

# Swelling Measurements of a Low Rank Coal in Supercritical CO<sub>2</sub>

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## ABSTRACT

Coal swelling in the presence of water as well as CO<sub>2</sub> is a well-known phenomenon, and these may affect the permeability of coal. Quantifying swelling effects is becoming an important issue to verify the suitability of particular coal seams for CO<sub>2</sub>-enhanced coal bed methane recovery projects. In this report, coal swelling experiments using a visualization method in the CO<sub>2</sub> supercritical conditions were conducted on crushed coal samples. The measurement apparatus was designed specifically for the present swelling experiment using a visualization method. Crushed coal samples were used instead of block coal samples to shorten equilibrium time and to solve the problem of limited availability of core coal samples. Dry and wet coal samples were used in the experiments because there is relatively limited information about how the swelling of coal by CO<sub>2</sub> is affected by water saturation. Moreover, some coal seams are saturated with water in initial reservoir conditions. The maximum volumetric swelling was around 3% at 10 MPa for dry samples and almost half that at the same pressure for wet samples. The wet samples showed lower volumetric swelling than dry ones because the wet coal samples were already swollen by water. Experimental results obtained for swelling were comparable with other reports. Our visualization method using crushed samples has advantages in terms of sample preparation and experimental execution compared with the other methods used to measure coal swelling using block samples.

**Keywords:** Coal Swelling Experiments; Visualization Method; CO<sub>2</sub>-Enhanced Coal Bed Methane Recovery; Low Rank Coal

## 1. Introduction

CO<sub>2</sub> capture and storage (CCS) is a method expected to reduce emission of CO<sub>2</sub> into the atmosphere. In fact, geological sequestration is most likely to be the only option that will allow CO<sub>2</sub> storage in large enough quantities over long geological periods of time. One of the geological sequestration options is injection of CO<sub>2</sub> into coal seams. This option is considered to be a safe and effective method for permanently storing CO<sub>2</sub> with the added value of enhanced coal bed methane recovery (CO<sub>2</sub>-ECBMR). It has been suggested that the CO<sub>2</sub> sorption capacity of coal seams is typically between 2 to 10 times higher than that for CH<sub>4</sub> depending on coal rank [1-3].

CO<sub>2</sub> injection into a coal seam increases the rate of CH<sub>4</sub> production considerably [4]. However, CO<sub>2</sub> is also known as a swelling agent for coal. Coal swelling in the presence of water or CO<sub>2</sub> is a well-known phenomenon, and may affect the permeability of coal. Because the

permeability of a coal is perhaps the most important factor controlling the economic viability of CO<sub>2</sub>-ECBMR, thus quantifying the effect of swelling on permeability has become an important issue.

A number of studies investigating CO<sub>2</sub>-induced swelling have been published. The maximum volumetric swelling has been reported as 1% - 5% depending on coal rank [5-7]. Reduction of CO<sub>2</sub> injectivity has been observed in some CO<sub>2</sub>-ECBMR field trials. One of these projects was carried at Yubari, Hokkaido, Japan. The Yubari pilot project for CO<sub>2</sub>-ECBMR identified the problem of low injectivity of CO<sub>2</sub> caused by a reduction in permeability induced by coal swelling [3]. The decay ratios of daily CO<sub>2</sub> injection rate measured in the Yubari pilot test are shown in **Figure 1**.

A field experiment of CO<sub>2</sub>-ECBMR carried out in the upper Silesian basin of Poland is another example of a pilot project that observed decreasing injectivity caused by swelling. Evaluation of wellhead pressure and temperature data during the pressure buildup and fall-off of

the intermittent injection periods, considering phase behavior and density changes of the CO<sub>2</sub>, showed that the buildup time decreases, and the fall-off time increases over time. In addition, the steep decrease at the beginning of the curve during start-up (with water down the hole) disappeared in November and December 2004. Both observations indicate that permeability reduced over time [8]. The reduced injectivity was presumably a result of swelling of the coal after contact with CO<sub>2</sub>.

