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Factors Affecting Sorption Induced Strain of Coal Specimens During Carbon Dioxide Injection: A Review Study

[1]Mustafa Abunowara, [1]Usama Eldemerdash and [2]Mariyamni Awang
[1]Department of Chemical Engineering,
[2]Department of Petroleum Engineering,
Universiti Teknologi PETRONAS, Tronoh, 31750, Perak, Malaysia

Abstract: Carbon dioxide (CO_2) capture, utilization and storage (CCUS) are considered as a potential approach to mitigate carbon dioxide emissions into geologic formations. Deep unmineable coal seams have been identified as a possible option because it has large CO_2 adsorption capacity, long time CO_2 trapping and extra enhanced coal-bed methane recovery (ECBMR) and CO_2 sequestration. The current practice for recovering coal bed methane is to depressurize the bed, usually by pumping water out of the reservoir, the desorption of gas from coal surface, diffusion of gas to the fracture systems and flow of the gas through the fractures to the wellbores. Hence, an alternative approach is to inject CO_2 into the coal bed seams to increase the mobililbity of methane recovery. As a result induced adsorption strain (swelling) is one of the main difficulties which face CO_2 sequestration in coal seams. This phenomenon occurs, particularly when the injected carbon dioxide adsorbs on surface of the coal pores and interacts with coal in chemi-physical adsorption isotherm under extreme conditions, which causes the coal to swell. This swelling in confined conditions leads to a closure of coal matrix pores and cleat system, which hinders further CO_2 injection. However, swelling will decrease permeability and adsorption capacity of coal seams and increases CO_2 injectability potential complications. The degree of swelling would be affected by many parameters such as coal rank, water content and petrographic content, mechanical properties (e.g., stress levels and confinements), operating conditions (e.g., gas injection pressure and temperature), free gas and fluid type. Thus these parameters have significant affect on CO_2 continuous injection process in coal seams in long term. This study is a reviewing for the main parameters which have influence on coal swelling during carbon dioxide injection in coal specimens.

Key words: Coal seams, CO_2 sequestration, swelling, gas sorption, supercritical CO_2

INTRODUCTION

Coal is known to swell when gases adsorb chemically and physically onto its surface (Day *et al*., 2010; Karacan, 2007; Kelemen and Kwiatek, 2009; Majewska *et al*., 2009) and when CO_2 interacts with wet coal. Swelling of the coal during adsorption of CO_2 is one of the obstacles of CO_2 sequestration in coal seams as it causes coal seam permeability to be significantly reduced (Perera *et al*., 2011). Coal sorption induced strain (swelling) depends on a number of factors including coal rank, coal seams pressure and temperature, gas nature and type and duration of gas injection and stress and confining pressure. In unconfined coal, swelling in CO_2 is typically less than about 5% by volume but this is nevertheless important when considering enhanced coalbed methane (ECBM) production or CO_2 sequestration because it can affect the gas transport properties of coal seams. Theoretical (Pan and Connell, 2011) and experimental (Harpalani and Mitra, 2010; Wang *et al*., 2010) studies predict that swelling decreases seam permeability and recent field trials confirm that gas flows are reduced when CO_2 is injected into deep coal seams (Durucan *et al*., 2009; Van Bergen *et al*., 2009; Fujioka *et al*., 2010). Because of the potential of swelling to seriously affect large scale ECBM and sequestration projects. This is one the difficulties which face CO_2 injection into Yubari field pilot test (Kiyama *et al*., 2011). However, the sorption of CO_2 in coal causes deformation of the coal matrix and affects the dynamic permeability and then decrease transport of gases in coal fracture, determining both the rate and capacity of the coalbed seam. In addition to that CO_2 adsorption and CH_4 desorption and can significantly affect the volumetric change of micro and macropores in coal leading to coal swelling or shrinkage and evolution of permeability and then controlling the transport and flow of gases through coal matrix (Wang *et al*., 2010).

