Research activities in University of Alberta based on International Training Program (ITP)

(*30 June – 20 Sept 2009)*

Phung Quoc Huy, Ph.D Student
Resources Production Engineering, Dept. of Earth Resources Engineering
Faculty of Engineering, Kyushu University

Theme: **Visualization Study on Swelling of Coal Matrix in High Pressure CO_2**

Based on International Training Program (ITP) supported by Japan Society for the Promotion of Science (JSPS), I had carried out research activities around 2.5 months (30th Jun ~ 20th September 2009) in University of Alberta, Edmonton city, Canada.

I. INTRODUCTION OF EDMONTON

Edmonton is the capital of the Canadian province of Alberta. The city is located on the North Saskatchewan River in the central region of the province, an area with some of the most fertile farmland on the prairies. It is the second largest city in Alberta after Calgary, and is the hub of Canada's sixth-largest census metropolitan area.

In the Canada 2006 Census, the city had a population of 730,372, and its census metropolitan area had a population of 1,034,945, making it the northernmost North American city with a metropolitan population over one million. The 2009 civic census showed a population of 782,439.

At 684 km^2, the City of Edmonton covers an area larger than Chicago, Philadelphia, Toronto, or Montreal. Edmonton has one of the lowest population densities in North America, about 9.4% that of New York City. A resident of Edmonton is known as an *Edmontonian*.

Edmonton serves as the northern anchor of the Calgary-Edmonton Corridor (one of four regions that together comprise 50% of Canada's population) and is a staging point for large-scale oil sands projects occurring in northern Alberta and large-scale diamond mining operations in the Northwest Territories.

Edmonton is Canada's second most populous provincial capital (after Toronto) and is a cultural, governmental and educational centre. It plays host to a year-round slate of world-class festivals, earning it the title of "The Festival City." It is home to North America's largest mall, West Edmonton Mall (which was the world's largest mall for a 23 year period from 1981 until 2004.), and Fort Edmonton Park, Canada's largest living history museum. In 2004, Edmonton celebrated the centennial of its incorporation as a city.

Map of Edmonton city

II. LIVING IN EDMONTON

The applicant has been to Edmonton City (Alberta, Canada) where University of Alberta is located. In the first week, applicant stays in Hi-Edmonton Hostel and looking for the rental room in two months. Then, applicant moves to the 5 bed rooms house which shared with other students. From the second week, applicant proceed the experiment at University of Alberta. During the staying period in Edmonton, applicant joint the field trip to Genesee coal mine, EPCOR power plant, Japan Canada Oil Company (JACOS) and Syncrude company.

5 BR house

Mining & Petroleum Dept.

Syncrude company visit

Genesee coalmine visit

Jacos company visit

EPCOR power plant visit

III. RESEARCH IN UNIVERSITY OF ALBERTA

INTRODUCTION

Many projects of CO_2 sequestration into un-mineable or deep coal seams have been started as a method for reducing greenhouse gas emissions to the atmosphere. However, some of projects had a problem of reducing permeability after CO_2 injection. Coal is well known to swell with CO_2, and this is likely to affect gas transport through coal seams. It has been suggested that in situ coal swelling may close the seam cleat system, thus it make reducing permeability (Larsen 2004). As well as effects which occur within the coal seam, coal swelling also may affect to the accuracy of estimating sorption capacity because the volume of the coal may change during the measurement of the adsorption isotherms. Authors observed the volume change before and after CO_2 gas adsorption as 1.74 to 1.93 % with Vietnamese coal samples (Huy et al., 2006). Coal is expected to be swelled by two mechanisms depending on the fluid (Sevket Durucan, 2009). The first mechanism is the sorption of a liquid-like layer on pore walls creates a pressure gradient that is large enough to cause measurable structural changes in the solid. At elevated pressures, CO_2 is expected to behave as a high density liquid, especially inside microporous materials. The force exerted by the adsorbed layer will part the macromolecular clusters. The second one is the imbibition of fluid into the solid structure causes expansion of the porous matrix. In this case, interaction of the fluid with the solid through hydrogen bonding, electron transfer, etc. can result in relaxation of the molecular layers in the coal. It is suggested that the expansion of coals in high-pressure CO_2 environments is attributable to a combination of these mechanisms. However, it has been difficult to account properly for the amount of expansion due to the extent of chemical versus physical interaction of the fluid with the solid.

As described above, swelling of coal matrix depends on many parameters such as coal characteristics (ash, moisture, volatile content); pore structure; type of fluid and adsorption capacity (see figure 1).

The aim of this study is to clarify the swelling phenomenon caused by CO_2. In this study, the visualization method was applied to measure the volume expansion by providing CO_2 in gas, liquid and supercritical states. Comparisons of the volume expansion with other methods were discussed.

SAMPLE PREPARATIONS

Coal samples for visualization study were prepared from coal blocks or small coal lump (see figure 2). To fit the dimensions of the visualization cell, size of coal samples was shaped to be less than 8 mm in diameter and 6 mm in thickness.

Coal samples used for present measurements are listed in Table 1. Four kinds of coal (Japanese, Australian ,Vietnames, Chinese were used.

