History matching of enhanced coal bed methane laboratory core flood tests
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Abstract
Enhanced coalbed methane (ECBM) involves the injection of a gas, such as nitrogen or carbon dioxide, into the coal reservoir to displace the methane present. Potentially this strategy can offer greater recovery of the coal seam methane and higher rates of recovery due to pressure maintenance of the reservoir. While reservoir simulation forms an important part of the planning and assessment of ECBM, a key question is the accuracy of existing approaches to characterising and representing the gas migration process. Laboratory core flooding allows the gas displacement process to be investigated on intact coal core samples under conditions analogous to those in the reservoir. In this paper a series of enhanced drainage core floods are presented and history matched using an established coal seam gas reservoir simulator, SIMED II. The core floods were performed at two pore pressures, 2 MPa and 10 MPa, and involve either nitrogen or flue gas (90% nitrogen and 10% CO\textsubscript{2}) flooding of core samples initially saturated with methane. At the end of the nitrogen floods the core flood was reversed by flooding with methane to investigate the potential for hysteresis in the gas displacement process. Prior to the core flooding an independent characterisation program was performed on the core sample where the adsorption isotherm, swelling with gas adsorption, cleat compressibility and geomechanical properties were measured. This information was used in the history matching of the core floods; the properties adjusted in the history matching were related to the affect of sorption strain on coal permeability and the transfer of gas between cleat and matrix. Excellent agreement was obtained between simulated and observed gas rates, breakthrough times and total mass balances for the nitrogen/methane floods. It was found that a triple porosity model improved the agreement with observed gas migration over the standard dual porosity Warren-Root model. The Connell, Lu and Pan hydrostatic permeability model was used in the simulations and improved history match results by representing the contrast between pore and bulk sorption strains for the 10 MPa cases but this effect was not apparent for the 2 MPa cases. There were significant differences between the simulations and observations for CO\textsubscript{2} flow rates and mass balances for the flue gas core floods. A possible explanation for these results could be that there may be inaccuracy in the representation of mixed gas adsorption using the extended Langmuir adsorption model.
1. Introduction

Coal seams are dual porosity reservoirs with a natural fracture system, known as cleats, forming a macroporosity and the intervening coal matrix, the microporosity. The main storage mechanism for gas in coal seam reservoirs is adsorption within the coal matrix with the quantity adsorbed a function of the gas species pressure. Gas drainage from coal reservoirs is commonly performed to establish safe conditions for underground mining, recover coalbed methane (CBM) for energy, and/or reduce fugitive emissions from coal mining. Conventional gas drainage involves drilling wells into the target coal seam and lowering seam pressure, typically through water production, leading to gas desorption and drainage from the seam. The recovery of gas is limited by the degree to which the seam pressure can be lowered. This is determined by a range of factors including reservoir properties such as permeability and operational considerations such as well spacing and drainage lead time. However, with conventional (primary) recovery large volumes of residual gas can be expected to remain in the seam.

Another gas drainage strategy is enhanced coalbed methane (ECBM) which involves the injection of a gas, such as nitrogen or carbon dioxide, into the coal reservoir (Puri and Yee, 1990; Reeves and Oudinot, 2004). The injected gas displaces the reservoir gases from the coal cleat system creating a partial pressure gradient between the cleat and coal matrix. Injected gas migrates into the coal matrix and becomes adsorbed while reservoir gas migrates to the cleat system from the matrix. Since the continued flow of injected gas keeps the cleat partial pressure of reservoir gas low, there is the potential for more complete recovery of the reservoir gas than that through pressure drawdown. ECBM also acts to sustain gas drainage rates as the injected gas maintains reservoir pressure. Furthermore, if CO$_2$ is used as the injectant the process can also serve as a greenhouse gas mitigation strategy as CO$_2$ can be stored in the coal while enhancing gas recovery (Mavor et al., 2004).

While ECBM has received considerable attention for gas production its application to coal mining would have different objectives. In particular CO$_2$ storage in coal seams to be mined cannot be considered a benefit as the gas would be released upon mining. In addition CO$_2$ rich coals often represent a significant mining hazard due to high gas contents and other effects. The benefits of enhanced drainage for mining over primary drainage relate to the reduction of residual gas at mining and higher rates of gas drainage due to pressure maintenance of the reservoir. For open cut mining where coal seams are shallow and pore pressures low, enhanced drainage could help to sustain drainage rates and reduce fugitive emissions.
during mining. The potential injectant gases for enhanced gas drainage for mining are nitrogen and for open cut drainage, nitrogen or flue gas, which is primarily nitrogen but with approximately 10% CO₂. Sander et al. (2010) presents an investigation into enhanced drainage for coal mining which combines reservoir simulation with economics to determine the potential role this strategy could play in managing fugitive emissions from open cut coal mines. Packham et al. (2010) presents a simulation study of the application of enhanced drainage for coal mine gas management.

