > 1.7\%$ are characterized by the lowest values of $P_L$ of all the examined seams. For these coals, adsorption of methane
Fig. 5. Adsorption isotherms at 303 K for coal samples with different degrees of metamorphism. (a) low-metamorphised coal samples ($0.5 < R_o < 0.9\%$); (b) medium-metamorphised coal samples ($0.9\% < R_o < 1.7\%$); (c) highly metamorphised coal samples ($1.7\% < R_o < 2.03\%$)
occurs most intensively at low equilibrium pressures of methane. The observations made in our study are in line with those reported in work of [36], which discussed various factors that affect the values of the Langmuir isotherm parameters for different coalification stages.

The paper [14] shows the factors that can determine the adsorption capacity of coals with different coalification degrees. The trends of changes in the Langmuir parameters obtained in Fig. 6 and Fig. 7 allow to estimate their values for any coal sample with a known value of $R_o$. Then, the knowledge of Langmuir parameters enables the reconstruction of the adsorption isotherm in the range of up to 10 bar, and on its basis the adsorption capacity of coal at the selected pressure. Adsorbed methane is the dominant component of methane bearing capacity. Hence, the trends obtained in Fig. 6 and Fig. 7 make it possible to predict the content of coalbed methane based on the knowledge of $R_o$ value. Sometimes, without the need to carry out a time-consuming measurement of $R_o$, it is enough to know the value of $V^\text{daf}$ which is in close correlation with reflectance $R_o$ (Fig. 3).

**Fig. 6.** The effect of the degree of coalification on the maximum methane adsorption capacity of coal

**Fig. 7.** The relationship between the Langmuir pressure and the degree of coalification
5. Conclusions

The adsorption capacity of coal in relation to methane and the values of the Langmuir isotherm equation parameters \(a_m\) and \(P_L\) change for coals with different degrees of metamorphism. As we moved from low-metamorphised coals to coals with a higher degree of coalification, an increase in the adsorption capacity of coal was observed for methane pressure up to 10 bar. The highest adsorption capacity within the range of the analyzed equilibrium pressures of methane was obtained for highly metamorphised coal samples with reflectance from 1.7% to 2.03%. These coals were also characterized by the highest values of \(a_m\) among all the investigated seams.

A U-shaped trend of the maximum adsorption capacity \(a_m\) was obtained with a minimum for the coal with reflectance \(R_o = 1.2\%\), whereby the coals with the lowest degree of coalification \((R_o \text{ from } 0.5 \text{ to } 0.9\%)\) were characterized by a decreasing trend with increasing degree of coalification, while for coals with reflectance \(R_o > 1.4\%\) the maximum adsorption capacity \(a_m\) increased almost linearly with coalification.

The \(P_L\) parameter of the Langmuir equation shows the tendency to decrease its value in the investigated range of coalification. The most metamorphised coals are characterized by the lowest values of the half-adsorption pressure \(P_L\) of all the tested seams. For these coals, methane adsorption at low equilibrium pressures of methane is the most significant.

The obtained results show that the course of methane adsorption isotherms and the values of Langmuir parameters undergo significant changes along with changes in the coalification degree. The trends of Langmuir parameters changes obtained in Fig. 6 and Fig. 7 enable the reconstruction of the coalbed methane content in the Upper Silesian seams based on the knowledge of the \(R_o\) value or the \(V_{daf}\) value closely correlated with reflectance. Knowledge of the adsorption properties of seams helps to identify those potentially dangerous for underground work or those that offer prospects for the implementation of new technologies.

Acknowledgments

The presented work was supported financially by the Polish Ministry of Science and Higher Education through the statutory research fund.

References