Corresponding Author: Mustafa Abunowara, Department of Chemical Engineering, Universiti Teknologi PETRONAS, Tronoh, 31750, Perak, Malaysia Tel: +6017 2902067

Although there are some differences in the results of varied studies, they are mostly consistent and may be due to different techniques and procedures which used by many researchers. The general conclusions can be briefly summarized as follows: Volumetric swelling increases as a non-linear function of injection pressure, approaching a maximum value, usually above 15 MPa and depending on the rank coal, maximum volumetric swelling induced by CO_2 in unconfined samples is generally within the range of about 1-5%, with highest swelling occurring in lower rank coals. However, recently it has been reported that much higher swelling is possible in compacted aggregates of coal (Van Bergen *et al*., 2011). In addition, other gases apart from CO_2 also swell coal to varying degrees gases such as ethane and xenon can swell some coals as CO_2 (Day *et al*., 2010) whereas methane induces about half as much swelling as CO_2 (Day *et al*., 2010; Van Bergen *et al*., 2011) and some gases can swell the coal more than CO_2, particularly H_2S, which could be a component of some flue gases destined for deep coal seam injection (Cui *et al*., 2007). In addition, the majority of coal seams are saturated with water that has been shown to influence swell of coal in different levels, in some cases by nearly as much as CO_2 (Fry *et al*., 2009). Often swelling is greater in the direction perpendicular to the bedding plane compared to the parallel direction (Day *et al*., 2008; Levine, 1996; Van Bergen *et al*., 2009) although some studies report that significant anisotropy was observed (Day *et al*., 2008). The mechanisms of coal matrix swelling base on (Qu *et al*., 2012) modeling results are at the initial stage of CO_2 injection under variable temperatures, matrix swelling due to gas sorption, thermal expansion and the change in adsorption capacity is localized within the vicinity of the matrix fracture. As the injection continues, the swelling zone is widening further into the matrix and the swelling becomes macro-swelling. When the swelling is localized, coal permeability is controlled by the internal fracture boundary condition and behaves volumetrically; when the swelling becomes macro-swelling, coal permeability is controlled by the external boundary condition (Qu *et al*., 2012). Moreover, regarding to what have mentioned above about sorption induced strain of coal. The following section will explain in more details the parameters which have impact on swelling phenomenon during carbon dioxide injections in coal seams.

FACTORS INDUCED COAL SWELLING

CO_2 injection into coal seams under in situ conditions urges chemi-physical adsorption and chemical reactions under confined conditions lead to swelling of coal seam. The degree influence of swelling relies on coal rank and water content and temperature and stress levels and confinements. Swelling leads to volumetric change of intact coal matrix and then leads to fracture cleats blockage which alter coal permeability and decrease CO_2 injection rate. Thus there are many factors which could control swelling strain such as coal rank, water content, petrographic contents, operating conditions (e.g., injection pressure and temperature), mechanical properties (stress and confining pressure). In addition, these parameters play a viable role on altering gas permeability of coal seams and then reducing CO_2 injection rate and coal adsorption capacity.

Coal rank and moisture content: Day *et al*. (2011) observed that swelling was greater in carbon dioxide (CO_2) than methane (CH_4). Experiments conducted on four Australian coals specimens at high injection pressure (up to 15 MPa) and temperatures (up to 55°C). It is observed that lower rank coals swell more than higher rank material and the presence of moisture significantly reduce the amount of additional swelling by the gas compared to dry coals and the degree to which the swelling of the coals was affected by moisture depended on the rank of the coal. In the lowest rank coal, maximum swelling was about 5% in CO_2 under dry conditions compared to about 2% in the highest rank sample (Day *et al*., 2008, 2011). In the dry samples swelling was greater in CO_2 than CH_4 by a factor about 1.7, regardless of the rank of the coal. Meanwhile, in moist coal the ratio was about 2.5, reflecting the proportionally greater effect of water on swelling in methane. It is noticed that moisture significantly reduced the degree of gas-induced swelling in both CO_2 and CH_4. However, the mount to which the swelling was reduced was a function of the rank and nature of the coal. In lower rank coals, the effect of moisture on the amount of swelling is high and swelling was depressed by about 55% whereas the highest rank coal was barely affected (Day *et al*., 2011). Although moist coals specimens swell less than dry samples and this is not rely on coal rank which the same result was reported by N. Siemons and A. Busch as shown in Fig. 1. No specific trend can be observed for the coals containing water while for the dry coals the increase in coal volume decreases at low rank and increases again at higher rank with a minimum vitrinite reflectance at 1.1 to 1.3%. In particular, the low rank coals show a large increase in volume (~11-13%). Coals containing water show no rank dependency. The volume increase for all samples varies between 4 and 8% (Siemons and Busch, 2007).

Thus, if this pre-swelling is induced, the total swelling of coals is high than that induced by the gas in dry coal (Day *et al*., 2011). Swelling was found to be entirely elastic, even after the coal had been subject to multiple exposures to CO_2 phases (Day *et al*., 2008).

