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## THE INFLUENCES OF THE RANK OF COAL ON METHANE SORPTION CAPACITY IN COALS

## WPŁYW RZĘDU WĘGLA NA POJEMNOŚĆ SORPCYJNĄ METANU W WĘGLACH

Methane sorption capacity is of significance in the issues of coalbed methane (CBM) and depends on various parameters, including mainly, on rank of coal and the maceral content in coals. However, in some of the World coals basins the influences of those parameters on methane sorption capacity is various and sometimes complicated. Usually the rank of coal is expressed by its vitrinite reflectance  $R_o$ . Moreover, in coals for which there is a high correlation between vitrinite reflectance and volatile matter  $V^{daf}$  the rank of coal may also be represented by  $V^{daf}$ . The influence of the rank of coal on methane sorption capacity for Polish coals is not well understood, hence the examination in the presented paper was undertaken. For the purpose of analysis there were chosen fourteen samples of hard coal originating from the Upper Silesian Basin and Lower Silesian Basin. The scope of the sorption capacity is: 15-42 cm<sup>3</sup>/g and the scope of vitrinite reflectance: 0,6-2,2%. Majority of those coals were of low rank, high volatile matter (HV), some were of middle rank, middle volatile matter (MV) and among them there was a small number of high rank, low volatile matter (LV) coals.

The analysis was conducted on the basis of available from the literature results of research of petrographic composition and methane sorption isotherms. Some of those samples were in the form (shape) of grains and others – as cut out plates of coal. The high pressure isotherms previously obtained in the cited studies were analyzed here for the purpose of establishing their sorption capacity on the basis of Langmuire equation.

As a result of this paper, it turned out that for low rank, HV coals the Langmuire volume  $V_L$  slightly decreases with the increase of rank, reaching its minimum for the middle rank (MV) coal and then increases with the rise of the rank (LV). From the graphic illustrations presented with respect to this relation follows the similarity to the Indian coals and partially to the Australian coals.

**Keywords:** methane, rank of coal, sorption capacity

Metanowa pojemność sorpcyjna węgla ma istotne znaczenie w zagadnieniach metanu pokładów węgla (CBM) i zależy od różnych parametrów, w tym głównie od rzędu węgla i zawartości meceralów w węglu. Jednakże zależność ta w wielu zagłębiach węglowych na świecie jest różnorodna i na ogół skomplikowana. Zazwyczaj rząd węgla jest wyrażany przez jego refleksyjność wityrynitową  $R_o$ . Ponadto

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w węglach, w których występuje wysoka korelacja między rzędem, a zawartością części lotnych  $V^{daf}$ , wówczas rząd węgla może być reprezentowany również przez  $V^{daf}$ . Wpływ rzędu na metanową pojemność sorpcyjną dla polskich węgli nie jest dobrze rozpoznany, dlatego w tej pracy podjęto próbę zbadania tego zagadnienia. Do przeprowadzenia analizy wybrano czternaście prób suchego węgla kamiennego pochodzącego z zagłębia górnośląskiego oraz dolnośląskiego. W tych węglach metanowa pojemność sorpcyjna mieściła się w zakresie 15-42 cm<sup>3</sup>/g, a refleksyjność wityrynitowa była w zakresie 0,6-2,2%. Większość z tych węgli była niskiego rzędu, o wysokiej zawartości części lotnych (HV), kilka było średniego rzędu, o średniej zawartości części lotnych (MV) oraz wśród nich była mała liczba węgli wysokiego rzędu, o niskiej zawartości części lotnych (LV).

Przeprowadzono analizę opartą o wyniki wcześniejszych badań spotykanych w literaturze, dotyczących wysokociśnieniowych izoterm sorpcji metanu oraz składu petrograficznego tych węgli. Niektóre z tych prób były w kształcie ziaren, a inne były płytkami wyciętymi z węgla. Wysokociśnieniowe izotermy sorpcji metanu uzyskane wcześniej w cytowanych pracach były analizowane w celu określenia pojemności sorpcyjnej na podstawie równania Langmuire. W efekcie tej pracy okazało się, że dla niskiego rzędu (HV) metanowa pojemność sorpcyjna węgla  $V_L$  nieznacznie maleje ze wzrostem rzędu, osiągając minimum dla średniego rzędu (MV), a następnie wzrasta ze wzrostem rzędu (LV). Z prezentowanych ilustracji graficznych odnośnie do tej zależności widać podobieństwo do węgli z basenu indyjskiego oraz częściowo do węgli z basenu australijskiego.