The only successful CO<sub>2</sub>-ECBMR project so far was run between 1995 and 2001 at the Allison Unit in the San Juan Basin in New Mexico, USA, but no monitoring project was associated with this pilot [10]. It has been recognized from the pilot test results that reduction of permeability induced by coal swelling is one of the main technical issues that needs to be resolved to put economical, large-scale CO<sub>2</sub>-ECBMR into practice worldwide.

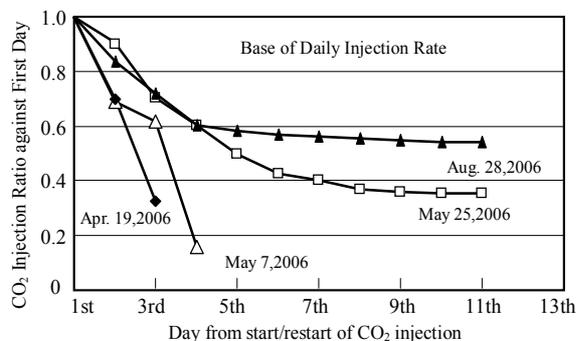
In the case of CO<sub>2</sub>-ECBMR, adsorption of CO<sub>2</sub> gas, which has greater sorption capacity than CH<sub>4</sub>, would cause coal matrix swelling and thus, in contrast to CH<sub>4</sub> desorption, potentially reduce the cleat permeability of a coal seam. Therefore, it is important to quantify the swelling of coal induced by CO<sub>2</sub> adsorption.

Most swelling experiments have been conducted using core/block coal samples [6,7,11-13], so the equilibrium time was long depending on core block size. Indonesia has many coal resources as well as coal bed methane, so shows potential for CO<sub>2</sub>-ECBMR sites. However, there is a lack of research of coal behavior after introducing CO<sub>2</sub>; in particular, coal swelling caused by CO<sub>2</sub> adsorption. Thus, our objective was to investigate the swelling characteristics of Indonesia low rank coal (LRC) up to 10 MPa. Crushed coal samples that were treated as block samples have been used in the experiments to allow faster equilibrium time and to solve the problem of limited availability of core samples from deep underground.

## 2. Samples and Methods

### 2.1. Samples

Coal core samples taken from coal seams deep under-



**Figure 1. Decay ratio of daily CO<sub>2</sub> injection rate monitored at Yubari ECBM-pilot test [9].**

ground are very limited. It's costly and technically difficult to obtain representative core samples from deep underground. Furthermore, coal that is anisotropic causes other difficulties with regard to analysis and reproducibility. Therefore, to overcome these difficulties, crushed coal samples were used instead of core samples.

Three coal samples of Indonesia LRC were used in the swelling experiments. Their properties are summarized in **Table 1**.

### 2.2. Apparatus

A visualization method was used to observe the coal swelling evaluated by surface movement of a column of coal particles. **Figure 2** shows a schematic diagram of the measurement apparatus. The measurement apparatus was designed specifically for this experiment, and consisted of a high-pressure cell with two glass windows, a hot water bath with a circulation pump, an electric pressure gauge with a precision of 0.01 MPa (GE Druck Ltd, DPI 282), thermocouples, an illumination source and line valves. The cell temperature was kept at 48°C by circulation of hot water through a circular jacket covering the cell, and the apparatus was positioned in a thermally insulated box.

A digital camera was used to take photographs continuously during the experiment. All measurement data from the camera, pressure transducer and thermocouple were logged into the data acquisition system continuously.

### 2.3. Procedures

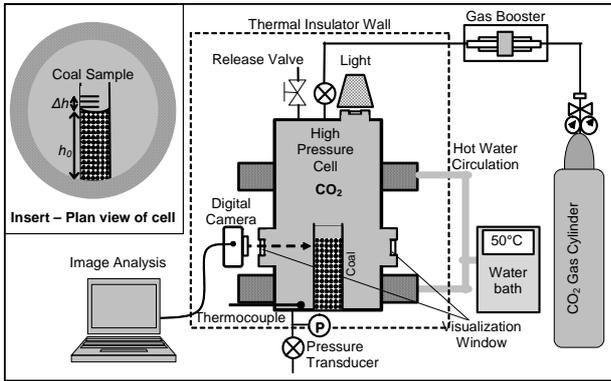
The procedure for coal swelling experiments was as follows:

#### Step 1. Sample preparation

Measurements were performed in a glass cylinder 7.35 mm in diameter and 50 mm in length with a high precision scale. Two cylinders were placed in the pressure cell

**Table 1. Detail coal properties for CO<sub>2</sub> swelling experiments.**

	Adb	SK-01	SK-02	SK-03
Moisture, % wt	10.68	11.63	7.53	
Ash, % wt	4.19	2.44	10.31	
Volatile matter, % wt	40.34	41.72	45.17	
C, % wt	55.18	57.09	53.76	
H, % wt	4.22	4.49	4.62	
N, % wt	0.72	0.79	0.73	
Sulfur, % wt	0.12	0.19	0.25	
R <sub>0</sub> , %	0.47	0.47	0.45	



**Figure 2. Schematic diagram of coal swelling measurement system using visualization method.**

for each measurement. Each cylinder was filled with dry or water-saturated crushed coal particles.

The crushed coal samples were sieved into grain sizes of 200 - 250 μm. For dry samples, crushed coal was put into an incubator at 60°C for 48 h under vacuum. After complete drying, approximately 1 g of sample was placed into a glass cylinder and centrifuged at 3000 rpm for 10 min. These conditions were equivalent to a relative centrifugal force (RCF) of 554.4 as defined by Equations (1) and (2). The results shown in **Figure 3** confirm that there was no difference in the coal column after 3000 rpm.

$$RCF = \frac{r\omega^2}{g}, \quad (1)$$

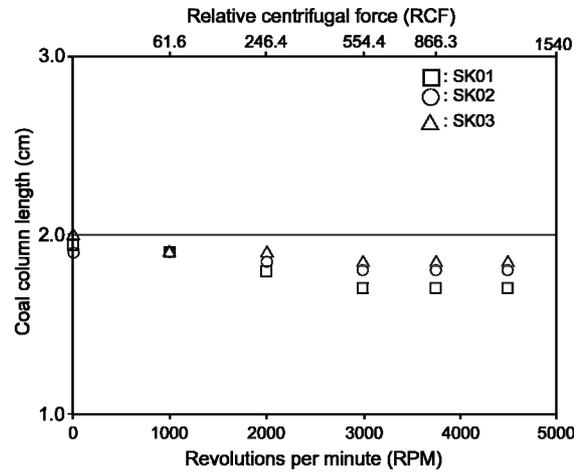
$$RCF = 1.12 \times 10^{-5} r_{cm} N_{RPM}^2, \quad (2)$$

where  $g$  is the gravitational acceleration of earth,  $r$  is the rotational radius,  $\omega$  is the angular velocity in radians per unit time,  $r_{cm}$  is the rotational radius measured in centimeters (cm) and  $N_{RPM}$  is rotational speed measured in rpm.

For saturated samples, approximately 1 g of crushed sample and 1 mL of pure water were filled in the cylinder and then it was centrifuged at 3000 rpm for 10 min to introduce air and ensure close packing of the sample. Centrifugation was used to allow accurate measurement of initial column height and ensure that the sample was fully saturated with water. Both samples were placed in the pressure cell for 24 h before CO<sub>2</sub> injection.

#### Step 2. Swelling Measurement

The cell temperature was set at 48°C and the CO<sub>2</sub> injection pressure was increased in a series of stepwise up to 10 MPa. The system was maintained at each pressure step at least for 48 h and even more. The equilibrium time required for swelling of coal induced by CO<sub>2</sub> adsorption depends on the size of the sample. A bigger coal sample will take longer to reach equilibrium and *vice versa*. In our experiments, most of the swelling occurred



**Figure 3. Column length of change as a function of relative centrifugal force in 10 minutes.**

within 24 h for each pressure step. After measurement at the maximum step, the pressure was decreased to atmospheric conditions to allow comparison of final and initial heights of the coal column.