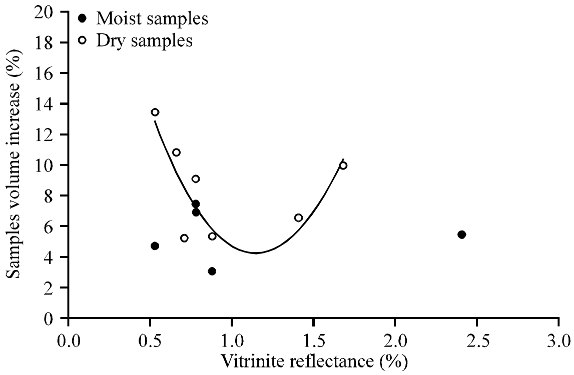

Fig. 1: Show coal volume increase for dry and decrease water-containing samples (Siemons and Busch, 2007)

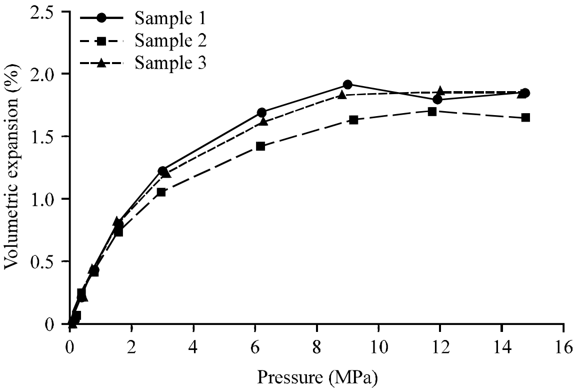

Fig. 2: Volumetric swelling of the three coal samples at 40°C as a function of CO_2 pressure (Day *et al*., 2008)

Below a few atmospheres pressure, swelling is low and generally unaffected by the amount of gas adsorbed, but at increased pressure, swelling becomes roughly linearly proportional to the amount of CO_2 adsorbed. Above about 8 MPa, this relationship was no longer linear, adsorption continued to increase but swelling did out as shown in Fig. 2. Volumetric swelling strain at 15 MPa ranged from about 1.9-5.5% in CO_2 and 1.0-2.5% in CH_4 depending on the rank of coal and the proportion of CO_2 in the gas mixture. Experimental results depict that there is no further enhanced swelling in mixed gases above that would be observed in the pure CO_2 at the same total pressure (Day *et al*., 2012).

Kiyama *et al.* (2011) conducted two laboratory experimental tests to simulate Yubari field pilot test on coal specimens and to understand the change of coal physical properties (permeability) during continuous injection of liquid and supercritical CO_2 and N_2 gas by measuring strain, elastic wave velocity and permeability evolution under stress-constrained conditions. In test I, liquid CO_2 was injected into a water-saturated coal specimen and then heated and injected as supercritical CO_2 and a volumetric swelling strain 0.25 to 0.5% was observed after injecting liquid CO_2. In test II, supercritical CO_2 was injected into a coal specimen saturated with N_2 and then N_2 and CO_2 were repeatedly injected and the swelling strain was about 0.5-0.8% after injecting supercritical CO_2. Following further injection of N_2 in test II, slow strain recovery was observed in the coal and this test was to simulate the case of N_2 injection and CO_2 re-injection at Yubari. The water-saturated coal specimen swelled by 2500-5000μ during liquid CO_2 injection as depicted in Fig. 3. The N_2 saturated dry specimen from the same block swelled by 5000-8000μ during supercritical CO_2 injection as depicted in Fig. 4. The difference between results was a consequence of different injected gas type, different media injection and adsorption capacity of the wet and dry coals specimens. In addition, subsequent N_2

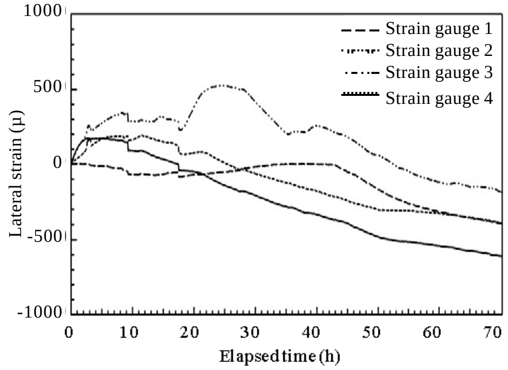

Fig. 3: Change of the lateral strain of coal sample saturated with water (Kiyama *et al*., 2011)

