IMPLICATIONS OF VOLUMETRIC SWELLING/SHRINKAGE OF COAL IN SEQUESTRATION OF ACID GASES

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ABSTRACT

A series of volumetric swelling and shrinkage experiments using N₂, CH₄, CO₂, and H₂S and subsequent numerical modelling was carried out on coals from the Western Canadian Sedimentary Basin in order to assess potential permeability damage or enhancement due to injection of various gases for enhanced coalbed methane production or for sequestration of acid gases. Investigated coals vary in rank from subbituminous ‘C’ (Rₒ₅ 0.46%) to medium volatile (Rₒ₅ 0.62%) and experiments were carried out on samples on an as received basis. Adsorption of N₂, CH₄, CO₂ and H₂S resulted in marked swelling of the coal matrix. The order of volumetric swelling at 0.6 MPa in decreasing order is H₂S (1.4%-9.3%) > CO₂ (0.26%-0.66%) > CH₄ (0.09%-0.30%) > N₂ (0.004% - 0.026%). Volumetric strain is strongly and positively correlated with quantity of adsorbed gas. Overall the sorption capacity and volumetric strain increases with rank however the comparative sorption capacity and volumetric strain vary markedly with coal rank and type of gas. Desorption of CH₄ results in shrinkage of the coal matrix from 0.4% to 0.9% depending on coal rank. The volumetric shrinkage during CH₄ desorption (per cc basis) is significantly greater (1.4 – 4.4 times) than the volumetric swelling caused by N₂ adsorption, therefore N₂ injection will enhance CH₄ production from coalbeds due to increase in N₂ partial pressure coupled with increased permeability of coal beds because of net shrinkage. Injection of pure H₂S or CO₂ will significantly reduce the permeability of coal seams as injection of H₂S causes swelling up to 5 times and CO₂ causes swelling more than 2.2 times the shrinkage created by CH₄ desorption.

Since volumetric strain varies with volume and type of fluid sorbed, drilling and completion fluids may result in swelling or shrinkage of the coal matrix and thus enhance or reduce permeability. Our numerical modeling results indicate that the variation in cleat permeability during acid gas injection into coal seams is strongly affected by the initial cleat porosity and the mechanical properties of coals. Injection of CO₂ can reduce the permeability by 50% for coals with a permeability of 10 md and cleat spacing of 0.5 cm and by two or more orders of magnitude for coals with a lower initial permeability (1 md) and large cleat spacing (1 cm). Injection of pure H₂S results in such marked swelling of the coal that the in-situ coal permeability will be rapidly reduced to such low values that significant sequestration will be impossible. Up to a 5 times increase in permeability is predicted for injection of N₂. Therefore, the injection of a mixture of N₂ and CO₂ or N₂ and H₂S may facilitate sequestration of acid gas.

Based on the results of our studies we have developed a new permeability model for acid gas sequestration taking into account coal matrix shrinkage and variable swelling property of coals with different gases during sequestration. Our model suggests that an optimum coal seam for sequestration of acid gases would have a high initial permeability and, most importantly, a large initial cleat porosity to reduce the effects of permeability reduction induced by coal swelling on the efficiency of the acid gas sequestration.
INTRODUCTION

Coal is in many areas the cheapest and most available energy source. Coal usage is projected to increase by more than 50% over the next three decades [1]. The ability to capture and store the effluent gases produced during the utilisation of coal is necessary if coal is to be utilised as an energy source without contributing to the build-up of greenhouse gases in the atmosphere. In addition, gas sequestration may enhance coalbed methane (CBM) production from low permeable reservoirs [2]. Hence carbon sequestration is a key technology for sustained use of coal as an energy resource.

There are numerous issues which are not fully understood in the sequestration of gases, such as effects on permeability damage or enhancement of the coal beds, when CO₂, H₂S, SO₂, NO₂ or other gases are injected. A very limited body of work exists on volumetric swelling/shrinkage of coals by gas adsorption in the literature and most studies consider only CH₄ and CO₂. In order to assess potential permeability damage or enhancement due to injection of various gases for enhanced coalbed methane production or for sequestration of acid gases, a series of volumetric swelling and shrinkage experiments and subsequent numerical modelling were carried out on variety of coals at The University of British Columbia. In this paper we discuss a new permeability model for acid gas sequestration based on new information generated from our coal matrix swelling and shrinkage experiments on various rank coals with N₂, CH₄, CO₂, and H₂S. The results of the model on implications of CO₂/H₂S sequestration and their effects on ECBM production are also discussed.

EXPERIMENTAL DESIGN

An experiment was set-up to measure the volumetric strain within the coal matrix under changing gas pressure. The design and experimental procedure is similar to the work done by Harpalani and Schraufnagel [3]; and George and Barakat [4] but differs in equilibrium timings and gas dosage methods. The equipment we designed is fully automated for gas dosage (valve opening and closing) and equilibrium timings with a proprietary software interface to minimize the error in deciding the equilibrium conditions, which is critical for the experiments.