**Słowa kluczowe:** metan, rząd węgla, pojemność sorpcyjna węgla

## 1. Introduction

Poland possesses significant deposits of coalbed methane (CBM) as an economic resource. Thus far the attempts to obtain this methane from hard coal mines were difficult. Methane is often strongly bound with microporous (submicroporous) coal matrix. Moreover methane is absorbed as dissolved gas in coal matrix. In certain types of coal the internal surface area in micropores is large. CBM reservoir characterization is complex due to high degree heterogeneity and variation in petrographic composition of coals.

An important parameter in the CBM issues is the methane sorption capacity of coals. The coal type is described by the maceral content, including mainly the vitrinite and inertinite content. In addition, the methane sorption capacity of coal is also influenced by such factors, as: mineral matter content and moisture content.

In research on the influence of the petrographic composition on the methane sorption capacity of hard coal it was found that there are differences between bright and dull coals (Levine et al., 1993). Bright – vitrinite rich coals have a higher sorption capacity than dull inertinite rich coals of the same rank (Bustin & Clarkson, 1998). In a work (Laxminarayana & Crosdale, 1999) on the Australian coals a rather complicated nature of this relation was observed. Whereas, from the result of a work (Laxminarayana & Crosdale, 2002) on the Indian coals it follows, that methane sorption capacity and vitrinite content are unrelated with each other. A similar result can be reached under the conclusions of the work (Olajossy, 2013), which determines a weak relation between those two parameters with respect to low and middle rank Polish coals.

Coal rank may be determined by physical and chemical properties, which are influenced by coal type. To minimize the effect of the coal type, in the cited study the use of the vitrinite reflectance  $R_o$  as the rank parameter was chosen. For such types of coal, for which there is a high correlation between volatile matter  $V^{daf}$  and vitrinite reflectance, also the volatile matter can express the rank of coal. The lack of this correlation means that  $V^{daf}$  content is strongly influenced by coal type variations and the presence of carbonates.

A relationship between increasing sorption capacity and increasing rank of coal has been previously established (Yee et al., 1993), (Laxminarayana & Crosdale, 1999). However other study (Levy et al., 1997) reports a wide spread of data with almost no change in sorption capacity up to semianthracite rank followed by a sharp increase in adsorbed gas.

The relation of Polish coal rank to methane sorption capacity is not fully understood. In order to determine this relation, samples of hard coal from Upper Silesian Basin and Lower Silesian Basin were chosen. Their maceral contents and high pressure sorption isotherms were established previously in studies: (Ceglarska-Stefańska, 2000) and (Nodzeński, 2000). On the basis of the available from literature results of those works and also studies of few other authors (cited in the references of the present paper) the required analysis and discussion of their results were conducted. The obtained results may be compared with the conclusion of the research on the relation of methane sorption capacity from the rank of coal regarding the samples of other coals in the World.

## 2. Effect of the rank of coal on the sorption capacity of coals originating from number of different coal basins in the World

The methane sorption capacity is usually expressed by the Langmuire volume  $V_L$  ( $\text{cm}^3/\text{g}$ ), which depends on the rank of coal presented by the vitrinite reflectance  $R_o$  (%) of the dry coal. There were chosen three known from the literature types of those influences with respect to three coal basins in order to better present the differences indicated in the previous point.

The first analysis concerns dry coal samples from the lower Permian Barkan/Harabari Formation in India (Laxminarayana & Crosdale, 2002). These samples of dry, ash free hard coals are characterized by their high sorption capacity: 25-40  $\text{cm}^3/\text{g}$ , relatively low and middle rank:  $0,6 < R_o < 1,5\%$ . Mineral matter in Indian coals appears not to be significantly contributing to their methane sorption characteristics. However, there was observed a clear relation of sorption capacity with the moisture of those coals.

Vitrinite reflectance  $R_o$  has been used as an indicator of rank of coals in preference to the volatile matter  $V^{daf}$ , which is strongly influenced by coal type variations. Fig. 1 presented a decrease in the Langmuire's volume  $V_L$  with increasing rank as  $R_o$  for dry coals, India (Laxminarayana & Crosdale, 2002).