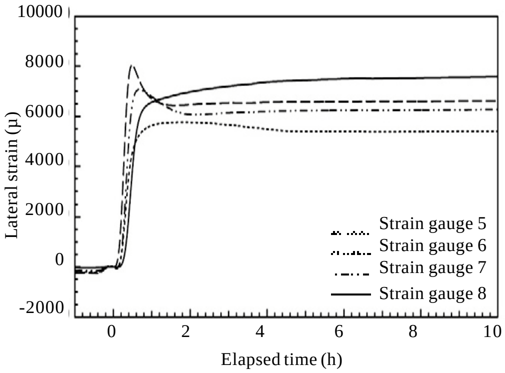

Fig. 4: Change of the lateral strain of coal sample saturated with N_2 gas (Kiyama *et al*., 2011)

flooding tests following CO_2 injection showed a little strain reduction, suggesting that N_2 displaces the adsorbed CO_2 in the coal matrix. Hence, the permeability of the coal specimen was also recovered after N_2 injection, although it declined rapidly after CO_2 injection. These results suggest that when liquid CO_2 was injected into the water-saturated coal specimen, it did not completely displace the water in the coal mixture and indicate that coal swelling is likely to be the main cause for the permeability change in the Yubari field tests and swelling strains were difficult to measure (Kiyama *et al*., 2011).

Fujioka *et al*. (2010) conducted a micro-pilot test with a single well and multi-well CO_2 injection tests, involving an injection and production wells in the period between May 2004 and October 2007. However, there were a variety of tests conducted in the injection well, includes an initial water injection fall-off test and a series of CO_2 injection and fall-off tests (Fujioka *et al*., 2010). Although gas production rate was obviously enhanced by CO_2 injection, water production rate was not clearly affected by CO_2 injection. Several injection tests that injectivity of CO_2 into the virgin coal seam saturated with water was eventually increased as the water saturation near the injector was decreased by the injected CO_2. As result, it was estimated that low injectivity of CO_2 was caused by the reduction in permeability induced by coal swelling. N_2 gas flooding test was performed in 2006 to evaluate the effectiveness of N_2 injection on improving well injectivity and the N_2 flooding test showed that daily CO_2 injection rate was boosted, but only temporarily. Moreover, the permeability did not return to the initial value after CO_2 and N_2 were repeatedly injected. It was also indicated that the coal matrix swelling might create a high stress zone near to the injection well. In addition to that the last estimation on the fracture opening pressure, boost of injection pressure up to 19MPa was tired at the final stage of the pilot test, which exceeded predetermined 15.6 MPa of limit injection pressure and it was possible to inject CO_2 at an injection rate of over 8t/day, but the last value of CO_2 injection rate at 15.6 MPa was below 4 t day^{-1} (Fujioka *et al*., 2010).

Van Bergen *et al*. (2009) observed different swelling behaviors of coal with different substances which carbon dioxide leads to higher strain than methane while exposure to organ leads to very little swelling. The experiments on moisturized specimens seem to confirm the role of moisture as a competitor to gas molecules for adsorption sites. A re-injection of carbon dioxide, after intermediate gas release, results in higher strains which indicate that drying effect of the carbon dioxide on coal specimens Van Bergen *et al*. (2009).

Balan and Gumrah (2009) revealed that swelling increased CO_2 breakthrough time and decreased displacement ratio and CO_2 storage for all ranks of coal. In addition, low-rank coals affected more negatively than high-rank coals by swelling and dry coal specimens are more influenced by swelling than saturated wet coals and saturated wet coals are more suitable for eliminating the negative effects of CO_2 injection and these results agree with the results that found by (Day *et al*., 2008, 2011). Thus it is possible to reduce swelling effect of CO_2 on cleat permeability by mixing it with N_2 before injection but this could happen temporary (Balan and Gumrah, 2009). Swelling phenomenon is sensitive to coal rank and moisture and water content.

Pressure and temperature: Perera *et al*. (2012) carried out experiments on naturally fractured bituminous coal specimens under five gas injection pressures (8, 9, 10, 11 and 13 MPa) under two different confinements (20 and 24 MPa) and five different temperatures (25, 30, 40, 50 and 70°C). The results showed increase in the permeability of naturally fractured black coal with increasing temperature (over 40°C) for supercritical CO_2 injection at higher injection pressures (more than 10 MPa) for any confinement and the permeability increment increases with increasing injection pressure. However, temperature has no much effect on permeability for low injection pressures (less than 9 MPa). In contrast, at low temperatures (less than around 40°C) CO_2 permeability decreases with increasing injecting pressures and this due to supercritical CO_2 adsorption-induced swell as shown in Fig. 5. Meanwhile, at higher temperatures (more than 50°C), permeability increases with increasing injecting pressures. The influence of temperature on N_2