The upward movement of the column surface was measured against pressure and termed change of column length ( $\alpha$ ). The proportion length of change,  $\alpha$  (%), and volumetric swelling,  $\beta$  (%), are defined by Equations (3) and (4).

$$\alpha = \frac{\Delta h}{h_0 [1 - K(p - p_0)]} \times 100\% \quad (3)$$

$$\beta = \frac{(\pi r^2 \Delta h)}{V_0 [1 - K(p - p_0)]} \times 100\% \quad (4)$$

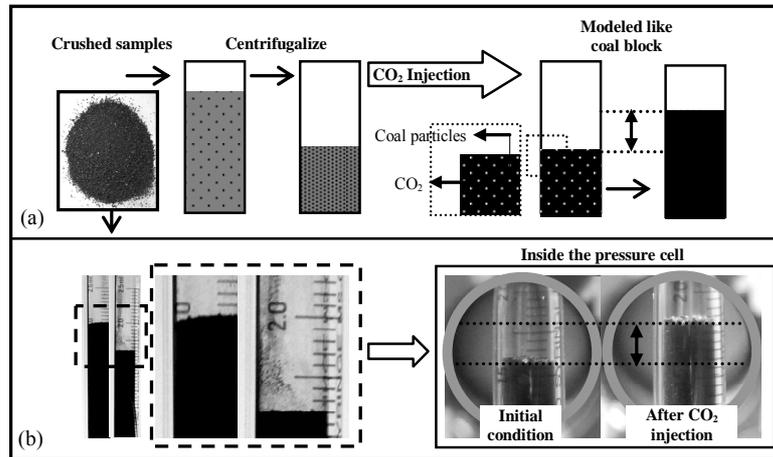
where  $\Delta h$  is increase length caused by CO<sub>2</sub>-induced swelling,  $K$  is compressibility factor (MPa<sup>-1</sup>),  $V_0$  is initial volume (cm<sup>3</sup>),  $r$  is radius of cylinder (cm),  $p$  is pressure (MPa) and  $p_0$  is initial pressure (MPa).

## 3. Results and Discussion

### 3.1. Swelling Experiments

Crushed samples with particle sizes of 200 - 250 μm were used to avoid particle-packing problems. The samples were centrifuged to ensure that the particles packed into interstitial spaces and were distributed evenly. In addition, Busch *et al.* [14] reported that sorption rate did not change significantly with increasing grain size >100 μm, thereby increasing ash content during grinding can be eliminated.

Swelling was represented by upward surface movement of the coal packing column, as shown in **Figure 4**. As shown in Equation (5), the total initial volume ( $\bar{V}_0$ ) is equal to the volume of each particle ( $v_i$ ) and CO<sub>2</sub> gas filling the interstitial space. Because swelling of coal



**Figure 4.** (a) Model showed the experimental procedure for crushed samples which was modeled like coal block; (b) Picture taken during experiment showed different column height after put in the centrifuge (left) and inside the pressure cell before and after CO<sub>2</sub> injection (right).

samples is proportional to adsorbed CO<sub>2</sub> gas molecules and CO<sub>2</sub> mass was kept constant at each pressure step, it was considered that the total initial volume was equivalent to the total volume of each particle. Moreover, the swelling ratio was the ratio of the total volume of the particles at a certain pressure ( $P_n$ ) to the initial volume of the particles, as given in Equation (6).

$$\bar{V}_0 = \sum_{i=1}^n v_{i0} + \bar{V}_{CO_2}, \quad (5)$$

$$\beta = \frac{\sum_{i=1}^n v_i P_n}{\sum_{i=1}^n v_{i0}} = \frac{\bar{V} P_n}{\bar{V}_0}. \quad (6)$$

Because upward surface movement of the coal column was assumed to be the total expanded volume for each particle, consequently, crushed samples were modeled like coal block. A diagram illustrating this model is shown in **Figure 4**.

Most of the swelling in the samples occurred within 24 h of exposure to CO<sub>2</sub>; after that, swelling increased slightly over a considerable length of time, as depicted in **Figure 5**. The length of both dry and wet sample columns increased with increasing CO<sub>2</sub> pressure. A rapid increase in length was observed in the low pressure range ( $\leq 6$  MPa), and the maximum length was reached between 8 and 10 MPa.