It is worth to mention, that in those samples of coals there were no high rank coals and anthracites. Certain differences between the bright and dull coals can be observed with respect to the examined relation.

The second analysis concerns the results obtained on the samples of dry coals from Bowen Basin, Australia (Laxminarayana & Crosdale, 1999). Those coals are characterized by high sorption capacity, within 35-50  $\text{cm}^3/\text{g}$  and a broad scope of changes of rank:  $0,5 < R_o < 3\%$  and lack of the content of liptinite. In those coals volatile matter  $V^{daf}$  is strongly correlated to vitrinite reflectance  $R_o$ , thus both of those parameters can represent the rank of coal. Trends in Langmuire sorption capacity  $V_L$  in relation to coal rank ( $R_o$ ) are presented on Fig. 2 (Laxminarayana & Crosdale, 1999).

This result indicates that  $V_L$  for bright and dull coal types follows discrete, second – order polynomial trends, with increasing rank. This non-monotonically function has a minimum  $V_L$  beside some value  $R_o$  for bright, low volatile coal and has other minimum  $V_L$  at some value  $R_o$  for dull, medium volatile coal. For high ranks of coal however the sorption capacity of both of

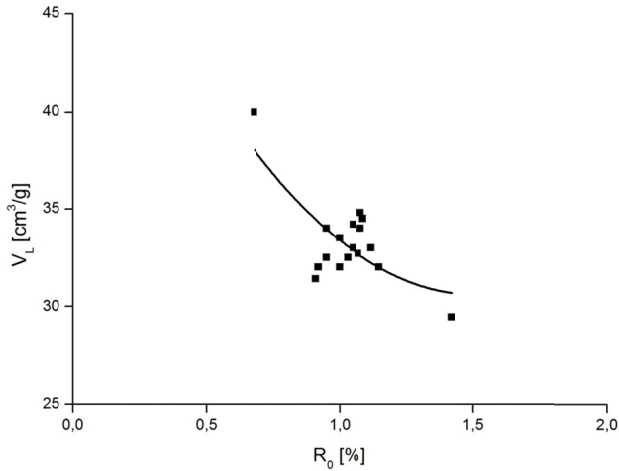


Fig. 1. The relation between methane sorption capacity and rank coal (India)

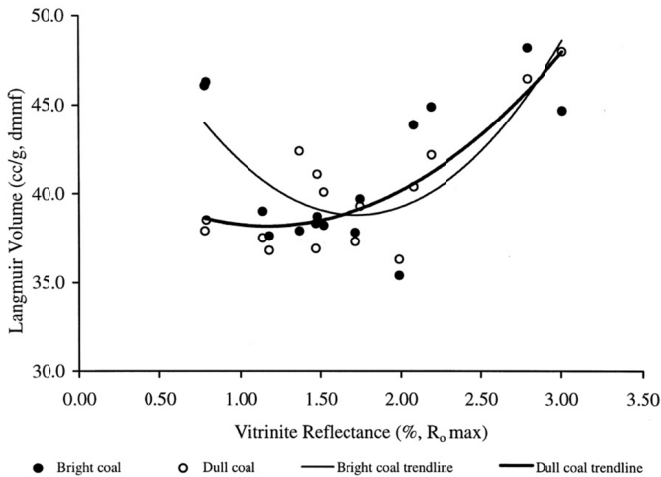


Fig. 2. Rank effects on adsorption capacity of dry coal (Australia)

those types of coals increases rapidly with the rise of  $R_o$  in a similar manner, thus the differences between bright and dull coals disappear. This conclusion is interesting.

The third analysis concerns the results of the research of dry hard coal samples from location within North America (Chalmers & Bustin, 2007). The coals have the values of methane sorption capacity in scope of changes: 5-25  $\text{cm}^3/\text{g}$  with scope of change of rank:  $0,6 < R_o < 4\%$ . The presence of mineral matter in the researched coals did not turn out to be significant. In those studies the presence of liptinite in the composition of macerals was separately considered.

Fig. 3 presents the relation between the methane sorption capacity (for chosen value of pressure 6 MPa) and a coal rank (Chalmers & Bustin, 2007).