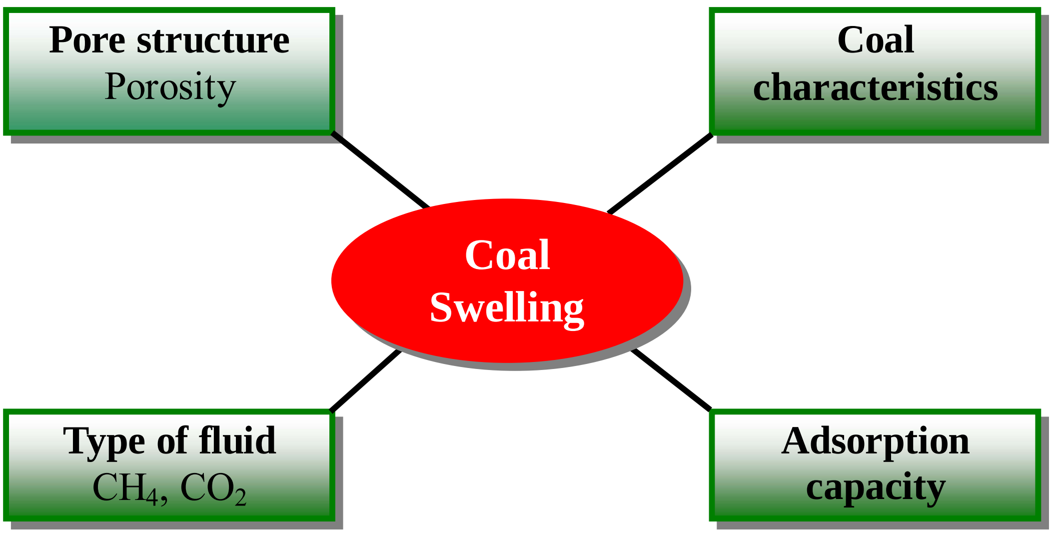


Figure 1: Parameters affecting coal swelling

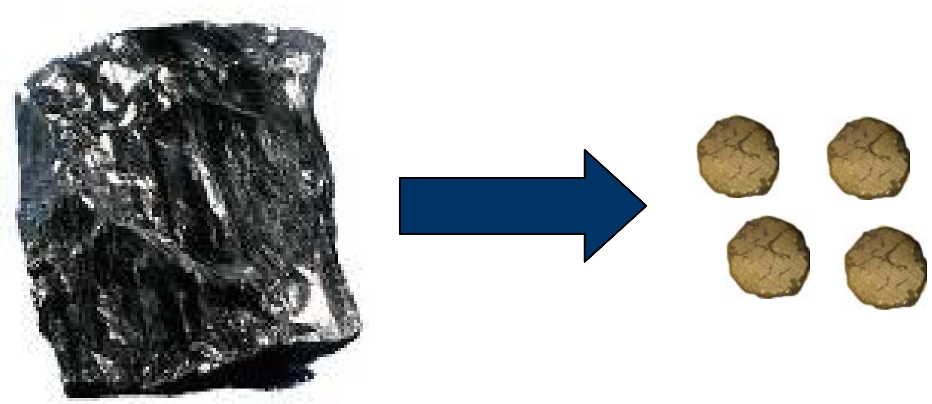


Figure 2: Preparation of coal samples

Table 1: Coal sample characteristics

Name of sample	Location	Diameter mm	Thickness mm	Weight g	Ash content %	Moisture content %	Volatile matter %	Fixed carbon %
JP-3	Japan	7.0	3.1	0.141	0.4	25.2	25.3	49.1
AUS-1 1	Australia	6.5	3.1	0.114	4.2	9.9	36.0	50.2
MK-1	Vietnam	7.0	2.9	0.118	29.7	4.5	6.5	59.3
CH-1	China	6.8	2.8	0.115	15.2	5.3	25.3	54.2
JP-2	Japan	6.7	2.9	0.152	1.2	6.3	26.9	65.6