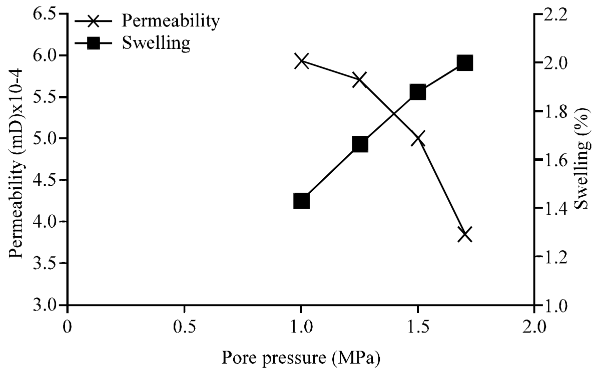

Fig. 5: Effect of pore pressure on gas permeability and swelling at 7 MPa effective stress for CO_2 (Jasinge *et al*., 2012)

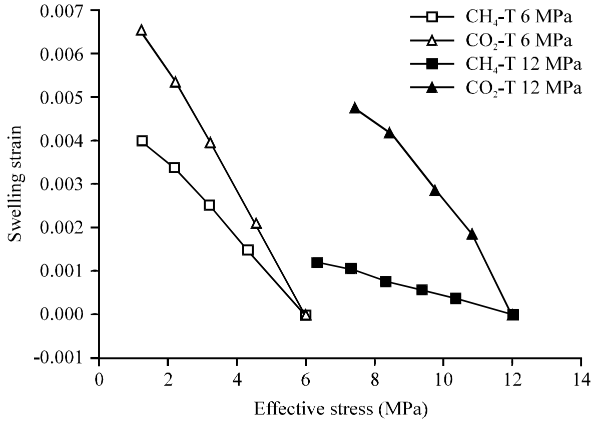

Fig. 6: Effect of applied effective stress on swelling for coal stresses at stepped magnitudes of 6 and 12 MPa and at constant applied confining (Wang *et al*., 2011)

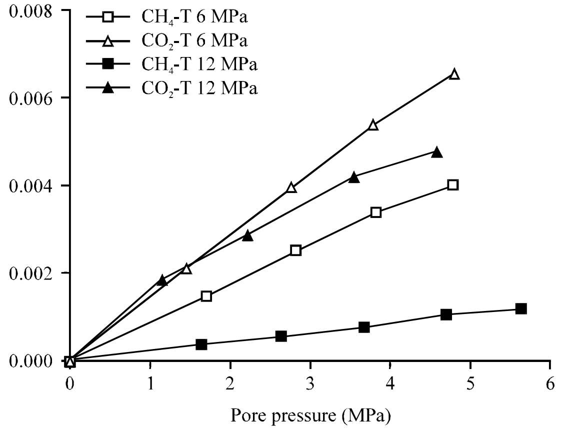

Fig. 7: Effect of applied pore pressure on swelling at stepped magnitudes of 6 and 12 MPa for constant applied confining stresses (Wang *et al*., 2011)

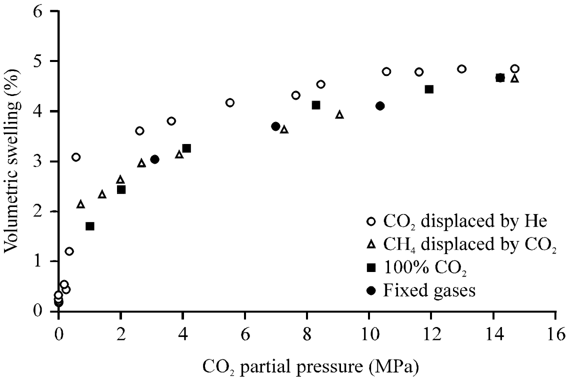

Fig. 8: Volumetric swelling of Coal as a function of CO$_2$ partial pressure (Day *et al*., 2012)

permeability was negligible compared to the CO$_2$ permeability, which is basically due to the fact that N$_2$ is a non-reactive gas (inert) which does not make any adsorption or swelling effect in coal matrix (Perera *et al*., 2012). In addition to that, temperature did not directly affect the maximum amount of swelling, however, the swelling tended to occur at lower pressures with decreasing temperature. Moreover, expressing the swelling as a function of gas density rather than gas pressure showed that swelling was independent of temperature (Day *et al*., 2008).