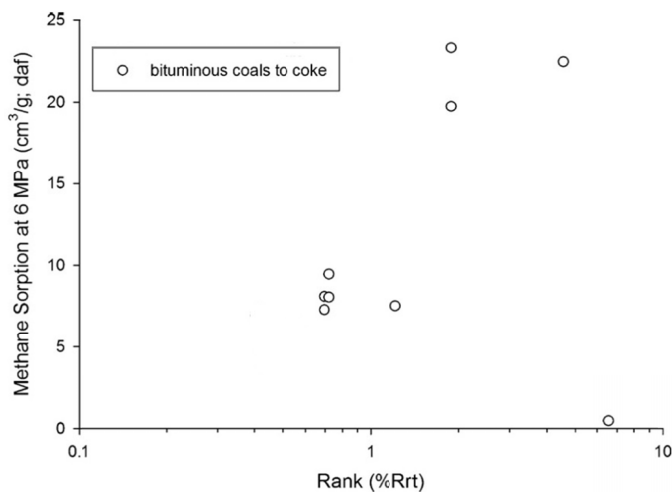


Fig. 3. Relationship between the methane sorption capacity and rank coal (North America)

The examined samples were divided in two groups: bituminous coals with low rank and higher rank groups, although not numerous, in order to show a positive relation between the methane sorption capacity and the rank of coal.

From the analyzed results of the cited papers there follows a varied character of relation  $V_L$  ( $R_o$ ). Therefore it is only natural to attempt to research this relation with regard to Polish coals.

### 3. The rank of coal and methane sorption capacity of dry coal (Poland)

For examination of the relation  $V_L$  ( $R_o$ ) and making of appropriate comparisons there were chosen fourteen samples of hard coals, including nine obtained from the Upper Silesian Basin and five from the Lower Silesian Basin. Some properties of those coals were presented in the study (Olajossy, 2013). Most of them are low rank, high volatile (HV) coals, some are middle rank, middle volatile (MV), whereas only few are high rank, low volatile (LV) and anthracite. The results of the research of isotherm of methane sorption on those coals and the results of the petrographic analysis of those coals obtained previously by various authors, mainly (Ceglarska-Stefańska, 2000; Nodzeński, 2000) were used.

Moreover the results of other works were used (Ceglarska-Stefańska & Brzóska, 1995), (Ceglarska-Stefańska & Zarebska, 2006; Nodzeński, 1998). On this basis, an analysis of methane sorption capacity was conducted (Olajossy, 2013) in which the Langmuire equation was used:

$$V = \frac{PV_L}{P_L + P}$$

or the linear form:

$$\frac{P}{V} = \frac{P_L}{V_L} + \frac{P}{V_L}$$

where  $V$  is the volume of adsorption gas at the pressure  $P$ ,  $V_L$  is Langmuire volume,  $P_L$  is Langmuire pressure. Some of the values of  $V_L$  were obtained within the scope of high pressures up to 8 MPa and others up to 4,5 MPa.

The relation between the rank of coal of these coals and their volatile matter is presented on Fig. 4. High correlation  $r^2 = 0,98$  can be presented with the assistance of the linear approximate:

$$V^{daf} = - 21,32 R_o + 51,62$$

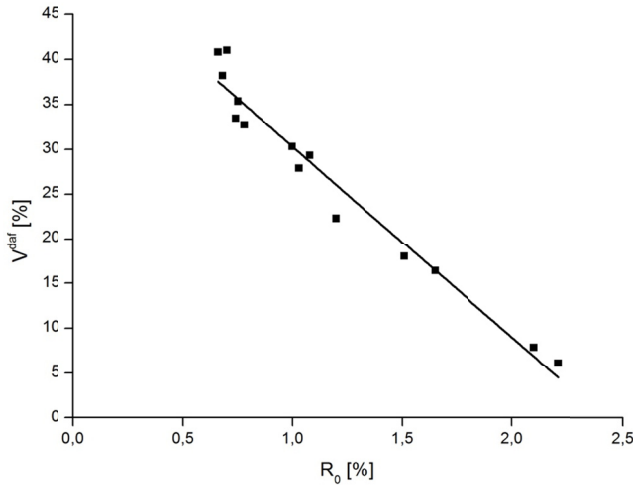


Fig. 4. Relation between volatile matter content  $V^{daf}$  and vitrinite effectance  $R_o$  (Poland)

Therefore, the rank of coal can be also expressed by the volatile matter. A similar result of linear equation (3) regards also Australian coals (Laxminarayana & Crosdale, 1999) in the scope of  $0,6 < R_o < 2,0\%$ .