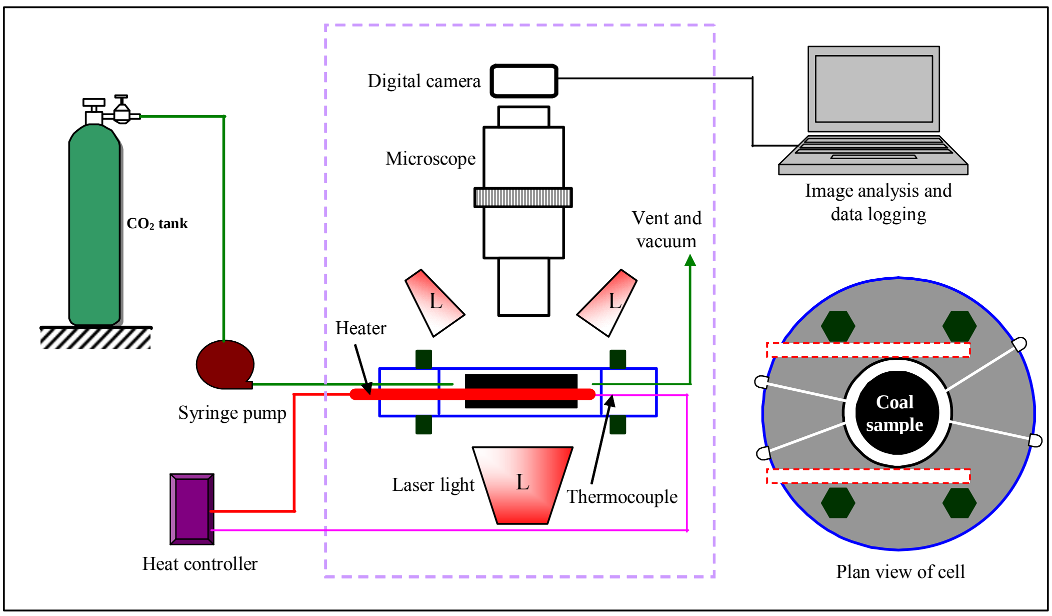


Figure 3: Diagram of experimental set up

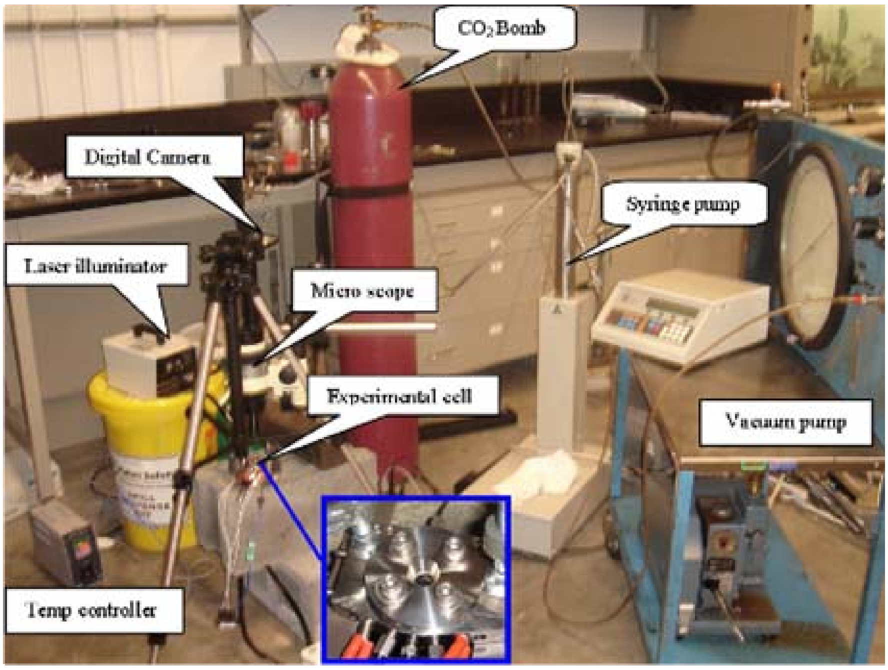


Figure 4: Photo of experimental apparatus

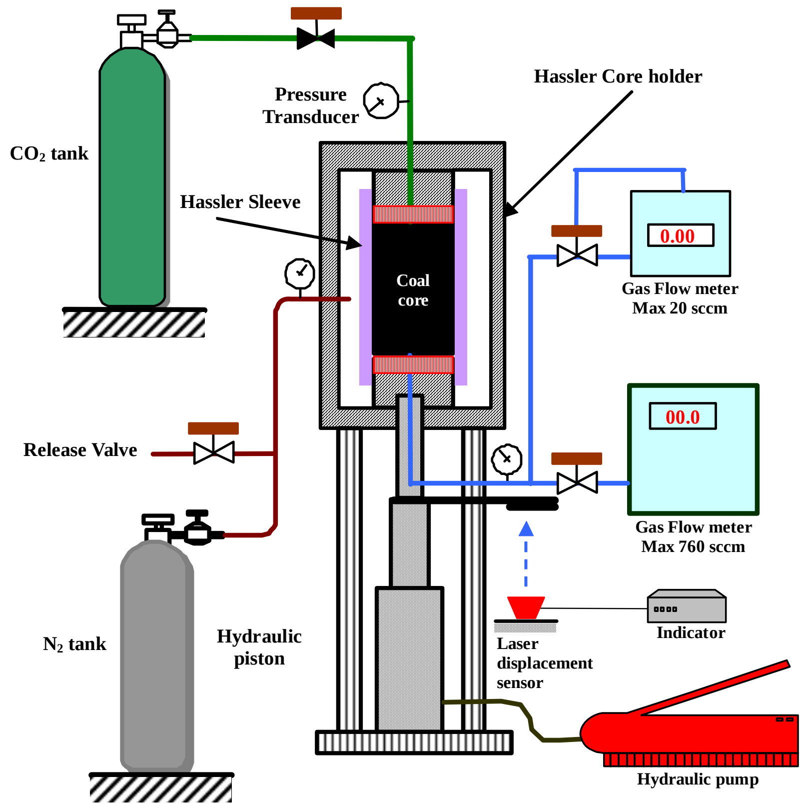


Figure 5: Experimental apparatus to measure core strain and permeability

Figure 6: Australian core sample (AU-14) for measuring core strain and permeability

APPARATUS AND PROCEDURE

Flow diagram and photo of present measurement apparatus for swelling are shown in Figures 3 and 4. It is comprised of a high pressure cell with transparent glass window through which the sample was observed directly using with microscope (Wild M75 Heerbrugg) or digital camera (Canon D50), laser lighting system, heater, sensors and CO_2 tank. The pressure cell dimension is 8mm in diameter and 6mm thick. The system can apply a wide range of pressures (from 0 MPa to 15 MPa) using a syringe pump (ISCO 500D). The heater system was used to maintain samples and CO_2 temperature for range of 20 to 60°C. The cell temperature was monitored by a thermocouple inserted in the cell.