The results of volumetric swelling for three coal samples as a function of pressure and gas density are plotted in **Figure 6**. Previous studies observed that CO<sub>2</sub>-induced coal swelling fitted the Langmuir model [11,13]. The Langmuir model has been widely used and is used as a standard by many industries. However, it gave a poor fit in the pressure range above 6 MPa [6]. As an alternative, Day *et al.* [6] suggested that the Dubinin-Radushkevich

(D-R) adsorption model can give a better fit. The D-R model uses gas density instead of pressure according to Equation (7) [7]:

$$\varepsilon = \varepsilon_{\max} e^{-D[\ln(\rho_a/\rho_g)]^2}, \quad (7)$$

where  $\varepsilon_{\max}$  is the maximum volumetric swelling of the coal,  $\rho_g$  is the density of the gas at the temperature and pressure,  $\rho_a$  is the density of the condensed phase on the coal surface, and  $D$  is a constant that is related to the enthalpy of adsorption.  $\rho_a$  for CO<sub>2</sub> is 1028 kg m<sup>-3</sup> and both  $D$  and  $\varepsilon_{\max}$  are considered empirical curve-fitting parameters. The gas density ( $\rho_g$ ) at each pressure step was calculated using the PROPHAT program. **Figure 6** shows a plot of the D-R model. A summary of the results for both the D-R and Langmuir models is presented in **Table 2**.

Based on these results, the maximum volumetric swelling ratio measured is very similar to ones reported by [5,6,15-17], who used blocks and crushed coal samples. The maximum swelling from [6] with Ro 0.89% was ~2.6% at around 8 MPa and 40°C. Moreover, the maximum swelling of lignite samples reported by [5] was around 4.18% at 1.5 MPa.

Day *et al.* [6] used block coal with dimensions of 30 × 10 × 10 mm, while [5] used pencil-shaped samples that were 10 mm in length and 4 mm in diameter. Such samples are difficult to prepare because LRC tends to break when cut into small pieces using a rock cutter with water as a lubricant. Therefore, swelling experiments using crushed coal are advantageous in terms of sample preparation and experimental apparatus compared with ones using blocks.

CO<sub>2</sub> adsorption equilibrium is reached faster for coal of smaller grain size [18]. For the crushed samples used in this study, swelling equilibrium time, which correlates

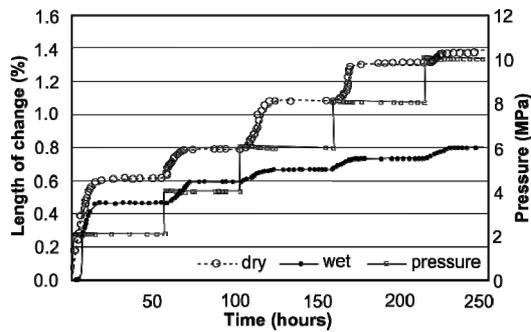


Figure 5. Length of change as a function of time and pressure.

Table 2. D-R and Langmuir's parameters for three coal samples.

	SK-01		SK-02		SK-03	
	Dry	Wet	Dry	Wet	Dry	Wet
<i>D-R's Model</i>						
$\epsilon_{max}$ (%)	2.81	1.45	3.33	1.57	3.04	1.51
D	0.0859	0.0827	0.0761	0.0910	0.0827	0.0953
<i>Langmuir's Model</i>						
$\epsilon_L$ (%)	3.53	1.96	4.53	2.34	3.95	2.17
$P_L$ (MPa)	4.53	5.07	4.82	6.58	4.63	6.46

strongly with adsorption equilibrium time, was shorter than that for blocks. Reucroft and Sethuraman [5] employed an exposure time of 200 h in their study. In our experiments, maximum swelling occurred within 12 - 24 h for each pressure step. This was another benefit of using crushed samples.

One of the remaining concerns of using crushed samples was whether the samples return to their original position or not. Coal swelling experiments using block samples showed that all of the coal samples return to their original dimensions after removal from CO<sub>2</sub> [6,7]. In the present study using crushed coal, we observed the same tendency. Unlike some liquid solvents, exposure to gas does not impart enough molecular mobility to allow relaxation of the structure of coal. Thus, the coal remains elastic, and the swelling process is reversible [19].