A new volumetric adsorption apparatus was constructed to do adsorption and strain gauge measurements. The equipment is designed using high precision pressure transducers (0 to 15 MPa) and data were collected by utilizing a computer interface. The equilibrium at each isothermal point is decided by the proprietary software where the conditions are pre-specified before the experiment. Equilibrium at each isothermal point is assumed to have been reached if the pressure reading was stable (∆P = -0.001 and +0.002) for more than 16 hours (980 minutes) and is monitored by the software. It may take longer time for coals with slow diffusion and shorter time for fast diffusion coals depending on the gas type and temperature. The programme automatically compares the data after 980 minutes and when ever the set conditions are met, it proceeds to next isothermal step and gas is dosed automatically otherwise it continues to compare for next cycle i.e. another 980 minutes for the condition to be met. For our strain experiments on an average each pressure step took between 6 – 10 days to reach equilibrium. Thus for a 12 step adsorption-strain experiment the time required would be between 72 – 120 days depending upon the coal-gas adsorption system. All the steps are performed according to the pre-determined pressure steps. Once all the steps are finished the experiment stops automatically. The data is filtered using a macro where the equilibrium data points for isotherm and strain are collected. Volume adsorbed at each pressure step is calculated using gas compressibility determined with the Peng-Robinson Equation of State and the liquid density of gas at its boiling point. The temperature of the bath is maintained at target temperature (in this case it is 25°C) ± 0.1°C using a PID temperature controller.
Specimen preparation for strain experiments

Core samples from field drilling programme was utilised (3” diameter core Fig. 2) and where cores were not available, lump coal samples were cored in the laboratory to 37 mm in diameter and about 100 mm in length and the specimen ends ground parallel. The specimens were then sealed in polyethylene bags and kept frozen to prevent oxidation or loss of moisture. Two sets of core samples were prepared, one set was used for non-toxic gases i.e. N₂, CH₄ and CO₂ and the other set for H₂S strain experiments. The samples were analysed for moisture and ash content (Table 1).

Four resistance type strain gages were used per specimen; they were adhered 180° apart, two axial and two radial for each sample. The lengths of strain gages were such that they covered nearly 90% of sample length so that representative strain is measured. The strain gage wires from the sample were soldered to another set of wires passing through a hermetically sealed gland assembly and to the computer via an interface to collect the strain data along with isotherm data (Figs. 1 & 2).

Strain experimental procedure

Prior to the start of the experiment strain gauges were calibrated for temperature variation coefficient. The samples attached with gauges were maintained at atmospheric pressure and strains set to zero. The bath temperature was gradually varied from 15°C to 55°C using the temperature controller and the change in strain was recorded continuously. The average axial and radial strains for each cell were then plotted with temperature and the temperature correction coefficients were derived from the plots for each cell. These corrections were incorporated for axial and horizontal strains into the proprietary strain measuring software for calculating the volumetric changes during adsorption and desorption process.

After the temperature calibration the bath temp was brought to 25°C (the experimental temperature) and the samples were evacuated for one-half hour and the system was allowed to reach equilibrium for 4 hrs. Coal sample volume was determined with helium expansion runs up to 6 MPa in increasing steps and also to measure the dead volume inside the vessel which is used for adsorption isotherm calculations. After the void volume runs, the samples were evacuated for one-half hour and system was allowed for equilibrium. The strains were set to zero and the N₂ adsorption runs began with increasing pressure steps up to 5 MPa. The strains were continuously recorded. After the adsorption runs finished, desorption runs were started with decreasing pressure steps to atmosphere. Once the N₂ test was finished, the samples were evacuated thoroughly and the same procedure was adopted with CH₄, CO₂ and H₂S adsorption runs.

CH₄ adsorption runs were started with increasing pressure steps up to 8 MPa. Since we were more interested in CH₄ desorption runs i.e. volumetric shrinkage during methane production, the adsorption cycle for CH₄ was done in two steps to save time. Hence CH₄ isotherm was run for two high pressure steps. The strains were continuously recorded. After the adsorption runs finished, desorption run was started with gradual 14 decreasing pressure steps for methane.

For CO₂ and H₂S adsorption experiments pressures steps varied. For CO₂ the adsorption steps were carried out up to 5 MPa with 14 steps. For H₂S 11 steps up to 1.5 MPa was programmed but at about 0.6 MPa the H₂S began reacting with strain gages and the experiment was stopped.

Data Analysis

The Langmuir model [5] was used for adsorption isotherm data analysis as is conventionally used in the field of coalbed methane. The equation is generally expressed as:
\[ V = \frac{V_L P}{P_L + P} \]  
\[ \text{Where } V = \text{volume of gas adsorbed} \]
\[ P = \text{pressure} \]
\[ V_L = \text{Langmuir Volume} \]
\[ P_L = \text{Langmuir pressure} \]

Since the pressure vs strain data follows the trend of isotherm shape i.e. pressure vs volume of gas adsorbed, an equation having the same mathematical form as the Langmuir equation was used to model the strain data [6] and gives a fairly good fit to the strain experimental data (Fig. 3).

\[ \varepsilon_v = \frac{e_L P}{p + p_e} \]  
\[ \text{Where, } \varepsilon_v \text{ is the volumetric strain at a given pressure ‘p’ and } e_L \text{ and } p_e \text{ are the model constants which are derived by best fitting a linearised form of Eq. 2 where } \frac{P}{\varepsilon_v} \text{ versus } p \text{ is plotted and have the similar meaning as of Langmuir model constants } V_L \text{ and } P_L \text{ derived for isotherm i.e. } e_L \text{ represents maximum theoretical strain at infinite pressure and } p_e \text{ is the pressure at 50% of the maximum strain.} \]

**Volumetric strain calculation**

The volumetric strain was calculated by taking the average of vertical and horizontal strains with temperature corrections applied. The following volumetric strain (\( \varepsilon \)) equation for the cylindrical samples was derived assuming isotropic swelling/shrinkage in the sample during adsorption/desorption process.

\[ \varepsilon = \frac{\Delta V}{V} = \varepsilon_a^2 + 2\varepsilon_a + \varepsilon_r^2 + 2\varepsilon_a\varepsilon_r + 2\varepsilon_a \]  
\[ \text{where } V = \text{original volume} \]
\[ \Delta V = \text{change in volume} \]
\[ \varepsilon_a = \text{axial strain} \]
\[ \varepsilon_r = \text{radial strain} \]