Visualization measurement procedure

Step 1: Checking leakage for the whole system using leakage checking liquid.

Step 2: Place the coal sample into the pressure cell.

Step 3: Evacuate the cell and lines to a vacuum for 2 hours.

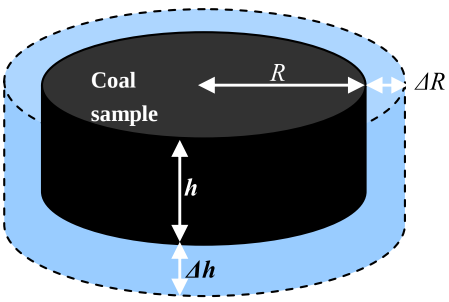
Step 4: Inject CO_2 into the pressure cell.

Step 5: Adjust the microscope and capture the image using the digital camera.

In this study, it was impossible to measure length change in the vertical axis. We therefore assumed the length expansion in the vertical and horizontal axes of the coal sample is the same value (see figure 7). This means that Δh is equal to ΔR. The volume expansion can be calculated using the following equation:

$$V + \Delta V = \pi (R + \Delta R)^2 (h + \Delta h) \quad (1)$$

$$\Delta V = \pi (R + \Delta R)^2 (h + \Delta h) - V \quad (2)$$



R: Radius of coal sample
ΔR: Expansion of radius
h: Thickness of sample
Δh: Expansion of thickness

Figure 7: Coal sample parameters

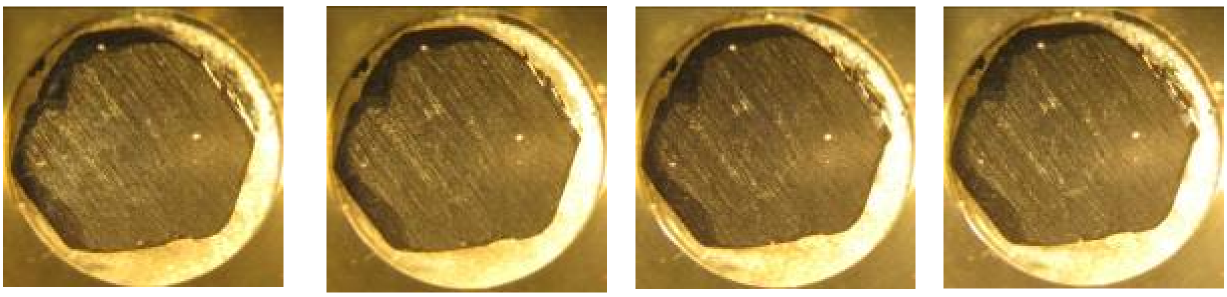
The expansion length of Australian coal core samples (AU-14) was measured simultaneously with permeability. The measurements apparatus shown in Fig. 6 and methodology have been reported by Huy et al. (2009) and Ichikawa et al. (2009). Thus, in this paper it is not described. The core is 38mm in diameter and 52.3 mm in length.

RESULTS AND DISCUSSION

Figure 8 shows the image of AUS-11 coal samples at certain time using microscope and digital camera. Because the size of the measurement cell was small (d = 8 mm), it is difficult to clarify differences between images. The sample cross-sectional expansion was recognized by using image analysis software.

Volume expansion versus cell pressure calculated using the visualization method is shown in Fig. 9. The results show that volume of almost coal samples increased from 0.1 to 1%. Volume of the JP-3 sample increased from 0.2% to 0.39 % with a cell pressure of 1 MPa and 5 MPa, respectively. Similarly, volume expansion of JP-2 was 0.12 % to 0.98 % with cell pressure range from 3 to 10 MPa. The Australian coal sample (AUS-11) has the smallest volume expansion from 0.031% to 0.153%, with a cell pressure range from 0.1 to 15 MPa. Figure 9 also shows that volume expansion increases with increasing cell pressure. This phenomenon can be explained by the adsorption process. In this case, volume expansion was caused by adsorption, with CO_2 adsorbed into the coal matrix to cause swelling. With high CO_2 pressure, CO_2 density changed very quickly at around the sub-critical and super critical point (P = 7.3 MPa, T = 304.1 K = 31.1 $^{\circ}$C). Coal swelling also occurred violently at the sub and supercritical CO_2 states.

Conversely, the volume of MK-1 and CH-1 coal samples reduced with increasing pressure. When injecting CO_2 into the sample cell both swelling and shrinkage occurred at the same time, causing volume increase and decrease, respectively. For the JP-2, JP-3 and AUS-11 coal samples, the swelling effect is much greater than the shrinkage effect so that volume expansion had a positive value.