Otake and Suuberg [17] reported that the packing density of particles could affect swelling measurements and introduce significant artifacts. To solve this problem, we used small particle sizes (200 - 250 μm) and sample columns were centrifuged to ensure that the particles packed into the interstitial spaces and were distributed evenly. Our results suggest that the processes of swelling are similar in crushed and block coal samples.

The experimental results shown in Figure 6 indicate that volumetric expansion of wet coal is lower than that of dry samples. Swelling of wet coal decreased by

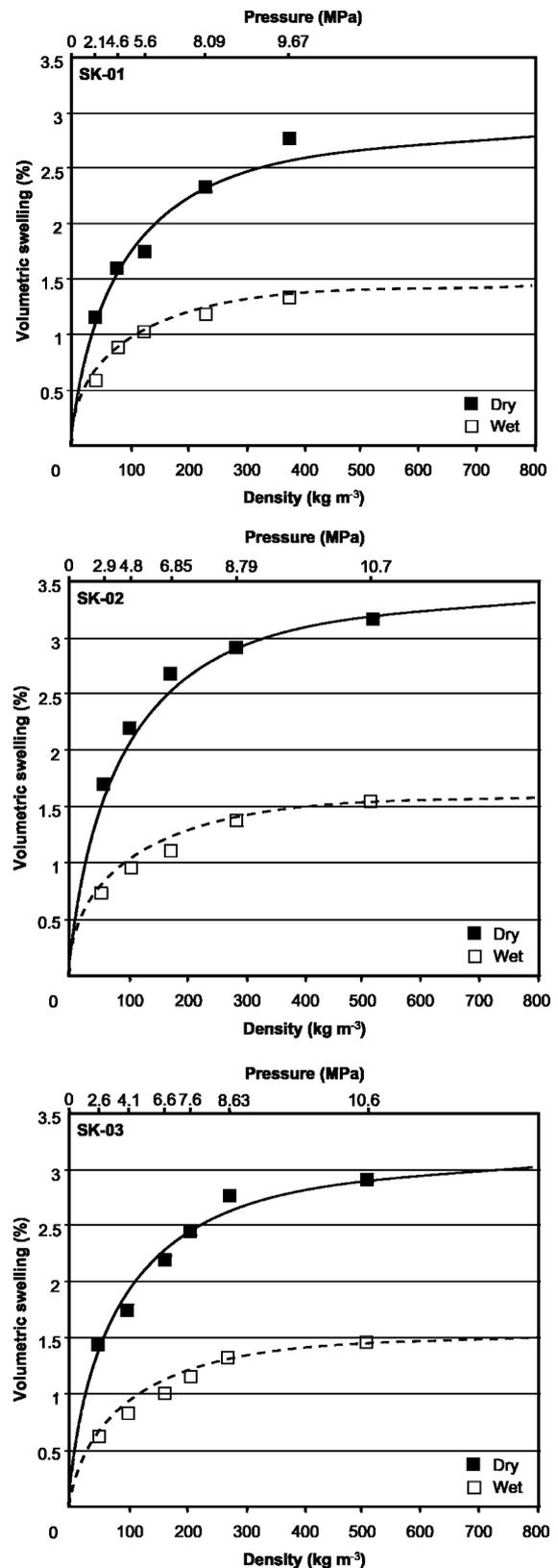


Figure 6. Volumetric expansion of the three coal samples at 48' under exposure to CO<sub>2</sub>, the line plot represents the modified D-R model results.

around 50% compared with that of dry coal. In accordance with [7], the effect of moisture on the amount of swelling is higher in LRC samples.

When considering the wet samples, one should remember that they are actually already swollen because of the presence of moisture. It has been suggested that the mechanism of swelling is the same in both water and gas [20]. Therefore, the total volumetric swelling of wet coal is the sum of swelling induced by both water and gas.

Considering the crushed coal samples used in this study, instead of swelling both perpendicular and parallel to the bedding plane, experimental results only showed “iso-swelling”. The term iso-swelling was used to show that volumetric swelling was calculated without considering the difference between linear swelling parallel and perpendicular to the bedding plane.

In fact, based on some previous studies, linear swelling is significantly different between perpendicular to bedding plane and parallel to the bedding plane. Swelling varies by 30% - 400% depending on the coal properties, and perpendicular swelling is always greater than parallel [6,11,12,21].