The effect of swelling strain on coal permeability was investigated using two types of Australian coal specimens and tests were carried out under low different gas injection pressures (2, 2.5, 3 and 3.4 MPa) and under confining pressures (6, 8, 10 and 11 MPa). In addition to that CO$_2$ and N$_2$ gases were used as injection media. However, gas injection was carried out with two stages of N$_2$ injection, prior to and after CO$_2$ injection. As result, for all specimens, the second N$_2$ injection showed a clear permeability reduction compared to the first N$_2$ injection, after exposure to CO$_2$ (Jasinge *et al*., 2012). The coal swelling percentage increased on exposure to carbon dioxide compared to exposure to N$_2$ and this effect increased as gas injection pressure increased and exposure of the coal specimens to CO$_2$ has contributed to a detrimental injection pressure increased. Increasing pore pressure increases swelling and decreases permeability as shown in Fig. 6 (Jasinge *et al*., 2012).

Injection pressure and pore pressure have highly influence on increasing coal swelling at low temperatures and effective stress decreases swelling strain at constant confining pressure as shown in Fig. 6 and 7 (Wang *et al*., 2011).

Swelling strain increases vastly at low CO$_2$ concentrations but increases steadily at high CO$_2$ concentrations and the same behaviour with CO$_2$ partial pressure in gases mixture that swelling strain increases vastly at low CO$_2$ partial pressure but increases steadily at high CO$_2$ partial pressure as demonstrated in Fig. 8 and Fig. 10. Nevertheless, Swelling decreases by time as shown in Fig. 9.

CO$_2$ phase and time: Perera *et al*. (2012) have conducted experiments on naturally fractured black coal specimens at 2-20MPa injection pressures under 10 to 24 MPa confining pressures for sub/super critical CO$_2$ and N$_2$ gas at 33°C. The experimental results depicted that the

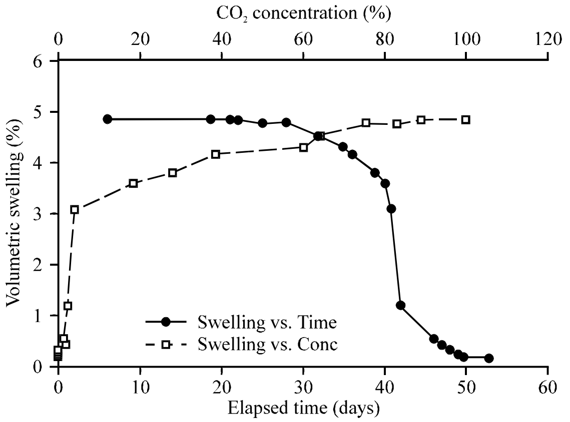

Fig. 9: Volumetric swelling of coal as a function of time during the experiment where CO_2 was displaced by He gas (Day *et al.*, 2012)

permeability of naturally fractured black coal is significantly reduced due to matrix swelling which starts as quickly as within 1 h of CO_2 injection. A further reduction is then observed and the maximum swelling rate occurs within the first 3-4 h of CO_2 adsorption. The amount of coal matrix swelling due to CO_2 adsorption clearly depends on the phase condition of the CO_2 as depicted in Fig. 8. And super-critical CO_2 adsorption-induced swelling is about two times high than that induced by sub-critical CO_2 adsorption (Perera *et al.*, 2011;Day *et al.*, 2012). Interestingly, although a fractured coal specimen which has already fully swelled under sub-critical CO_2 adsorption can swell significantly more under super-critical CO_2 adsorption. However, after that conditions no further swelling effect occurs under any CO_2 pressure or phase condition as depicted in Fig. 9. Moreover, the swelling process continues longer under super-critical CO_2 adsorption. It is concluded that super-critical CO_2 adsorption can induce more matrix swelling than sub-critical CO_2 adsorption under the same adsorption pressure. If the effect of adsorption time on swelling is considered, the swelling rate decreases drastically with time as shown in Fig. 9. Furthermore, the maximum swelling rate can be observed within the first 3-4 h of CO_2 adsorption and this swelling process ends the third day of CO_2 adsorption process (Perera *et al.*, 2011).

Gas composition: During enhanced coalbed methane recovery or CO_2 sequestration, the composition of the gas within the seam will change with time while the total gas pressure remains approximately constant. Day *et al.* (2012) reported that swelling results show a pressure dependence on the ratio of CO_2/methane swelling as shown in Fig. 10 and 11. Despite the linear dependence of swelling on the CO_2 concentration for different coals as depicted in Fig. 13, slight differences in the slope of the