P = 0 MPa | P=7.5 MPa | P = 10 MPa | P = 15MPa

Figure 8: Visual results of coal analyses

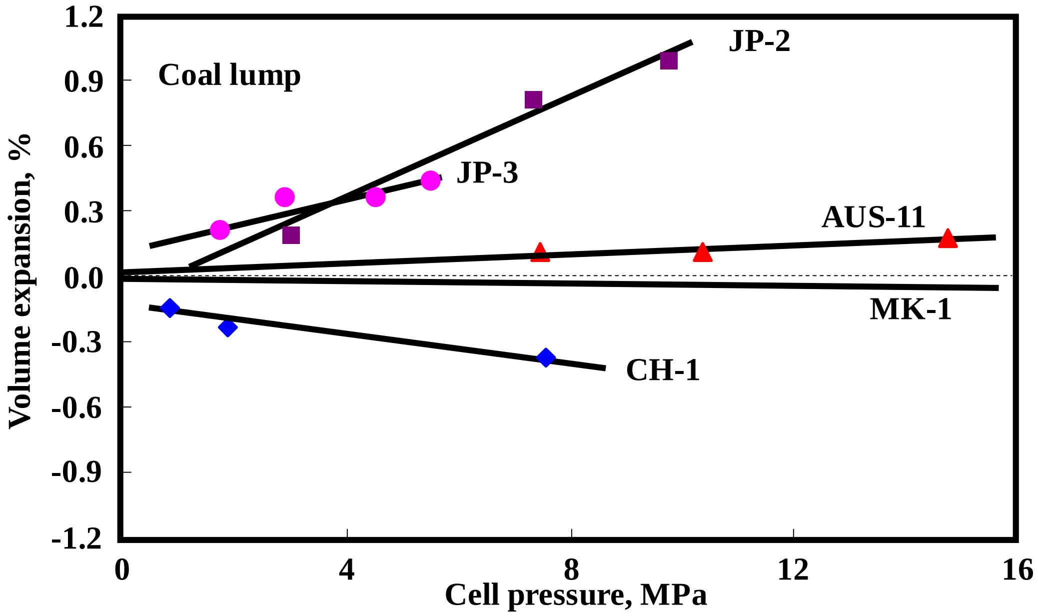


Figure 9: Volume expansion versus cell pressure using the visualization method

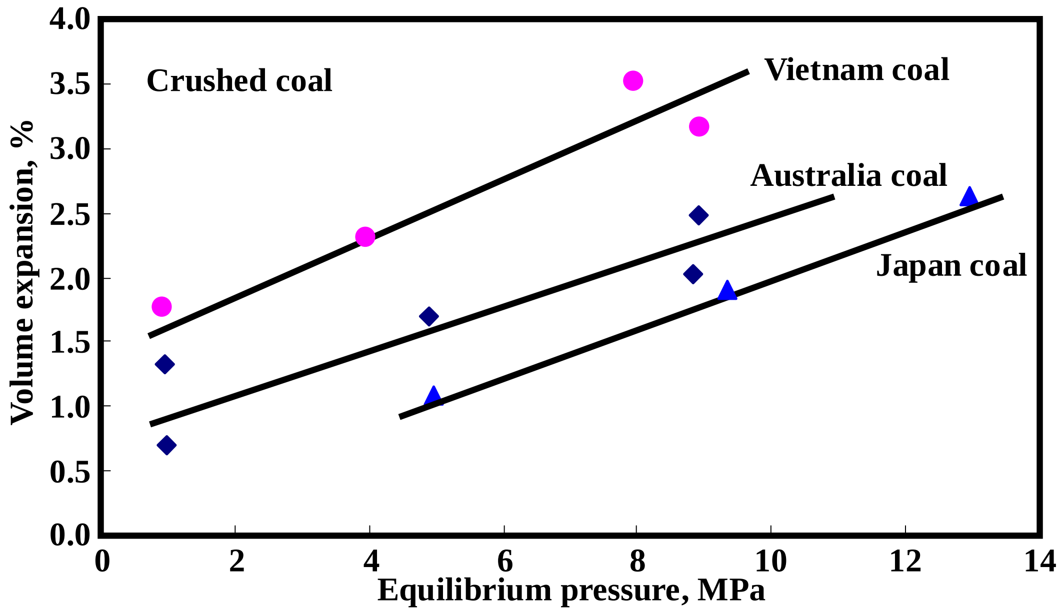


Figure 10: Volume expansion versus equilibrium pressure using the adsorption method

For MK-1 and CH-1 coal samples, the shrinkage effect might be much greater than swelling. Consequently the volume expansion had a negative value.

Negative volume expansion of can also be explained by coal sample characteristics such as ash, moisture, volatile matter content, permeability and fracture aperture. These parameters might affect volume expansion of coal samples.

Volume expansion was confirmed by using an adsorption method. The mass of the samples before and after adsorption at elevated equilibrium pressure was recorded. Results for the Japanese, Australian and Vietnamese samples showed that coal volume expansion increased with increasing equilibrium pressure. Volume expansion was from 1.65 to 3.5 % for

Vietnamese coal, from 0.63 % to 2.5% for Australian coal and from 1.07% to 2.6% for Japanese coal, with equilibrium pressure ranging 1 to 13 MPa (Fig. 10).

Samples used for adsorption measurements were crushed coal, with particle sizes from 1 to 2 mm. This can explained why the volume expansion results in Fig. 10 are larger than that of Fig. 9. CO_2 has a characteristic of better absorbing into the crushed coal matrix than into the coal lump, because of increased surface area.