**Measurement of matrix swelling/ shrinkage coefficient**

Matrix shrinkage or swelling coefficient (C_m) is a measure of strain change with change in pressure. This can be derived in two ways; one is based on the pressure vs strain relationship which is similar to Langmuir isotherm shape and can be derived by differentiating the Eq. 2 w.r.t. pressure ‘p’ which is

\[ C_m = \frac{d\varepsilon}{dp} = \frac{e_L p_e}{(p + p_e)^2} \]  
\[ \text{The coefficient derived by Eq.4 has unit psi}^{-1} \text{ which quantifies the volumetric strain change per unit change in pressure.} \]

The second method is based on the relationship between gas concentration vs strain which is linear and positively correlated in all the cases and for all gases. C_m is given by the slope of the plot between gas.
volume of gas adsorbed vs volumetric strain (Figs. 4a to 4d). We represent this coefficient as $C_g$ representing gas induced volumetric swelling or shrinkage to differentiate between the two coefficients, $C_g$ must be derived for each gas separately. This coefficient has unit cc$^{-1}$ which quantifies the volumetric strain change per unit volume of gas concentration at standard conditions. Our results show that these two values differs significantly for each gas as discussed in the model calculations considered in the following sections.

MODEL DEVELOPMENT

The coal volume swelling during gas adsorption has been reported in the literature by various people [3, 4, 6, 7]. After Gray [8] quantified the effect of coal volume swelling/shrinkage on cleat permeability, a number of studies have been focused on pore pressure dependent permeability, coal matrix shrinkage and several models have been proposed [9,10,11,12,13]. Among the various models the Palmer and Mansoori model has been widely used to history match the field production [14] and laboratory permeability test data [10]. However, there are draw backs in the model which needs further modification to give more precise predictions. For example, the model is derived for coalbed methane production and does not consider the sequestration process during the ECBM production and mostly for a single gas. Further the model uses a coal matrix shrinkage coefficient ‘$C_m$’ based on the pressure vs strain relationship which in turn assumes coal matrix shrinkage no matter whether gas is desorbed or not [15] which gives an over estimation of permeability in the case of under saturated reservoirs. For example the pressure in the cleats and coal matrix are not in equilibrium during draw down of reservoir pressure during production causing over/under estimation of permeability during CBM/ECBM production. Most of the models use either the relationship between volumetric swelling/swellage being proportional to volume of gas adsorbed/desorbed or to the sorption pressure. The detailed discussion of the various models is given elsewhere [16].

Our volumetric strain experiments show that on an unit concentration (per cc adsorption) basis, compared to CH$_4$, the strongly adsorbing gases such as H$_2$S and CO$_2$ causes a greater degree of matrix swelling than the weakly adsorbing gases such as He, N$_2$ relative to CH$_4$. This effect is termed “differential swelling” relative to CH$_4$. The ARI model [15] has proposed the differential swelling coefficient. However, their model incorporates matrix shrinkage coefficient ($C_m$) based on the pressure dependence rather than the gas concentration dependence which does not apply for under saturated reservoirs. Further they were not having the data on differential swelling coefficients for different gases. As discussed in the previous sections, the matrix shrinkage coefficient derived from pressure dependence assumes shrinkage or swelling based on changes in pressure irrespective of gas desorption takes place or not.

In this paper we propose a new model for acid gas sequestration combining the Palmer and Mansoori model concept with inclusion of the differential swelling coefficients derived from our experiments.

HYDRO-MECHANIC MODEL OF COALBEDS

For non-isothermal elastic porous reservoirs, the porosity ($\phi$) change responding to the variations of stress ($\sigma_{ij}$), pore pressure (p), and temperature (T) is given by [17]

$$
\frac{\partial \phi}{\partial t} = -\frac{1}{3} \left[ \frac{1}{K} - \frac{1}{K_s} \right] \frac{\partial \sigma_{kk}}{\partial t} + \left[ \frac{1}{K} - \frac{1}{K_s} - \frac{\phi}{K_s} \right] \frac{\partial p}{\partial t} + \left[ \alpha - (1 - \phi) \alpha_s \right] \frac{\partial T}{\partial t}
$$

(5)

where $t$ is time, $K$ is the drained bulk modulus of the porous medium, $K_s$ is the bulk modulus of the solid grains, $\alpha$ and $\alpha_s$ are the coefficients of bulk thermal expansivity of the porous medium and the solid grains, respectively, and $\sigma_{kk}$ is the sum of the main diagonal terms of the stress tensor. For most
For isothermal coalbeds, the volumetric strains caused by coal matrix shrinkage upon gas desorption or swelling upon gas adsorption can be analogous to the volumetric strain due to the temperature changes for thermo-poroelastic medium [9]. Therefore, Eq. 6 can be simply modified to describe the hydro-mechanic behaviour of isothermal coal seams during gas production or gas injection.

\[
\frac{\partial \phi}{\partial t} = \zeta \left( \frac{1}{M} + \frac{1}{K_s} \right) \frac{\partial p}{\partial t} + \left[ K \left( \frac{1}{M} + \gamma \frac{1}{K_s} \right) - (1 - \phi) \right] \frac{\partial T}{\partial t} 
\]

(6)

\[
\zeta = 1 - K / K_s 
\]

(6a)

\[
M = 3K(1-\nu)/(1+\nu) = E(1-\nu)/(1-2\nu)(1+\nu) 
\]

(6b)

\[
\gamma = 2(1-2\nu)/3(1-\nu) 
\]

(6c)

where \(\zeta\) is the Biot number, \(M\) is the constrained axial modulus, and \(E\) and \(\nu\) are the Young’s modulus and Poisson ratio of the porous medium, respectively.