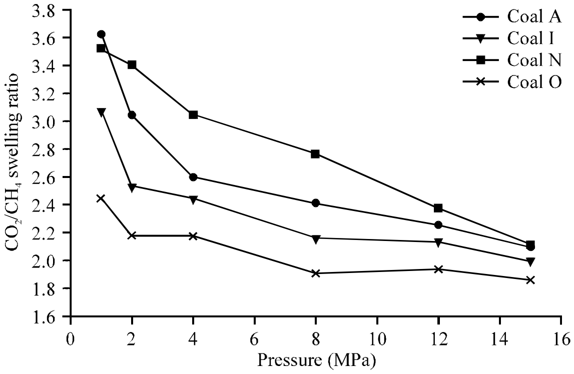

Fig. 10: The ratio of CO_2/CH_4 swelling as a function of pressure for the four coals (Day *et al.*, 2012)

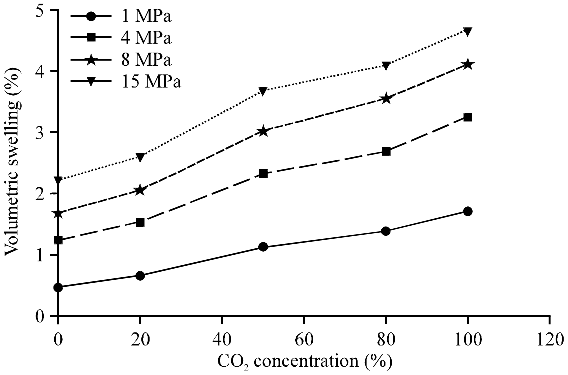

Fig. 11: Volumetric swelling of coal A as a function of CO_2 concentration at various pressures (Day *et al.*, 2012)

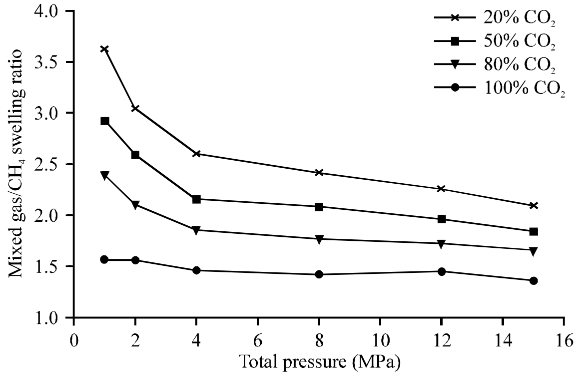

Fig. 12: Mixed gas/CH_4 swelling ratio and total pressure as a function of CO_2 concentrations (Day *et al.*, 2012)