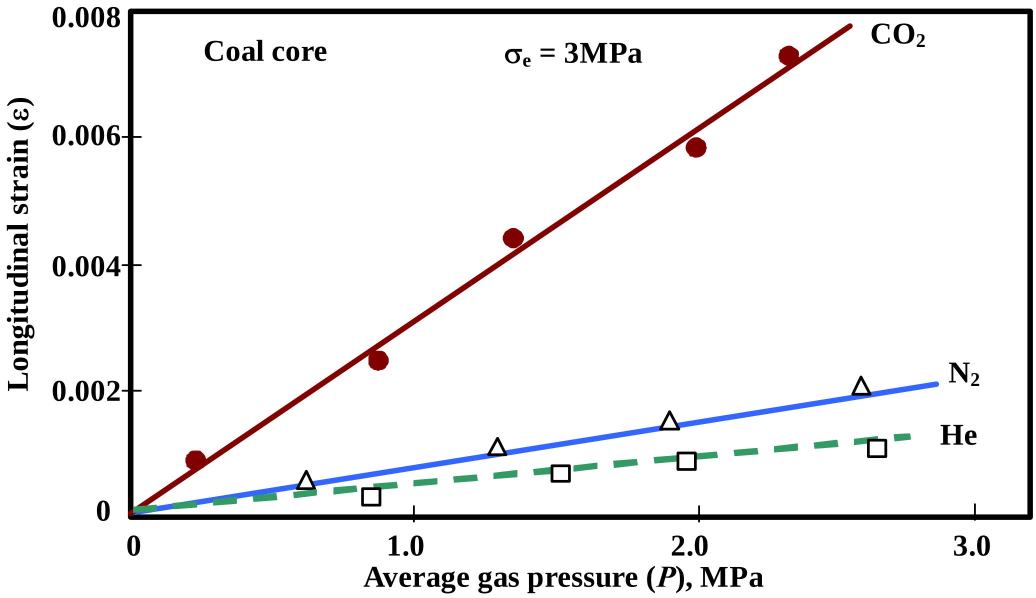


Figure 11: Gas pressure versus strain for three gases (AUS-14).

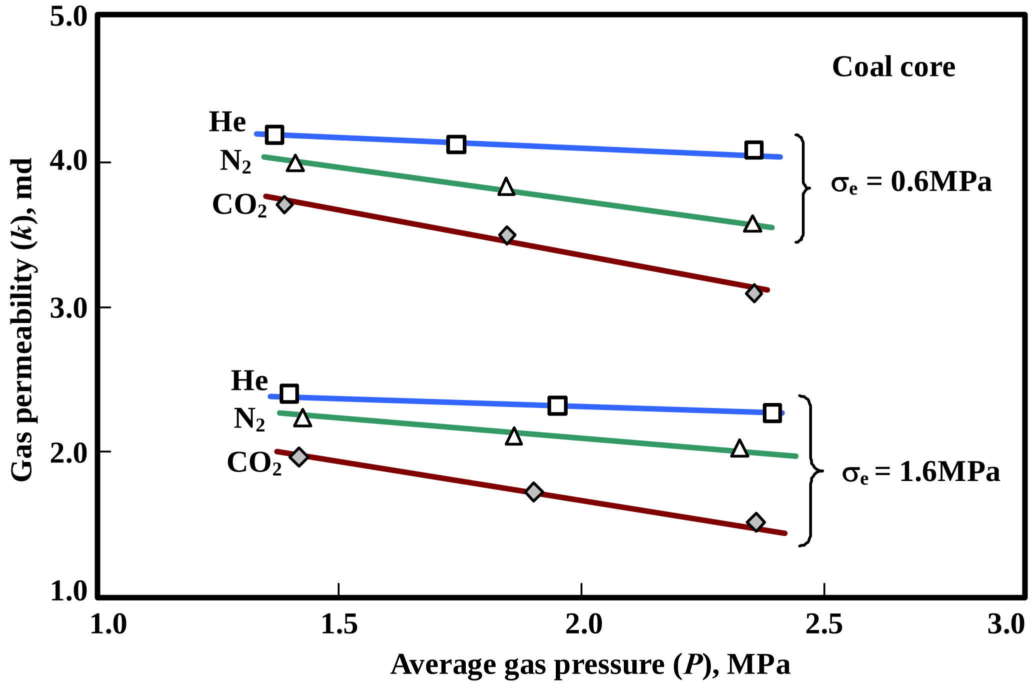


Figure 12: Permeability versus average gas pressure

The longitudinal strain of the core length for three gases (CO_2, N_2 and He) were measured (see Ichikawa et al., 2009). Figure 11 shows the measured results of Longitudinal strain, ε, against average gas pressure of coal core samples measured with effective stress (= confined stress – gas pressure), σ_e = 3 MPa. It is considered here that He has little adsorption on internal surfaces in coal matrices at room temperature (25°C). Thus, the incipient strain caused by He gas pressure increased with increasing mechanical stress, and was small compared with strains of N_2 and CO_2. On the other hand, the strains for N_2 and CO_2 gases depend on not only gas pressure, but also CO_2 gas adsorption which has 3.8 times larger adsorption capacity onto coal internal surfaces compared with N_2. In this analysis, the strain for CO_2 gas increased steeply with increasing gas adsorption as a function of pressure. As a result, the coal strain for CO_2 was about 8 times higher than that for He and about 4 times higher than for N_2. This difference in strains derives from swelling of coal matrices by gas adsorption. The results indicate that the permeability should be corrected by swelling conditions, which is a function of gas type and gas pressure.