The subscript ‘0’ represents the initial values for porosity, pressure, and adsorbed-gas volume. Equations 8 and 8a are similar to those given by Palmer and Mansoori (1998). However, Palmer and Mansoori consider the shrinkage coefficient \(C_m\) derived from pressure vs strain curve Eq 4 as discussed previously. The above model considers the strain component \(C_g V_g\) derived from gas concentration vs strain which also satisfies the Langmuir model conditions which is evident from our laboratory experiments.

For coal seams, the permeability for gas and water flow is mainly attributed to the cleat/fracture networks. Therefore, the cleat permeability change of fractured coal coalbeds due to fluid pressure and gas adsorption or desorption can be described as [18]

\[
\frac{\kappa}{\kappa_0} = \left( \frac{\phi}{\phi_0} \right)^3
\]

(9)
where $\kappa$ and $\kappa_0$ are the new and initial cleat permeability, respectively. The porosity of the coal beds ($\phi$) is given by Eqs. 8 & 8a.

Due to well-developed fractures or cleat, coal seams are often modelled by dual-porosity/match-stick model [18], which describes the coal seams as mainly microporous coal matrixes/blocks cut by orthogonal fractures/cleats, which are the major conduits for gas and water flow in coal seams. The equations 8-9 mainly describe the cleat porosity and permeability variations. During coalbed gas production or acid gas injection, the permeability change is a function of the coal mechanical properties (i.e. Young Modulus, E and Poisson’s ratio, $\nu$), the volumetric strain caused by gas adsorption, the initial reservoir pressure ($p_0$), and initial cleat porosity ($\phi_0$). The effect of gas adsorption or desorption on permeability is only controlled by the Poisson’s ratio ($\nu$), but the effect of pressure on the cleat permeability change is controlled by both Young’s Modulus E and Poisson’s ratio $\nu$ (see Eq. 8a). Our experimental data on pure components of CH$_4$, CO$_2$, H$_2$S, and N$_2$ indicate that the volumetric strain caused by the gas adsorption or desorption is strongly, positively correlated and is a linear function of the volume of adsorbate (Figs. 4a to 4d). However, during the sequestration of acid gas into coal seams, binary or multiple gases are likely involved and desorption/or adsorption of each component happens simultaneously. Therefore, the volumetric strain caused by adsorption or desorption of each gas must be accounted for. Because of the likely linear elastic behaviour of coal seams, we here assume that total volumetric strain is simply a linear combination of the volumetric strain caused by each gas component as given in Eq 10.

$$C_{gi}V_{gi} = \sum_{i=1}^{n} C_{gi}V_{gi}$$  \hspace{1cm} (10)

where $C_{gi}V_{gi}$ is the volumetric strain caused by adsorption of gas i, determined by experimental data, and ‘n’ is the total number of gas component considered, which ranges from one to three in this study. Since the volumetric shrinkage/swelling coefficient is different for different gases, $C_{gi}$ represents the swelling coefficient for each individual gas for unit gas adsorbed.

Incorporating the parameters discussed above i.e. differential swelling coefficient and combining equations 8 and 10, our new model is represented by Eq. 11.

$$\phi - \phi_0 = \frac{1}{M} (p - p_0) + \left( C_{gi}V_{gi} - C_{g0}V_{g0} \right)$$  \hspace{1cm} (11)

Where, the term $C_{gi}V_{gi} - C_{g0}V_{g0}$ represents differential swelling/shrinkage aspect of the coal matrix due to gas adsorption/desorption of gas component i.

Multiple gas adsorptions in coals can be modelled by the extended Langmuir isotherm which is represented as:

$$V_{gi} = \frac{V_{Li}b_{Li}y_i p}{1 + p\sum_{i=1}^{n} b_{Li}y_i}$$  \hspace{1cm} (12)

where $b_{Li} (=1/p_{Li})$ and $V_{Li}$ are the Langmuir constants for gas i, $y_i$ is the mole fraction of gas ‘i’ in the gas mixture, $p$ is the gas pressure in coals. During sequestration of acidic gas into coal seams, the injected gas penetrates into the coal seams through fracture or cleat networks of coal seams, diffuses into coal matrix, and is adsorbed. At the same time, methane can also be displaced out of coals. During such processes, the gas pressure in fracture/cleat network may not be the same as that in coal matrix pores unless the
equilibrium between the cleat network and coal matrix is reached. Therefore, the gas pressure ‘p’ and composition $y_i$ in Eq. 12 should be replaced by the gas pressure $p_m$ and gas composition $y_{mi}$ within coal matrix instead of those of cleat networks when dynamic variations in cleat permeability and porosity are considered during acid gas injection.

RESULTS AND DISCUSSIONS

Volumetric changes in coal due to adsorption/desorption of $N_2$, $CH_4$, $CO_2$ and $H_2S$

Volumetric strain experiments during adsorbing $N_2$, $CH_4$, $CO_2$ and $H_2S$ were carried out for coals varying in rank from sub-bituminous ‘C’ ($R_i = 0.46\%$) to medium volatile ($R_i = 0.62\%$). Whereas desorption runs were carried out for $CH_4$ only as we were interested in quantifying the coal matrix shrinkage during CBM production. With increasing adsorption pressure of $N_2$, $CH_4$, $CO_2$ and $H_2S$ resulted in marked swelling of the coal matrix. The volumetric strain and pressure follows the trend of the isotherm i.e. similar to pressure vs gas concentration and can be modelled using a Langmuir type equation (Eq.2) for strain modelling. The modelled data fits fairly well to the experimental strain data for all the gases (Fig 3). For comparison of strain at a common base, all the strains were calculated at 0.6 MPa using Eq. 2, since the $H_2S$ experiment was done at low pressures, up to 0.6 MPa. The order of volumetric swelling observed at 0.6 MPa in descending order is $H_2S$ (1.4%-9.3%) > $CO_2$ (0.26%-0.66%) > $CH_4$ (0.09%- 0.30%) > $N_2$ (0.004% - 0.026%) (Fig. 3a; Table 2). For clarity only one sample is shown in Fig. 3. All other samples followed similar qualitative trends, however, they are quantitatively different. The volumetric strain (swelling) with $H_2S$ is about 5-20 times higher than $CO_2$ and about 15-70 times higher than $CH_4$ shrinkage. Similarly swelling caused by $CO_2$ is 2-5 times higher than the $CH_4$ Shrinkage and $CH_4$ shrinkage is 10-25 times higher than the swelling caused by the $N_2$ adsorption for the sample suite investigated (Table 3; Fig 3a). The volumetric strain is strongly and positively correlated with adsorbed gas quantities and are linearly proportional to the amount of gas adsorbed (Figs. 4a to 4d). Overall the sorption capacity and volumetric strain increases with rank for the sample suite investigated (Fig 4e), however the comparative sorption capacity and volumetric strain varies markedly with coal rank and type of gas. This trend is expected because the adsorption capacity increases with increasing coal rank. It must be noted that the sample suite of this study has a small rank range however our work is in progress on a variety of coal ranks to confirm the trend.

