ABSTRACT

Coal permeability is very sensitive to the volumetric changes induced in coal matrix as a result of sorption of gas(es). Although there have been no studies to investigate the matrix swelling effect resulting from CO₂ injection, and its influence on coal permeability, there is a strong suspicion that there is such an interdependence. This paper presents the details of a laboratory study currently underway to investigate the effect of increasing the amount of CO₂ retained in coal on its volumetric strain, and hence, on its permeability. The experimental part of the study included measurement of volumetric strain to determine the effect of injecting CO₂ in deep coal with, and without, simultaneous displacement of methane. The experimental results obtained to date clearly indicate that coal swells significantly when exposed to carbon dioxide. There is also a clear indication that the strain induced by CO₂ is higher that that induced by methane. The results are used to determine the changes in the cleat aperture of coal, and hence, its permeability. Although the results are preliminary at this stage, there is no doubt that injection of CO₂ results in a significant decrease in permeability.

INTRODUCTION

Enhancement of oil recovery from oil-bearing rocks by injection of carbon dioxide (CO₂) is a standard practice [1]. Over the last decade, there has been some effort to use this technique to enhance the recovery of coalbed methane (CBM) from coal reservoirs. The effort has been augmented since the technique provides a means to sequester CO₂, a greenhouse gas, in deep and unmineable coals. In fact, this technique is currently considered the only practical and feasible alternative for CO₂ storage while other technologies are being studied in detail. Coalbed reservoirs are considered to have good potential to sequester large quantities of CO₂ because of the ability of coal to adsorb CO₂, the ease of availability of deep reservoirs throughout the world, and their proximity to power plants which are considered to be the main source of CO₂ emissions [2]. Also, a considerable amount of knowledge has been acquired, technology and models developed in the area of coalbed methane recovery, all of which can be easily adapted to CO₂ flow and storage. Thus the concept of CO₂ sequestration, coupled with enhancement of coalbed methane recovery to serve as an incremental energy source, is considered to provide a good synergy for long term benefits, one being environmental and the other economical. Furthermore, enhanced recovery could make marginal coal properties economically attractive and also make deep coal reservoirs viable targets. To increase the ultimate reserves significantly, the enhanced recovery process should result in a reduction in the amount of methane left adsorbed in the micropores at the economic limit of the production, and accomplish the reduction economically by injection of CO₂. Finally, if the rate of production is increased considerably, improvements in the rate of return on the investment of enhancement might justify the additional cost of injection [3]. The enhancement of coalbed methane production by CO₂ injection has, therefore, resulted in renewed thrust on the study of CBM enrichment.

It is well known that the storage and flow of gas in coal is related to its matrix structure and fracture network. It is also known that there is a significant change in the volume of coal matrix associated with desorption/adsorption of gas which, in turn, results in a wide variation in the permeability thus impacting continued production [4, 5, 6]. The influence of this volumetric change, and its impact on the flow behavior, of coal is a phenomenon with significant impact on recovery of methane from coalbeds. A number of studies, theoretical, numerical and experimental have been conducted in the past to assess
the changes in matrix volume due to desorption of methane. These studies have concluded that matrix shrinkage resulting from desorption of methane causes the coal to shrink, and this matrix shrinkage results in opening up of the cleat system causing the permeability to increase [4, 5, 6, 7, 8, 9, 10]. However, there have been no studies to investigate the matrix swelling effect resulting from injection of carbon dioxide and its influence on coal permeability, although there is a strong suspicion that there is such an interdependence since CO$_2$ is significantly more sorptive than methane [11]. Field observations to date support this suspicion. A complete understanding of the interaction of CO$_2$ with coal, displacement of methane by CO$_2$ because of its ability to sorb preferentially, the impact of adsorption on cleat aperture and coal permeability is, therefore, essential in order to predict the long-term effects of CO$_2$ injection.