Figure 12 shows the relationship between gas permeability and gas pressure using three types of gases (CO_2, N_2 and He) under constant effective stress (σ_e= 0.6 and 1.6 MPa). Theoretically, permeability is not dependent on gas pressure without a low pressure range with a slip effect of gas molecules. In the case of He gas, the permeability showed a constant value against gas pressure. In contrast, permeabilities with N_2 and CO_2 decreased considerably with increasing average gas pressure. With the range of average gas pressure from 1.5 to 2.5 MPa, the permeability reduction was 10.0 % to 13.6 % for N_2 and from 15.7 % to 17.8 % for CO_2. This phenomenal can be explained by the fact that N_2 and CO_2 may have been absorbed into internal surfaces of the coal matrix, creating a swelling effect. Consequently, the width of fractures became smaller and smaller, and permeability decreased with increasing average gas pressure. CO_2 can be absorbed into a coal matrix more strongly than N_2. Therefore, the permeability attenuation of CO_2 gas was much larger than that of N_2 gas. One of the methods used in numerical simulations in considering permeability reduction against gas pressure is the model by Palmer & Mansoori (1998), which expressed permeability reduction as shrinkage of micro-pores. The results shown in Fig. 11 will be basic data for the model.

The CO_2 adsorption volume at sub-critical and supercritical condition was calculated by using following equation:

$$Q_{ad} = \Delta\rho V_{void} \quad (3)$$

where: Q_{ad} = Adsorption amount, (g); $\triangle\rho$ = difference of CO_2 density at the initial and equilibrium pressures, (g/cc); V_{void} = Void volume of cell, (cc).

Once swelling effect occurs during adsorption measurement, volume of coal samples is increased gradually with increasing of cell pressure. This causes to reduce the void volume in adsorbate phase. Consequently, the actual adsorption volume is smaller than that calculated by equation (3). Following equation has been proposed to estimate the adsorption volume considering to the volume expansion caused by swelling effect.

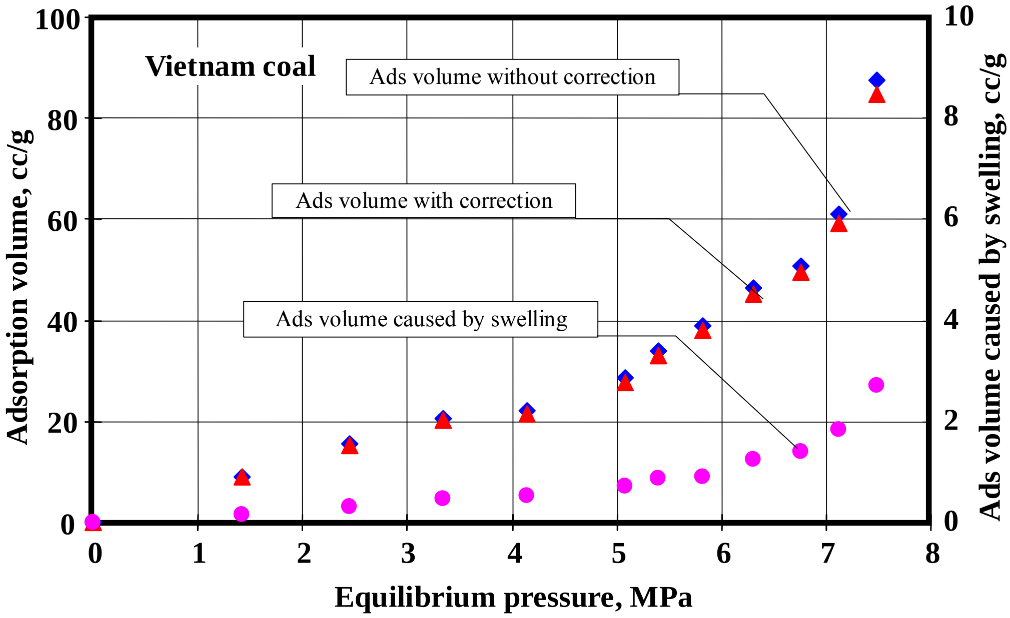


Figure 13: Correction of adsorption volume caused by volume expansion of Vietnam coal