Comparison of gas concentrations at the same pressure i.e. at 0.6 MPa on as received basis shows that $H_2S$ adsorbs 3-6 times the quantities of $CO_2$ and 5 to 50 times as much as $CH_4$ for the sample suite (Tables 2 & 3). Comparing the volumetric strain and gas adsorption per unit basis, the volumetric strain with $H_2S$ gas injection will create up to 5 times the volumetric swelling on a unit concentration (per cc) basis when compared to $CH_4$ shrinkage and more than 4 times the swelling of $CO_2$ gas concentration. For the case of $N_2$ injection, the shrinkage caused by $CH_4$ desorption is up to 4.4 times higher than the swelling caused by the $N_2$ per unit concentration basis (Table 3). Therefore $N_2$ injection will enhance the $CH_4$ production from coalbeds due to increase in $N_2$ partial pressure coupled with increased permeability of coal beds because of shrinkage by $CH_4$ and moisture desorption from coal matrix. Pure injection of $H_2S$ or $CO_2$ may completely or significantly reduce the permeability of coal seams as injection of $H_2S$ and $CO_2$ causes swelling many times higher than the shrinkage created by $CH_4$ desorption. Our model suggests that the injection of a mixture of $N_2$ and $CO_2$ or $N_2$ and $H_2S$ may inhibit the swelling of coals and may facilitate sequestration of acid gas. Detailed discussions of model results are given below.

PERMEABILITY CHANGE DURING ACID GAS INJECTION

We have run our analytical model and subsequently verified the results with the numerical model for the Wolf Mountain coal sample. Both the models give consistent results as discussed in the following section.
First we consider the possible permeability changes in the Wolf Mountain coal during the primary CH\textsubscript{4} recovery and subsequently acid gases injection under various conditions. Figures 5a to 5d shows the possible cleat permeability change during injection of acidic gases of CO\textsubscript{2} and H\textsubscript{2}S, or flue gases into the Wolf Mountain coal. The reference values of the Young’s modulus (\(E\)), Poisson’s ratio (\(\nu\)), initial porosity (\(\phi\)), and initial pressure (\(p_0\)) in 5a are 3 GPa, 0.3, 0.18\%, and 3 MPa, respectively. However, the reference initial cleat porosity in Fig. 5c is 1\%.

As expected from Eq. 11, the permeability variation relative to its initial values systematically decreases with the increase in Young’s modulus because coal seams with larger \(E\) can be stronger and hence less sensitive to variation in fluid pressure (Fig. 5a). Decrease in Poisson’s ratio also systematically enhances the variation of the cleat permeability when gas pressure is elevated or reduced. Because of the relatively high CH\textsubscript{4} adsorption capacity of this coal (\(V_L = 14 \text{ cc/g}; \) Table 2), the CH\textsubscript{4} desorption from the coal seam with an initial pressure of 3 MPa can cause strong permeability rebounds at relatively low pressures (Fig. 5a). Injection of CO\textsubscript{2} into this coal will make coal impermeable if the coal has an initial porosity of about 0.18\% or less. Elevating the gas pressure by twice its initial value causes no permeability recovery (Fig. 5b). Even with a large initial porosity of 0.5\%, the permeability does not show any recovery. Injection of H\textsubscript{2}S is likely to make the Wolf Mountain coal impermeable if the coal has an initial porosity less than 5\% due to strong swelling effects of H\textsubscript{2}S. About 80\% permeability reduction will be induced for coals with large Poisson’s ratio \(\nu=0.4\) (Fig. 5c). The reduced permeability does not rebound back when the pressure is elevated higher than its initial values after H\textsubscript{2}S displaces all the pre-adsorbed CH\textsubscript{4}. However, injection of N\textsubscript{2} into the Wolf Mountain coal enhances the cleat permeability by more than 5 times because of the small adsorption capacity and low adsorption affinity of N\textsubscript{2} to coal and displacement of pre-adsorbed CH\textsubscript{4} (Fig. 5d). However, the permeability enhancement is limited for coals with large initial porosity of 0.5\%.

Because of the strong enhancement of cleat permeability by injection of N\textsubscript{2}, the complete permeability loss during injection of CO\textsubscript{2} and H\textsubscript{2}S into the low cleat porosity coals may be avoided by injection of flue gases. Indeed, injection of a flue gas of 20\%CO\textsubscript{2} and 80\%N\textsubscript{2} only reduces the cleat permeability by 60\% for coals with initial porosity 0.5\% (Fig. 5b) and injection of a flue gas of 20\%H\textsubscript{2}S and 80\%N\textsubscript{2} will cause 90\% reduction of the cleat permeability (Fig. 5c).