This paper presents the results of an experimental investigation currently underway to estimate the changes in flow characteristics of coal as a result of volumetric strain associated with CO$_2$ injection. The measured volumetric strain is used to determine the changes in cleat aperture, and hence, the permeability of coal.

BACKGROUND

Most coalbed methane (CBM) operations employ the pressure depletion principle for recovering methane. This is known as the primary recovery method, and involves pumping off large volume of formation water to lower the reservoir pressure and allow the methane to desorb form coal [12]. Primary recovery methods cannot recover more than 50 percent of original gas in place, beyond which the flow rate becomes so low that the operation ceases to be economical [13]. Thus a substantial amount of methane is left behind in the coal. In order to improve the overall recovery from a CBM operation using a technique similar to that being used in the oil industry, the concept of enhanced coalbed methane recovery (ECBM) by injection of a second gas was developed. One of the ECBM techniques is the displacement of methane by introduction of a highly adsorbing gas like CO$_2$. The basis of the technique was the results of laboratory tests showing that coal adsorbs CO$_2$ preferentially over methane, the adsorption ratio of CO$_2$ and methane varying between 2 to 7 [11]. The basic principle behind the enhanced coalbed methane recovery by CO$_2$ injection is that coal releases the sorbed methane when it comes in contact with CO$_2$. With this method, it is possible to increase the CBM recovery up to 90%, or more, of gas-in-place [14]. An added advantage of using CO$_2$ is that it gets sequestered permanently in coal. The CO$_2$ generated from industrial activities can thus be injected into CBM reservoirs in order to enhance methane recovery, while simultaneously sequestering the CO$_2$, provided that CO$_2$ can be separated easily and economically. For long-term methane enhancement and CO$_2$ sequestration, the other important factor is the impact of injection of a second gas on the microstructure of the coal and its flow behavior.

Studies have already demonstrated that changes in coal matrix and cleat system due to desorption can have profound effect on the reservoir permeability, thus affecting the production performance [4, 5, 6]. Gray et al. [8] first proposed the shrinkage of coal matrix associated with desorption of gas and its impact on permeability, which was later demonstrated by Harpalani and Schraufnagel [4, 5]. Harpalani and Chen [7] measured the changes in the coal matrix volume associated with gas desorption, and calculated the corresponding change in coal porosity. Their results showed a linear relationship between coal matrix volumetric strain and the quantity of gas released. Several other researchers have shown a similar trend. Seidle et al. [9] measured coal matrix shrinkage due to gas desorption and showed that sorption induced volumetric strain is correlated with sorbed gas content. Levine [10] used a model to evaluate the matrix shrinkage effect on permeability, and the model results showed that predicted change in permeability is strongly dependent on the matrix shrinkage coefficient and the elastic moduli of the coal. Palmer and Mansoori [15] developed a shrinkage model that describes permeability as a function of matrix shrinkage, and showed that, apart from matrix shrinkage coefficient and the elastic moduli, cleat porosity also plays an important role.
Swelling of coal due to sorption of liquids is a known phenomenon, and has been reported by several researchers [16, 17]. Swelling of coal in the presence of an adsorptive gas has also been investigated in the past. Moffat and Weale [18] reported studying the swelling and shrinkage of coal with adsorption or desorption of methane in order to interpret the sorption isotherms correctly. The study concluded that sorption of gas results in a change in the coal volume. Another study was carried out to investigate how the surface area and pore structure of coal are altered when an adsorptive gas is used for these measurements. Reucroft et al [19] carried out dilatometric studies on coal samples in various gaseous environments and observed significant swelling or volume increase in a range of coal samples when exposed to carbon dioxide. This work was followed by another study to evaluate the effect of pressure on swelling behaviour of typical coals [20]. The results showed that swelling increased with increasing pressure. However, for this study, only the change in the specimen length was monitored. Assuming isotropic behaviour, the change in the specimen volume was estimated.