To investigate this phenomenon, some equations have been used to predict linear swelling parallel and perpendicular to the bedding plane. In swelling experiments using a strain gauge where volumetric swelling was calculated as twice the strain parallel to the bedding plus the strain perpendicular to the bedding [11,12], the following equation was used:

$$\frac{\Delta V}{V} = \epsilon_{vs} = \epsilon_{ls1} + \epsilon_{ls2} + \epsilon_{ls3}, \tag{8}$$

where  $\epsilon_{vs}$  is volumetric swelling, and  $\epsilon_{ls1}$ ,  $\epsilon_{ls2}$ , and  $\epsilon_{ls3}$  are the principal linear strain. The volumetric swelling can be determined by adding the linear strain,

$$\epsilon_{vs} = 2\epsilon_{lpar} + \epsilon_{lper}, \tag{9}$$

where  $\epsilon_{lpar}$  and  $\epsilon_{lper}$  are strains parallel and perpendicular to the bedding plane, respectively. To formulate Equation (9), it was assumed that the strain perpendicular and parallel to the bedding plane could be predicted using Equation (10),

$$\epsilon_{par} = \frac{\epsilon_{par1} + \epsilon_{par2}}{2}. \tag{10}$$

### 3.2. Dependence of CO<sub>2</sub> Swelling on Coal Properties

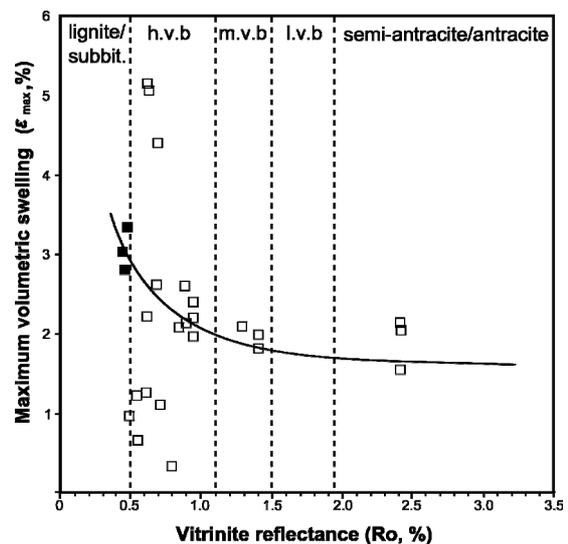
Because the samples used in these experiments have similar coal rank, the difference between swelling results was too small to allow meaningful comparison. Meanwhile, some authors [5,7,11,19] concluded that swelling decreased with increasing coal rank. **Figure 7** shows the maximum coal swelling ( $\epsilon_{max}$ ) as a function of coal rank

from our results and those reported [6,7,11-13,21,22]. There is a dependence between coal rank and swelling; higher swelling corresponds to lower coal rank.

Some considerations have to be made for the LRC where the data are scattered. Coal swelling by CO<sub>2</sub> injection is strongly controlled by CO<sub>2</sub> adsorption volume. Based on [23], the dependence of CO<sub>2</sub> sorption on coal properties is controlled not only by individual factors but also the combination by several factors; e.g., coal composition, TOC, ash content, dull and bright coal as well as coal rank. Meanwhile, gas sorption tends to increase with increasing rank, although some exceptions were found in [18]. Beulah-Zap lignite, which was the lowest rank of all samples, showed the highest sorption capacity.

Coal of higher rank has been subjected to a more severe stress history than LRC, so it in general exhibits more structural deformation and is thereby more difficult to swell. Larsen *et al.* [19] explained that higher rank coal rearranges in the opposite direction to LRC to form a less-associated structure. Thus, coal properties should be well characterized to evaluate coal swelling because swelling does not only rely on rank.

Within the sample set studied, a general tendency was observed for  $D$  to increase for wet samples, although sample SK-01 was an exception. Increasing  $D$  is related to the enthalpy of adsorption based on Wood’s equation (Equation (11)), whereas  $E$  of CO<sub>2</sub> is reduced in the presence of moisture [7,24]. This apparent reduction in enthalpy may be caused by higher energy adsorption sites being occupied by water molecules in the wet samples (by hydrogen bonding), thus restricting gas sorption to sites of lower energy [7].