The content of ash ( $A_o$ ) in coals evidences the amount of the mineral matter in this coal. Fig. 5 presents the relation between the Langmuire volume  $V_L$  and the ash content  $A_o$ .

In the examined samples of coals the volume of mineral matters was on the level of about 5%, which may be both of more or less significance.

The relation between methane sorption capacity and the rank of coal, which is material for the present paper, is presented on the Fig. 6.

Regardless of the volume of vitrinite and inertinite (bright coal or dull coal) for the low rank (HV) the Langmuire volume  $V_L$  decreases with increase of  $R_o$  and passes through minimum for middle rank coal (MV),  $R_o - 1,20$ . Subsequently,  $V_L$  rises with the increase of  $R_o$  for the higher rank coal to semianthracite and anthracite (while sorption capacity semianthracite is lower than the expected – perhaps due to the low content of vitrinite).

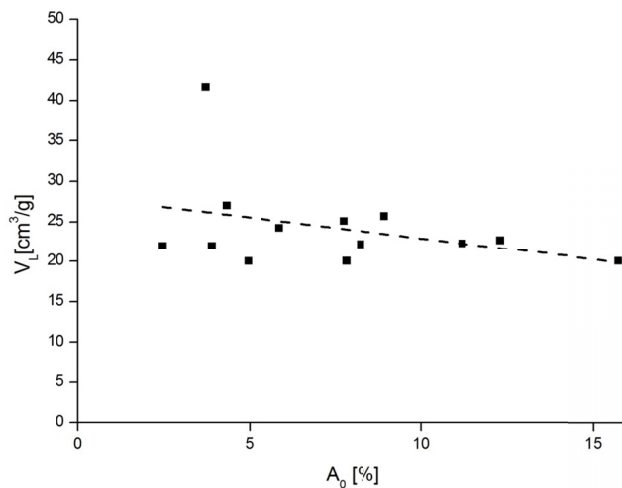


Fig. 5. Relation between sorption capacity  $V_L$  and ash content  $A_o$  in coals (Poland)

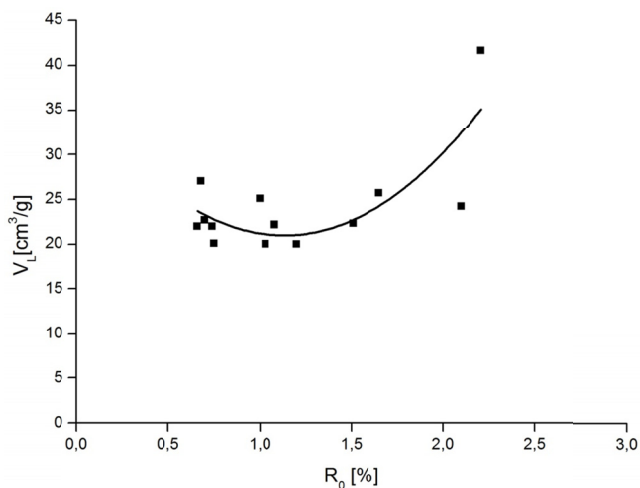


Fig. 6. Rank effects on methane sorption capacity of coals (Poland)

This relation can be presented with assistance of second – order polynomial trends:

$$V_L = 3,48 R_o^2 - 0,59 R_o + 21,02$$

as nonlinear approximation with weak correlation,  $r^2 = 0,60$ .

By comparison one can observe here the similarity to the Indian coals (for low rank) and Australian coal (for middle and high rank coal). There is however no broad source basis for the comparative purposes due to a small number of Polish coal samples that were possible to be gathered for the analysis.

## 4. Summary

With respect to the relation of rank of Polish coal and the methane sorption capacity one should rather focus on the trend, not on a particular strict relationship. The opinion that with the increase of the rank of coal rises also the sorption capacity is untrue. This can be only the case for the high rank coal along with anthracites. Methane sorption capacity, expressed by Langmuire volume decreases for low rank (HV) and reaches minimum for values of  $R_o$  corresponding with the medium rank (MV). In the scopes of changes  $R_o$ : 0,6-2,2% and the sorption capacity 15-42 cm<sup>3</sup>/g the correlation between methane sorption capacity and rank of coal is weak. While for low rank (HV) a similarity to Indian coals was observed as well as for the high rank and middle rank a partial similarity to Australian coals.

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Received: 06 September 2013