Stefanska et al [21] conducted simultaneous sorption and dilatometric studies of coal cubes under the action of carbon dioxide and methane gases. The conclusion of the study was that the sorption-induced swelling or shrinkage for coal and carbon dioxide is reversible, but for coal and methane it is not. Also, the degree of swelling of coal was found to depend on its rank and amount of moisture in the coal. Karacan et al [22] studied changes in the sorbed gas concentration and matrix properties of confined Pittsburgh coal samples, when pressurized with CO2, using dual X-ray computed tomography. The quantified variations in bulk density and effective atomic number were used to calculate the amount of sorbed gas, changes in the coal matrix, and kinetics of the complex heterogeneous process occurring with CO2 injection. It was found that the sorption and swelling behavior of coal is heterogeneous and dependent on the microlithotypes present in coal. Larsen et al [23] studied the effect of dissolved CO2 on coal structure and concluded that significant changes in the structure of confined coals rapidly occur upon CO2 uptake.

Field experience with CO2 injection is limited to the Allison Unit (operated by Burlington Resources) in the San Juan Basin, Upper Silesian coal basin of Poland (RECPOL project), and Alberta Sedimentary Basin of Canada (by Alberta Research Council) [24, 25, 26]. RECPOL and Alberta projects are still in their development phase, although some preliminary assessment has been completed. The Allison unit CO2 injection operation started in 1995 and continued for over three years. Analysis of the injection data collected suggests a significant reduction in coal permeability [24]. However, further testing and field demonstration are needed to validate the results of the analysis.

It is evident from the discussion above that any volumetric changes in coal matrix affect the cleat aperture, and thus the gas flow characteristics of coal. Since there have been no in-depth studies related to the impact of sorption induced swelling on flow behavior of coal, a preliminary experimental study was initiated to estimate the changes in permeability as a consequence of adsorption of CO2. Although the primary objective of the current study was to determine the matrix volumetric change associated with CO2 injection and estimate the impact of matrix volumetric change on coal permeability, the sorption related volumetric strain was also measured for methane-CO2 exchange to understand the swelling relative to shrinkage induced by desorption of methane.

**EXPERIMENTAL WORK**

Based on past experience of the authors and a review of similar work carried out by other researchers, it was decided that the experimental setup would take into consideration the following factors:

1. The effects of matrix and fracture compressibility due to change in pressure will be eliminated so that the measured strain is the result of volumetric changes resulting from changes in the gas composition alone. For this reason, the experimental samples were subjected to constant total pressure throughout the experiment. The result was a constant effective stress but the measured strain was the result of sorption induced matrix strain alone.
2. Since the experimental temperature and moisture content of the sample affect the quantity of gas sorbed, experiments were carried out at constant temperature, representative of the reservoir, and controlled moisture conditions.
Sample Preparation

Large blocks of coal were collected from a coal mine. The blocks were kept under water to prevent oxidation during transportation and storage in the laboratory until it was time to prepare the test specimens. The blocks were cut using a handsaw and rectangular specimens were prepared. The samples with the least cleats were selected for experimental work. Subsequent to preparation, the test specimens were kept in an environmental chamber under controlled conditions of temperature and humidity.

Experimental Setup and Procedure

The experimental setup for the study was designed to enable measurement of volumetric strain due to changes in gas composition while keeping the total gas pressure constant. A schematic of the experimental setup is shown in Figure 1. The main components of the setup were pressure vessels capable of withstanding very high pressures, a data acquisition system to monitor strain, and a gas chromatograph (GC) to measure the composition of gas mixtures in the pressure vessels. As shown in the diagram, the setup consisted of three pressure vessels, capable of testing three coal samples simultaneously. Since sorption is very sensitive to temperature, the pressure vessels were placed in a constant temperature bath. Three strain gages were affixed to the surface of each sample in order to monitor strains in the three orthogonal directions. The gages were affixed using an epoxy recommended by the manufacturer. After attaching the gages, leads were attached to each strain gage. The entire assembly was then placed in the pressure vessel with the required outlets for gage connection to a data acquisition system. This procedure was repeated for all three test samples.