**Figure 7.** The maximum coal volumetric swelling ( $\epsilon_{max}$ ) as a function of coal rank (black cube represent swelling data from present experiment results).

$$D = \left[ \frac{RT}{\beta E} \right]^2 \quad (11)$$

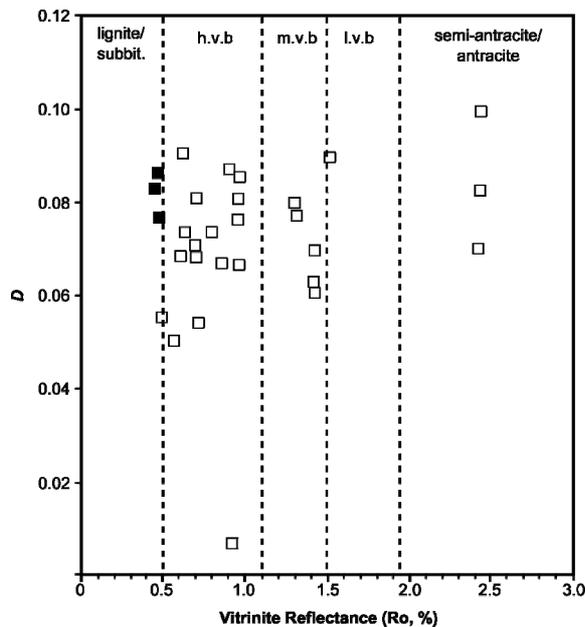
where  $R$  is the universal gas content,  $T$  is the temperature and  $\beta$  is a scaling factor to account for the different affinities of sorbates for the sorbent material.

In the case of published data [7,24], high rank coal with vitrinite reflectance ( $R_0$ ) more than 0.76 [24] and 0.62 to 1.40 for [7] was used. However, the lowest rank sample in [7] showed decreasing  $D$  value, which they attributed to moisture preferentially occupying polar sites on the coal surface. In dry samples, it appears that  $\text{CO}_2$  and  $\text{CH}_4$  can readily adsorb on these polar sites, but when water is present, they become unavailable to other sorbates. Thus, in moist LRCs with a high proportion of polar sites, much of the internal surface of the coal is already covered by water molecules. As a result, there is relatively little surface available for gas adsorption and therefore little swelling.

Based on our data using LRC, both decreasing and increasing  $D$  values were observed. **Figure 8** shows  $D$  value as a function of coal rank based on current experiments and reported data [6,7,11-13,21,22], which indicates that  $D$  value does not have a strong dependence on coal rank. Thus, because  $\text{CO}_2$  swelling induced by  $\text{CO}_2$  adsorption correlates strongly with the amount of  $\text{CO}_2$  adsorbed, care should be taken when considering the factor(s) causing coal swelling.

#### 4. Conclusions

In this study, a visualization method was used to measure



**Figure 8.**  $D$  value as a function of coal rank (black cube represent swelling data from present experiment results).

swelling of crushed coal samples in a pressure cell with  $\text{CO}_2$  pressure of 0.1 to 10 MPa and temperature of  $48^\circ\text{C}$ . Crushed coal samples provide benefits in terms of sample preparation and experimental ease compared with block coal samples. The other benefit of this method is the relatively short equilibrium time needed to reach maximum swelling.

Coal swelling was represented by upward surface movement of columns filled with crushed coal particles. The total expanded volume of coal particles was modeled like the swelling volume of coal block. Present swelling results were comparable with published data.

The maximum swelling volume ratios for the three coal samples achieved by  $\text{CO}_2$  adsorption were around 3% at 10 MPa of  $\text{CO}_2$ . Wet coal samples showed lower volumetric expansion than dry ones because moisture considerably reduced the degree of gas-induced swelling. Although wet coal swells less than dry samples, wet samples are in fact already partially swelled because of the presence of water. In addition to calculating swelling perpendicular and parallel to the bedding plane instead of volumetric swelling, the equation developed in this study can be used.

#### 5. Acknowledgements

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