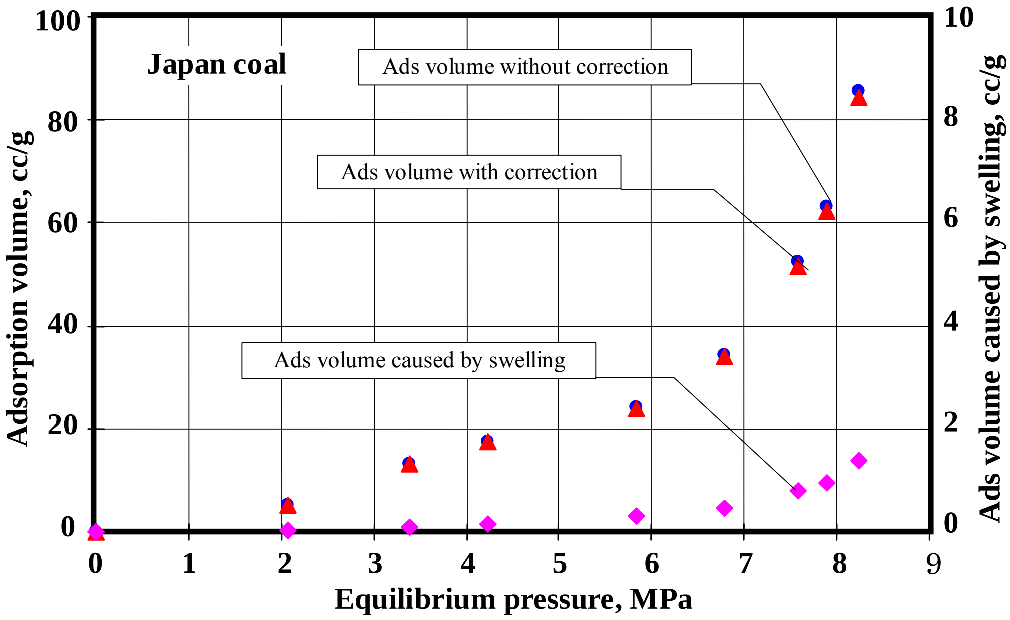


Figure 14: Correction of adsorption volume caused by volume expansion of Japan coal

$$Q_{ad} = \Delta\rho\left(V_{void} - V_{swell}\right) \qquad (4)$$

where: V_{swell} = expansion volume caused by swelling effect, cc

By applying equation 4, CO_2 adsorption volume of Vietnam, Japan and Australia coal has been corrected (see figure 13, 14 and 15). Actual adsorption volume with correction is shown in these graphs in red triangle marks.

In fact, adsorption volume caused by swelling effect is not large comparing with the total adsorption volume. Therefore, this parameter is usually neglected in the most estimation of adsorption. However, several simulators have been considered about shrinkage or swelling effect occurred during adsorption/desorption process to get more accuracy results. Thus, this study can contribute to clarify the swelling effect during CO_2 adsorption process.

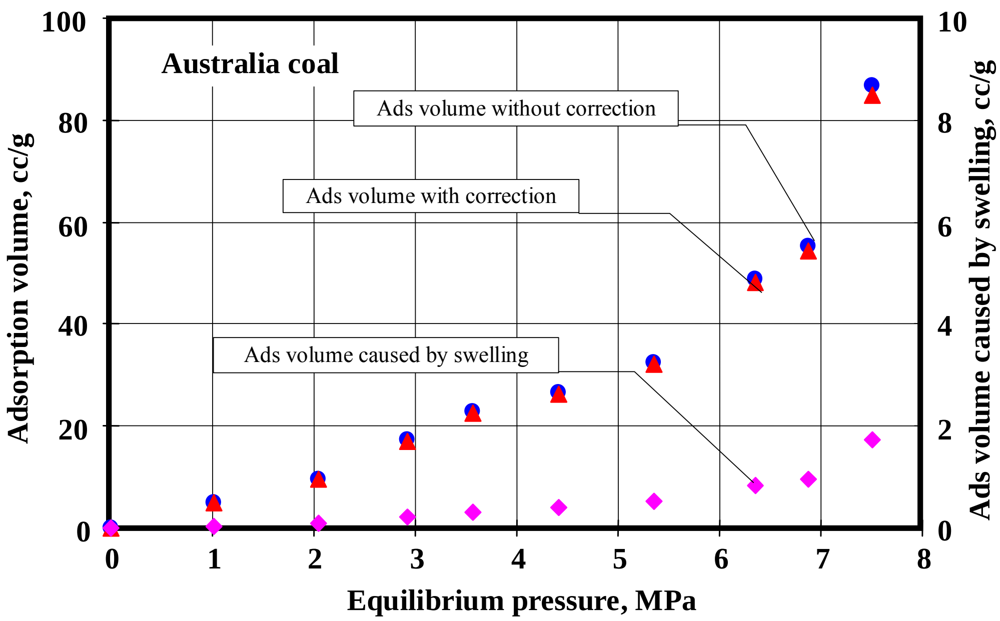


Figure 15: Correction of adsorption volume caused by volume expansion of Australia coal

CONCLUSIONS

An investigation of coal sample swelling in CO_2 (gas, liquid and supercritical) using a visualization method was undertaken at temperatures ranging 25 to 60°C and pressures up to 15 MPa. The swelling effect was observed by using visualization and adsorption results. The following results were obtained:

- Volume expansion phenomenon occurs in the most type of coals and it has been investigated and clarified by several methods.
- While volume expansion of Japanese and Australian coal lumps was from 0.1 to 1.0 %, negative value was observed in the Vietnamese and Chinese coal samples.
- Volume expansion of crushed Vietnamese, Japanese and Australian coals were from 0.5% to 3.6 %.
- Coal strain for CO_2 was about 8 times higher than that of He, and 4 times higher than that of N_2.
- The equation has been revised to estimate the adsorption volume considering to the volume expansion caused by swelling effect.

ACKNOWLEDGMENTS

We would like to thank G-COE and the International Training Project (ITP) of Kyushu University for their financial support.