All three samples were first subjected to increasing helium pressure in steps of 2.76 MPa (400 psi) and volumetric strain was measured at equilibrium for each step. Since helium is a non-adsorptive gas, the measured strain is purely due to the mechanical compression of the solid coal resulting from the change in pressure. After attaining equilibrium at 10.34 MPa (1500 psi), one of the samples was left alone to serve as the control test. The second sample was subjected to increasing concentration of methane, maintaining the total pressure constant at 10.34 MPa (1500 psi). This was achieved by injecting methane while bleeding helium/(helium + methane) mixture out every time methane was injected. This ensured that the solid and fractures in the samples were subjected to same external conditions throughout the experiment, the only difference being the composition of the gases within and around the sample. The procedure was continued until the gas in the vessel was pure methane. Hence, the sample was subjected to varying composition of methane, all the way from 0% to 100%. At each equilibrium step, a sample of gas was analyzed using the GC and concentration of methane was measured. For the third sample, same procedure was followed using CO₂ instead of methane. Once the second sample is completely saturated with methane, carbon dioxide will be injected to determine the volumetric strain for methane/CO₂ exchange.

RESULTS AND DISCUSSION

The first part of the experimental phase involved dosing all samples with increasing amount of helium in pressure steps up to a maximum of 10.34 MPa (1500 psi) while monitoring the strain continuously. The sample was considered to be in equilibrium if the strain remained constant for eight hours or more. This required three to four days for each pressure step. The entire experiment was carried out at a temperature of 45°C (113°F). Using the measured strain for the helium cycle, the volumetric strain was calculated for each sample. Figure 2 shows the volumetric strain with increasing helium pressure. As expected, the volume of coal matrix decreased with increasing gas pressure due to compression of the coal grains. The matrix, or grain, compressibility \((C_m)\), defined as the change in the volume of solid grains as a result of changes in external pressure, is given mathematically as:
\[ C_m = \frac{1}{V_m} \left( \frac{dV_m}{dP} \right) \]

where, \( V_m \) is the volume of solid coal and \( dP \) is the change in pressure. Using the measured strain for helium, this was calculated to be approximately \(-3E-4 \text{ MPa}^{-1} \) (-2.07E-6 psi\(^{-1}\)). The results are similar for all three samples.

For the second part of the experimental work, methane was injected into the second pressure vessel, maintaining the total pressure constant at 10.34 MPa (1500 psi). For the third vessel, helium was bled out to reduce the pressure to approximately 5.86 MPa (850 psi) since the maximum CO\(_2\) pressure in a gas cylinder is approximately 5.86 MPa (850 psi). Once the sample reached equilibrium at 5.86 MPa (850 psi), CO\(_2\) was injected, once again, maintaining the total pressure constant. With subsequent injections, volumetric strain was calculated for all samples for each step. Figure 3 shows the volumetric strain for the three samples for different pressure steps. As expected, there is no further strain in the sample left alone for the duration of the experiment. It is also evident from the graph that the swelling due to carbon dioxide adsorption is significantly greater than that for methane. This is consistent with the fact that the sorption capacity of coal for CO\(_2\) is significantly higher than that for methane [11]. Using the same approach as that used for estimating matrix shrinkage coefficient, the swelling coefficients for samples were calculated.

For methane injection, the swelling coefficient was calculated to be 9.47E-4 MPa\(^{-1}\) (6.53E-6 psi\(^{-1}\)) for pressure up to 7.41 MPa (1075 psi). For CO\(_2\) injection, swelling coefficient was calculated as 2.4E-3 MPa\(^{-1}\) (1.65E-5 psi\(^{-1}\)) for pressure up to 5.24 MPa (760 psi). Interestingly, the ratio of the two coefficients is 2.5, once again, suggesting a direct dependence of the quantity sorbed and the resulting strain.