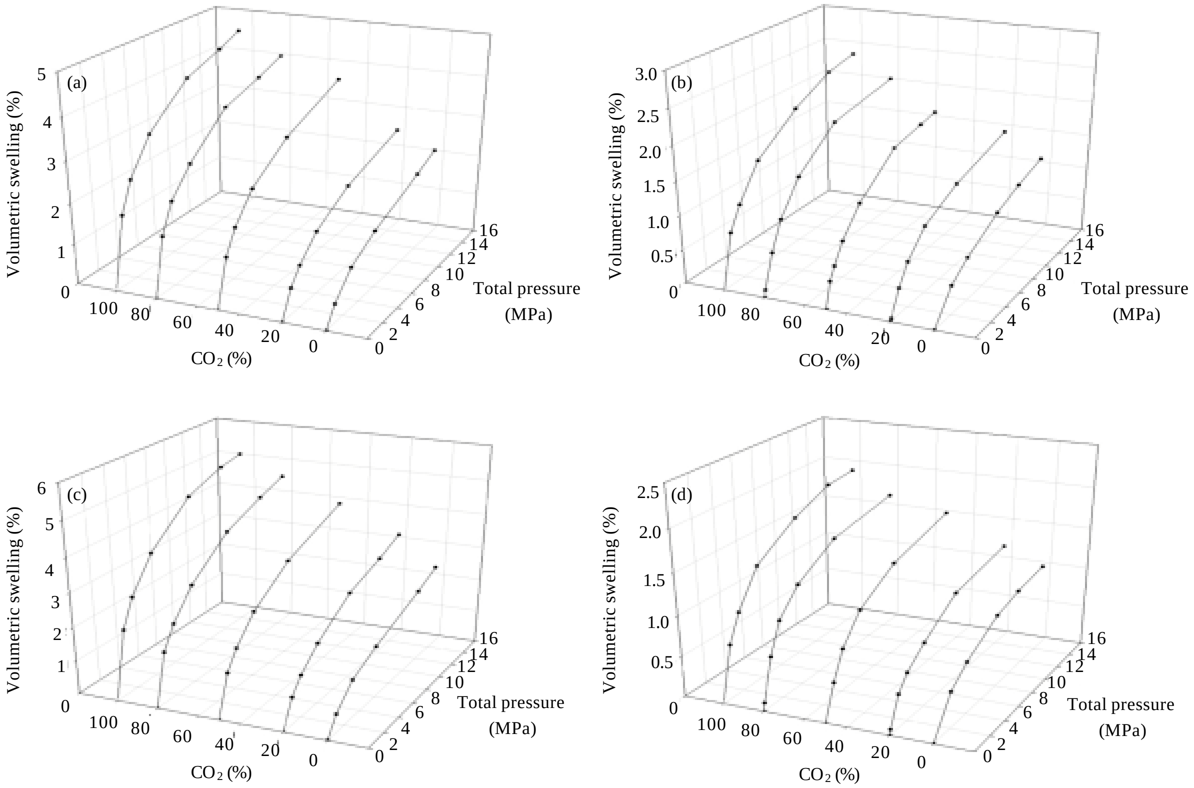

Fig. 13(a-d): Volumetric swelling as a function of pressure and CO_2 concentration in the bulk gas for the four coals (Day *et al*., 2012) (a) Coal A, (b) Coal I, (c) Coal N and (d) Coal O

relationship at different pressures leads to different CO_2/CH_4 swelling ratios across the pressure range and there is no enhanced swelling in fixed mixed gases (100% CO_2, 80% CO_2/20% CH_4, 50% CO_2/50% CH_4, 20% CO_2/80% CH_4 and 100% CH_4) would be observed in the pure critical CO_2 at the same total pressure (Day *et al*., 2012). In gases mixture of fixed composition, the swelling induced was between that pure critical CO_2 and methane with swelling increasing in proportion to the concentration of CO_2 in the mixture. It is also apparent that in mixed gases, the primary driver for swelling is the partial pressure of the component gases (Day *et al*., 2012) as demonstrated in Fig. 12. Exposure of coal specimens to other gases such as amines, H_2S or NO had no effect on the sorption characteristics of the coal. In contrast, SO_2 markedly reduced the CO_2 sorption capacity of the coal by 25% and SO_2 modified the minerals matter extensively and reacted with clays and carbonated and producing a range of sulfate minerals and amorphous materials (Sakurovs, 2012). In contrary, Bustin (2004) conducted experiments for adsorption isotherm and volumetric measurements on British Colombia coals with various gases. It is concluded that the overall the sorption capacity and volumetric strain increase with increasing pressure steps until they reach their saturation pressures. Comparison of strain at about 0.6 MPa pressure step show that the volumetric strain (swelling) with H_2S is very high and about 20 times higher than CO_2 and about 70 times higher than CH_4. The order of volumetric swelling at 0.6 MPa in decreasing order is H_2S (up to 10.0%)> CO_2 (up to 0.66%)> CH_4 (up to 0.29%)> N_2 (up to 0.026%) (Bustin, 2004).

CONCLUSION

Swelling phenomenon occurs during carbon dioxide (CO_2) injection into coal seams at in situ conditions and triggers complex chemo-physical adsorption and chemical reactions within coal seams. Thus coal rank, moisture/water content, injection gas pressure, confining pressure, effective stress and temperature and CO_2 injection duration time have a significant influence on coal sorption induced strain and coal adsorption capacity. However, the relationship between swelling and CO_2 injection pressure are not constant particularly with coal specimens saturated with water meanwhile swelling in dry coal specimens increases with increasing CO_2 injection pressure. Nevertheless adsorption capacity increases by increasing CO_2 injection pressure. However, temperature has no much effect on permeability for low injection pressures. The moistured coal specimens swell less than dry coal specimens and the degree to which the swelling of the coals was affected by moisture depended on the coal rank. Super-critical CO_2 can induce more matrix swelling than sub-critical CO_2 adsorption under the same adsorption pressure and if the effect of adsorption time on swelling is considered. Swelling phenomenon is sensitive to effective stress. Hence swelling strain decreases with increasing effective stress. The volumetric strain (swelling) with SO_2 and H_2S were markedly very high compared to volumetric strain caused by CO_2 and CH_4. Similarly swelling caused by CO_2 is 2-5 times higher than the CH_4 Shrinkage and CH_4 shrinkage is 10 times higher than the swelling caused by the N_2 adsorption. Due to SO_2, CO_2 and H_2S have strong adsorption and swelling effects, sequestration of SO_2 or H_2S or CO_2 into coal specimens significantly reduced cleat permeability and affected strongly the injection efficiency. Particularly, SO_2 and H_2S injection will likely causes the coal to become impermeable. The volumetric strain is one of the main difficulties exists during CO_2 injection field trial into coal seams and it is difficult to measure.

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