**NUMERICAL MODELLING**

To verify the simple analytic results presented in the preceding section, we developed a two-dimensional finite difference model that can rigorously simulate the dynamic changes in permeability during acid/flue gas injection into the example coals. The governing equations solved include the mass conservation equations of water and gas in coal-seam cleats and coal matrix. Only the Darcy’s flow of water and gas in cleat networks are considered and gas diffusion between cleat and coal matrix is approximated by linear-driven force model [19, 20]. The diffusion coefficients of gases in coal matrix are determined by our adsorption-rate experimental data. Gas and water relative permeability relation is taken from Gash [21]. The model has been tested against the results provided by other models [22]. The results from our models are consistent with results predicted by those different simulators.

Here we only give a few typical modelling cases to demonstrate how permeability varies during acid/flue gas injection into the Wolf Mountain coals, which initially contains only CH\textsubscript{4} and 65\% water in cleats. During simulations, a 5-spot well pattern is applied (Fig.6), but the simulated part is only one quarter of the whole domain with a rectangular mesh of 21 by 21. The dimension of the simulated domain is 200 x 200 x 5 m. For simplicity, all properties, including fluid pressure, gas concentration and others, are assumed to be homogeneous and the injection well and/or production well fully penetrate the coal seam. Influences of gravity on gas and water flow are neglected because of the relatively small thickness and
large horizontal dimensions of the coal seams. The effects of temperature and moisture are not considered here. Other parameters are given Table 4.

Modelling results of injection of 99%CO₂ + 1%N₂ into the Wolf Mountain coals (the reference model in Fig. 5b) are partly shown in Fig. 7a. The CO₂ injection rate is significantly reduced from 10,000 to ~10 m³/day in about 10 days because of dramatic decrease in the permeability caused by the CO₂-adsorption. Since the swelling caused by CO₂ is significantly higher than the shrinkage caused by the CH₄ desorption, the production rate of CH₄ is reduced significantly (Fig. 7a & 7d). The bottom-hole pressures of both injection and production wells are constant at 6 and 0.5 MPa, respectively (Fig 7b). Due to the strong CO₂ adsorption, the low injection rate, and the large cleat spacing of the coal seam (1 cm), CO₂ breaks through after 4 years but with very low concentration. However, the N₂ break through is seen in very early stages (Fig. 7c). The permeability of the coal seam at different location changes dynamically with injection of CO₂. The permeability of coals near the injection well firstly is enhanced marginally because of the elevated cleat pressure by CO₂ injection accompanies by minor adsorption of CO₂ at the early stages. Thereafter, their permeability declines as the coals adsorb more CO₂ until reaching the maximum reduction when coal is saturated with CO₂ (Fig. 7d). The permeability of coals near the production well, however, undergoes enhancement with time because of the CH₄ desorption with a decrease in pressure and essentially no adsorption of CO₂.

Injection of flue gas (20%CO₂+80%N₂) into the same coals avoids the significant reduction in the injection rate because of the much lower decline in permeability (Fig. 8). Also the methane recovery is significantly enhanced with a peak production rate that is about two times that for the injection of 99% CO₂ (compare Fig. 7a and 8a). With injection of flue gas, the N₂ break through is significant and occurs at very early stages. However CO₂ breakthrough occurs very late and in small volumes (Fig. 8c). The permeability of coals close to the injection well increases 1.5 times because of the elevation of cleat pressure and then decrease with adsorption of CO₂, but it only decreases to half of its initial values (Fig. 8d). The permeability of coals close to the production well increases with time at early stage because of desorption of CH₄ but begins to decline at later stages because of adsorption of CO₂ (Fig. 8d).

As shown in figure 5c, injection of H₂S into the Wolf Mountain coals will result in total loss of permeability if the initial porosity is less than 1%. Here we simulate the injection of H₂S into the coals with an initial cleat porosity of 5% and a cleat spacing of 2 mm and other parameters are the same as shown in Fig. 5c. Because of the strong swelling effects of H₂S, the injection of pure H₂S into the coals causes nearly complete loss of permeability of the injection well block in about a week (Fig. 9b), but the rest of the area is not affected because of the very limited H₂S injected (Fig. 9a). Fluctuations observed in the H₂S injection rate after reaching its lowest level of injection (in Fig. 9a) is scale effect and the injection rate fluctuates between 0.5 m³/d to 0.1m³/day, which is almost no injection rate. Recovery of CH₄ is mainly primary. Injection of flue gas (20%H₂S + 80%N₂) causes a smaller decrease in permeability of the injection well block but still results in 90% loss of permeability (Fig. 9d) in 60 days and thus low injection rate of the flue gas (Fig. 9c).

CONCLUSIONS

Our volumetric strain experiments with various gases provide critical information on the coal reservoir behaviour when acid gas is injected for sequestration or for ECBM purposes. The volumetric swelling/shrinkage is strongly and positively correlated with gas concentration in all coals and for all gases. The volumetric strain related to pressure can be reasonably modelled using the Langmuir type model equation similar to isotherm modelling. The parameters derived from modelling the strain can be
used for interpreting the strain caused as a result of pressure change in the reservoir and also the change in gas concentration using the relationship between volumetric strain vs gas concentration.

The order of volumetric swelling at 0.6 MPa in decreasing order is H₂S (1.4%-9.3%) > CO₂ (0.26%-0.66%) > CH₄ (0.09%-0.30%) > N₂ (0.004% - 0.026%). On unit concentration basis, the CH₄ desorption creates a significant volumetric shrinkage of coal which is 1.4 to 4.4 times higher than the volumetric swelling created by the N₂ adsorption. Therefore N₂ injection will enhance the CH₄ production from coalbeds due to an increase in N₂ partial pressure coupled with increased permeability of coalbeds because of shrinkage by CH₄ desorption. Similarly the injection of pure H₂S will create significant swelling effect on a unit concentration basis which is about 5 times higher than CH₄ desorption shrinkage and CO₂ injection will create a swelling effect of 2.2 times higher than the shrinkage effect created by the CH₄ desorption. Thus injection of these two pure gases will reduce the permeability of coalbeds significantly and can be critical to the CH₄ production.