On attaining equilibrium after each injection step, the concentration of methane and CO\(_2\) in the samples was determined using a gas chromatograph. This was used to calculate the partial pressure of the two gases. The results are shown in Figure 4. The plot shows the change in volumetric strain due to increasing methane and CO\(_2\) pressure in the samples. For pressures up to 5 MPa (725.2 psi), the volume of coal matrix increased by \(-0.6\%\) due to methane adsorption, and \(-1.25\%\) due to CO\(_2\) adsorption.

### Impact of Matrix Swelling on Cleat Porosity and Permeability

The swelling of coal matrix due to CO\(_2\) injection results in a change in cleat porosity, and hence, the cleat permeability. Following the procedure described by Harpalani and Chen [7], and briefly described below, changes in cleat porosity and permeability as a result of changes in the volume of coal matrix were calculated. The increase in the coal matrix volume due to swelling was considered equal to the decrease in cleat aperture \(b\) as shown in Figure 5. This is a reasonable assumption since the total volume of coal in situ (matrix and cleats) remains constant with degasification, or injection of a second gas. Once again, assuming a matchstick geometry [7] and, \(a_1 = a_2\), the initial coal porosity \(\phi_{\text{initial}}\) is given by \(2b/a\).

The change in matrix dimension, \(\Delta a\), as a result of swelling depends on the change in pressure and swelling behavior of the coal, and is given by \(\Delta a = a l_m \Delta P\), where \(a\) is the initial cleat spacing, \(l_m\) is the change in dimension of the coal matrix in the horizontal direction per unit pressure, and \(\Delta P\) is the change in pressure of the gas being injected. Since the flow of the gas is considered along the cleat systems perpendicular to the bedding plane, change in dimensions along the vertical cleats only has been taken into account necessitating the introduction of the term \(l_m\). After a pressure change of \(\Delta P\), the new cleat porosity \(\phi_{\text{new}}\) can, therefore, be written as

\[ \phi_{\text{new}} = \frac{2(b - \Delta a)}{(a + \Delta a)} \]

The ratio of the modified cleat porosity to the initial cleat porosity is then given as:
\[
\phi_{\text{New}} = \frac{2(b - \Delta a)}{a + \Delta a} / \frac{2b}{a}
\]

Substituting for \(\Delta a\) and simplifying gives the following:

\[
\phi_{\text{New}} = \frac{1 - \left(2l_m \Delta P / \phi_{\text{Initial}}\right)}{(1 + l_m \Delta P)}
\]

Hence, using estimates for initial porosity and the measured change in matrix dimensions of coal, change in cleat porosity with changes in pressure can be estimated. Considering the initial cleat porosity to be 1%, the change in porosity with change in pressure was estimated as shown in Figure 6. At CO2 pressure of approximately 2.75 MPa (400 psi), for coal with an initial cleat porosity of 1%, the cleat porosity is reduced to zero. Negative values are obtained for higher pressures since the swelling is greater than the starting porosity of coal. However, since this is a physical impossibility, the results beyond this pressure can be interpreted as suggesting that the flow is taking place through the coal matrix and can no longer be considered fracture, or cleat, flow. With 0.5% as the initial cleat porosity, the decrease this occurs at 1.37 MPa (200 psi). The results are being re-analyzed to ensure that this is, in fact, the case. Furthermore, the results clearly indicate a linear relationship between porosity ratio and the pressure of gas, i.e., with increase in CO2 concentration in the coal, the porosity decreases linearly.