Our analytical and numerical model simulation results show that the injection of acid gases (CO₂ or H₂S) will result in total loss of permeability in the Wolf Mountain coal because the coal has low initial cleat porosity (<<1%). Permeability reduction is less marked for coals with larger cleat spacings because of higher initial cleat porosity for a fixed permeability. Coals which have low cleat porosity, the permeability decline with coal swelling is more marked. Mixing of N₂ with the acid gas can alleviate the permeability loss induced by injection of pure acid gas. Also injection of the flue gas causes the permeability reduction at later stages than injection of pure acid gas or carbon dioxide.

Our numerical model can be easily applied to estimate the feasibility for acid gas sequestration into various coals. Overall an optimum coal seam for sequestration of acid gases would have to have a high initial permeability and most importantly a large initial cleat porosity to reduce the effects of permeability reduction induced by coal swelling on the efficiency of the acid gas sequestration.

REFERENCES CITED


### Table 1: Composition of coal samples for strain experiments

<table>
<thead>
<tr>
<th>Samples</th>
<th>Rank &amp; Proximate analysis</th>
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<th></th>
<th></th>
</tr>
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<tbody>
<tr>
<td></td>
<td>Vitritine reflectance (Rv %)</td>
<td>Moisture (%)</td>
<td>Ash (%)</td>
<td></td>
</tr>
<tr>
<td>Ardley</td>
<td>0.46</td>
<td>8.04</td>
<td>32.8</td>
<td></td>
</tr>
<tr>
<td>Illinois</td>
<td>0.50</td>
<td>5.48</td>
<td>10.11</td>
<td></td>
</tr>
<tr>
<td>Quinsam</td>
<td>0.62</td>
<td>4.14</td>
<td>9.30</td>
<td></td>
</tr>
<tr>
<td>Wolf Mountain</td>
<td>0.62</td>
<td>2.26</td>
<td>5.05</td>
<td></td>
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### Table 2: Langmuir model constants for strain and isotherm experiments for the studied gases on solid core samples and reported as received basis

<table>
<thead>
<tr>
<th>Sample id</th>
<th>Gas type</th>
<th>L $\varepsilon$</th>
<th>$L p$</th>
<th>$L V$</th>
<th>$L P$</th>
<th>Cm/psi From pressure component</th>
<th>Cm/cc From volume component</th>
<th>Volumes adsorbed at 0.6Mpa (cc/g, as received basis)</th>
<th>Vol strain at 0.6 MPa with Pressure component</th>
<th>Different -ial swelling</th>
</tr>
</thead>
<tbody>
<tr>
<td>Wolf Mountain</td>
<td>H2S</td>
<td>18.05%</td>
<td>0.56</td>
<td>73.62</td>
<td>0.67</td>
<td>2.2E-03</td>
<td>2.7E-03</td>
<td>34.67</td>
<td>9.327%</td>
<td>2.1E-03</td>
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<tr>
<td>Quinsam</td>
<td>H2S</td>
<td>14.42%</td>
<td>0.43</td>
<td>82.59</td>
<td>0.91</td>
<td>2.3E-03</td>
<td>2.5E-03</td>
<td>32.84</td>
<td>8.378%</td>
<td>1.5E-03</td>
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<tr>
<td>Illinois</td>
<td>H2S</td>
<td>19.79%</td>
<td>1.18</td>
<td>48.27</td>
<td>1.47</td>
<td>1.2E-03</td>
<td>4.6E-03</td>
<td>13.99</td>
<td>6.676%</td>
<td>3.7E-03</td>
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<tr>
<td>Ardley</td>
<td>H2S</td>
<td>1.45%</td>
<td>0.02</td>
<td>32.00</td>
<td>0.27</td>
<td>4.3E-03</td>
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<td>1.402%</td>
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<tr>
<td>Wolf Mountain</td>
<td>CO2</td>
<td>6.74%</td>
<td>8.04</td>
<td>27.92</td>
<td>1.76</td>
<td>5.8E-05</td>
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<td>0.468%</td>
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<td>CO2</td>
<td>5.90%</td>
<td>4.80</td>
<td>38.84</td>
<td>2.06</td>
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<td>CH4</td>
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<td>CH4</td>
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<td>5.31</td>
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<td>0.41</td>
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<tr>
<td>Wolf Mountain</td>
<td>N2</td>
<td>0.42%</td>
<td>9.02</td>
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<td>14.02</td>
<td>3.2E-06</td>
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<td>0.59</td>
<td>0.026%</td>
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<td>19.86</td>
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<td>N2</td>
<td>0.09%</td>
<td>6.85</td>
<td>8.34</td>
<td>17.09</td>
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### Table 3: Ratios of adsorbed quantities and strains at 0.6 MPa as received basis

<table>
<thead>
<tr>
<th>Samples</th>
<th>Ratios of gas quantities adsorbed at 0.6 MPa</th>
<th>Ratios of volumetric strains at 0.6 MPa</th>
<th>Ratios of $C_m$/cc coefficient</th>
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<tr>
<td></td>
<td>$H_2S/CH_4$</td>
<td>$H_2S/CH_4$</td>
<td>$H_2S/CH_4$</td>
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<tr>
<td>Wolf Mountain</td>
<td>8.93</td>
<td>31.41</td>
<td>4.6</td>
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<td>9.51</td>
<td>68.00</td>
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<tr>
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<td>5.29</td>
<td>66.55</td>
<td>4.8</td>
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<tr>
<td>Ardley</td>
<td>53.06</td>
<td>15.34</td>
<td>1.3</td>
</tr>
<tr>
<td></td>
<td>$CO_2/CH_4$</td>
<td>$CO_2/CH_4$</td>
<td>$CO_2/CH_4$</td>
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<tr>
<td>Wolf Mountain</td>
<td>1.83</td>
<td>1.58</td>
<td>2.2</td>
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<td>2.53</td>
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<td>2.01</td>
<td>4.06</td>
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<tr>
<td>Ardley</td>
<td>9.45</td>
<td>2.86</td>
<td>1.9</td>
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<tr>
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<td>$N_2/CH_4$</td>
<td>$N_2/CH_4$</td>
<td>$CH_4/N_2$</td>
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<tr>
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<td>0.15</td>
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<tr>
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### Table 4: Parameters for all simulation cases*

<table>
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<tr>
<th>parameter</th>
<th>values</th>
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<tr>
<td>Initial reservoir pressure (MPa)</td>
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<tr>
<td>Maximum injection rate (full well, m³/day)</td>
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</tr>
<tr>
<td>Maximum injection well bottom-hole pressure (MPa)</td>
<td>6</td>
</tr>
<tr>
<td>Minimum production well bottom-hole pressure (MPa)</td>
<td>0.05</td>
</tr>
<tr>
<td>Initial cleat permeability ($k_0$, millidarcy)</td>
<td>5</td>
</tr>
<tr>
<td>Coal matrix porosity (%)</td>
<td>7</td>
</tr>
<tr>
<td>Coal density (g/cc)</td>
<td>1.34</td>
</tr>
<tr>
<td>Temperature (°C)</td>
<td>25</td>
</tr>
<tr>
<td>Coal seam thickness (m)</td>
<td>5</td>
</tr>
<tr>
<td>Well-radius (cm)</td>
<td>4</td>
</tr>
<tr>
<td>Initial water saturation in cleats</td>
<td>0.65</td>
</tr>
<tr>
<td>Initial gas in cleats and coal matrix (% CH₄)</td>
<td>100</td>
</tr>
</tbody>
</table>

* For CO₂-related simulations, the cleat spacing of coal seam is assumed to be 2 mm and the cleat porosity (0.49%) is determined with the match stick model; for H₂S related simulations, the coal seams is assumed to have a initial cleat porosity of 1% and a cleat spacing 1 mm.
**Figure 1:** Schematic layout of volumetric adsorption-strain experimental set up

**Figure 2:** Strain gauge attachment to the core sample. The core shown in the picture is 3” dia and 4.5” length Wolf Mountain coal.
Figure 3: Wolf Mountain volumetric strain with pressure. Experimental values are shown with symbols on the plot and the continuous lines shows the model fit using the Eq. 2. In case of H$_2$S the scale is to be multiplied by 5.

Figure 3a: This is X-axis scale enlarged for Figure 3 for comparing the strains at 0.60 MPa.
Figure 4 (a)-(d): Relationship between volumetric strain vs gas concentration for (a) H$_2$S, (b) CO$_2$, (c) CH$_4$ and (d) N$_2$. As can be seen the relationship between volumetric strain and volume of gas adsorbed for different gases show strong correlation with $r^2$ value mostly more than 0.90.
Figure 4e: Trend of volumetric strain with coal rank at 0.6 MPa. Even though the rank suite investigated is small range (i.e. 0.46% to 0.62% $R_0$%), follows the trend of gas adsorption capacities with coal rank suggesting the possible trend could be true. This trend needs to be confirmed using high rank coal volumetric strain experiments.
Figures 5a-5d: The possible permeability variation of the Wolf Mountain coals during CBM primary recovery or enhanced recovery by injection of CO₂, H₂S or N₂. The reference values of the Young’s modulus ($E$), Poisson’s ratio ($\nu$), initial porosity ($\phi_0$), and initial pressure ($p_0$) are 3 GPa, 0.3, 0.18%, and 3 MPa, respectively. However, the reference initial cleat porosity in $c$ is 1%. Flue gas injection is labelled out by the fraction of N₂. For cases considering gas injection to displace CH₄ (b-d), it is assumed that the pressure is the partial pressure of injected gas while the coal seam pressure is held at constant of initial pressure $p_0$. When the pressure becomes higher than the initial pressure, CH₄ is completely displaced and then the pressure is the total pressure of the coal seam. Higher pressure implies that more gas is injected into coals.
Figure 6: The five-spot pattern and the finite difference block applied in numerical simulates. Symbol I represents an injection well and symbol P represent the production wells.

Figure 7: Simulation results of CO₂ injection into Wolf Mountain coals. (a) gas injection and production rate; (b) Injection (i) and production (p) gas well bottom-hole pressure (i\textsubscript{bh}) and well block cleat pressure (i); (c) production gas composition; and (d) permeability variation ratio of coals at different locations along the diagonal of the I-P line in Fig. 6.
Figure 8: Simulation results of 20%CO₂ +80%N₂ injection into Wolf Mountain coals. (a) gas injection and production rate; (b) Injection (i) and production (p) gas well bottom-hole pressure (\(P_{bh}\)) and well block cleat pressure (\(P_b\)); (c) production gas composition; and (d) permeability variation of coals at different locations along the diagonal of the I-P line in Fig. 6.
Figure 9: Injection of H\textsubscript{2}S into Wolf Mountain coal. (a) H\textsubscript{2}S injection and CH\textsubscript{4} production rate; (b) permeability variation of coals at different locations along the diagonal of the I-P line in Fig. 6; (c) flue gas injection and coalbed gas production rate; (d) permeability variation of coals at different locations along the diagonal of the I-P line in Fig. 6.