The change in cleat porosity as a consequence of change in matrix dimension results in a change in cleat permeability. For the same matchstick geometry, and considering the same assumptions, the initial cleat permeability \(k_{\text{Initial}}\) is given by \(k_{\text{Initial}} = \frac{b^3}{12a}\). After a pressure change of \(\Delta P\), the new cleat permeability \(k_{\text{New}}\) can be written as:

\[
k_{\text{New}} = \frac{(b - \Delta a)^3}{12(a + \Delta a)}
\]

The ratio of the modified cleat permeability to the initial cleat permeability can then be written as:

\[
\frac{k_{\text{New}}}{k_{\text{Initial}}} = \frac{(b - \Delta a)^3}{12(a + \Delta a)} / \frac{b^3}{12a}
\]

Substituting for \(\Delta a\) and simplifying gives the following:

\[
\frac{k_{\text{New}}}{k_{\text{Initial}}} = \left(1 - \frac{2l_m \Delta P / \phi_{\text{Initial}}}{(1 + l_m \Delta P)}\right)^3
\]

The variation in permeability as a function of the initial permeability is shown in Figure 7. As expected, at CO2 pressure of 3.2MPa (465 psi), the permeability decreases to zero. The results do suggest a continuous decrease in permeability ratio with increasing gas pressure, i.e., with increase in CO2 concentration in coal, the permeability decreases continuously. This correlates well with the field observation at Burlington Resources’ Allison Unit site [24] where a reduction of \textit{in situ} coal permeability from 100 – 130 md to >1 md, a two orders of magnitude reduction, is suspected. However, these results indicating a decrease in permeability are due to changes in volume of coal matrix alone. Since the total
pressure was kept constant throughout the experiment, the change in effective stress was zero, and this effect is has been neglected.

CONCLUSION AND FUTURE WORK

- The most important conclusion of the study completed to date is that injection of CO₂ results in a swelling of the coal matrix, which can have a profound effect on coal permeability. The study further facilitated estimation of the matrix swelling coefficient due to changes in gas composition alone.
- The swelling induced due to CO₂ adsorption is more than twice that due to methane adsorption. This correlates well with the fact that CO₂ is significantly more sorptive than methane.
- The major impact of this swelling is a decrease in cleat porosity of coal. Depending on the initial cleat porosity and adsorption pressure, the cleat porosity decrease to almost zero with continued injection. This decrease in coal porosity results in a decrease of the permeability to almost zero, once again, the time taken to reach this stage depending on the initial porosity.
- The decrease in permeability discussed above is due to change in gas concentration only. Several studies have demonstrated the sensitivity of permeability to changes in effective stress [15]. If the two effects (swelling effect due to change in gas composition only and change in effective stress) are coupled, the overall impact on permeability will be reduced since the increase in effective stress associated with reduction in pore pressure will not occur.
- Since continued injection of CO₂ in coalbed methane reservoirs can induce a significant decrease in permeability of the cleat system, this effect calls for a more detailed study both in the laboratory and field.
- As a continuation of this effort, injection of CO₂ in to the sample saturated with methane will be carried out to estimate the volumetric changes due to methane/CO₂ exchange. The results obtained would be useful in situations where CO₂ is injected not only for sequestration but also for incremental recovery of methane from a coal reservoir. Finally, this will be followed by an experimental study to study the coupled effect of stress and CO₂ induced volumetric changes in the coal matrix. The results will assist in estimating the effect of CO₂ injection, and the resulting changes in effective stress, on the volume compressibility and permeability of the coal.

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REFERENCES


Figure 1: Schematic of the experimental setup to measure the sorption induced volumetric strain.

Figure 2: Volumetric strain of coal samples with increasing helium pressure.
Figure 3: Volumetric strain of coal samples with time for various pressure steps.

Figure 4: Volumetric strain of samples with respect to partial pressure of methane and CO$_2$. 
For the matchstick geometry, the initial cleat porosity \( \phi_{initial} \) of the coal if \( a_1 = a_2 = a \) is given by

\[
\phi_{initial} = \frac{2b}{a}
\]

where, \( b \) is the cleat aperture [27]

Figure 5: Change in cleat geometry with change in matrix volume.

Figure 6: Estimated variation in cleat porosity due to change in matrix volume.
Figure 7: Estimated variation in coal permeability due to change in matrix volume ($\phi = 1\%$).