A range of properties and operational conditions affect the enhanced drainage process. Key reservoir properties include the permeability, the gas adsorption characteristics and the properties which determine the rate of exchange of gases between cleat and matrix. An important factor is the sweep efficiency. Sweep efficiency relates to the proportion of the seam gas displaced by the injected gas and reflects the nature of the flow path through the reservoir. For example, a reservoir with a sparse but highly conductive fracture system is likely to have a low sweep efficiency compared to a uniform formation undergoing porous media flow. Sweep efficiency also influences the time to injectant gas breakthrough as it in part determines the flow velocity within the coal seam.

Core flooding experiments offer a means of observing the gas displacement process of enhanced recovery in the laboratory. These experiments involve placing a coal sample in a pressure cell and establishing an initial methane content by holding a methane pressure until adsorption has equilibrated. Gas, for example CO₂, is then injected at one end of the sample and outflow allowed to occur at the opposite end via a backpressure regulator. Monitoring gas rates and composition provides information on the enhanced gas drainage process. Fulton et al. (1980) and Reznik et al. (1984) present results from CO₂ floods of coal core initially saturated with methane and observed the potential for CO₂ to effectively displace the methane present. In Mazumder and Wolf (2008) CO₂ and flue gas core flood experiments on coal samples are presented. In Wolf et al. (2000) CO₂ ECBM core floods with Belgian and German coal samples were history matched using the STARS compositional reservoir simulator. In contrast to existing approaches to describing diffusive exchange between cleat and matrix which typically use the Warren and Root (1963) unipore model, Shi and Durucan (2003) applied a bidisperse pore diffusion model to the Wolf et al. (2000) core flooding results. In order to achieve a good history match Shi and Durucan used a linear relationship between the diffusivity and the total sorbate concentration. Jessen et al. (2008) history matched core flooding observations from coal that had been ground and reconstituted.
Since the coal particles were relatively small it was reasoned that diffusive exchange between primary and secondary porosities was not important.

This paper presents a series of core flooding experiments and their simulation designed to investigate the enhanced drainage process. The focus is on enhanced drainage relevant to mining conditions and as such the injectant gases used were nitrogen and flue gas (90% nitrogen 10% CO₂ was used). A key objective of the work is testing of the theory used in reservoir simulation of gas migration during enhanced drainage in particular the dual porosity diffusional exchange. Another aspect of interest in the work is characterisation of the sweep efficiency in the core flooding experiments. The simulator used for this study is a modified version of the commercial reservoir simulation program SIMED II (Stevenson and Pinzcewski, 1996).

2. Experimental methodology

Core flooding experiments were performed in a tri-axial cell as outlined in Figure 1. This arrangement enabled core samples to be tested at reservoir pore pressures and temperatures with confining pressures representative of the average stress under hydrostatic conditions. The radial strain was monitored using strain gauges. The confining fluid used in the tri-axial cell was water and the sample was jacketed with a thin lead foil and then a viton rubber membrane. Teledyne ISCO 500D syringe pumps were used to control the confining pressure and the gas inflow rates with two pumps used for the flue gas studies involving nitrogen and carbon dioxide injection. The pore pressure was maintained during the core floods using a back pressure control device and the gas rate measured using a Ritter Apparatebau MilliGascounter appropriate for the flow rates associated with the tests. Outflow gas composition was measured using a Shimadzu GC/MS QP2010. The core flooding pressure vessel and associated pipework is housed in a temperature controlled cabinet allowing tests to be performed at reservoir temperature. All flow rates and gas volumes presented in this paper are at standard conditions of 15.6 Celsius and 101.3 kPa.

The test procedure comprised the following steps;
1. Vacuuming of sample to remove residual gas.
2. Adsorption of gas (methane) by maintaining the pore pressure in the core sample and allowing gas adsorption to equilibrate (indicated by CH₄ injection rate equal to zero).
3. Injection of gas (nitrogen or flue gas) into one end of the sample and the control of a constant pore pressure using a back pressure regulator. The inflow and outflow gas rates were monitored and the gas composition measured using a GC/MS. The experiment is terminated when steady state concentration conditions are achieved, that is outflow concentration and rate is equal to inflow.

The core sample used in the core flooding program described in this paper is from the Bowen Basin, Australia. The physical characteristics of the coal sample is presented in Table 1. Prior to the core flooding experiments a series of measurements were performed on the core samples in a separate apparatus to determine the adsorption isotherm and swelling characteristics (described in the following section below).

The core flooding comprised a sequence of gas displacement experiments at pore pressures of 2 and 10 MPa, with confining pressures of 4 MPa and 12 MPa respectively, at a temperature of approximately 35°C to be representative of reservoir conditions. Two types of experiments are performed; binary flow experiments using CH$_4$ and N$_2$ and ternary experiments using a gas mixture (90% N$_2$, 10% CO$_2$), to replicate a combustion flue gas, to displace CH$_4$. For the CH$_4$/N$_2$ floods a core flood is then performed where CH$_4$ is the injectant displacing the N$_2$ in the core, so that initial methane saturated conditions are restored in order to investigate the reversibility of the displacement process. For the other core floods, at the end of the flood the core is vacuumed to remove gas.

For the Bowen Basin sample the following core floods were performed;

Binary experiments: CH$_4$ / N$_2$ floods
- 2 MPa
  - N$_2$ displacing CH$_4$
  - CH$_4$ displacing N$_2$
- 10 MPa
  - N$_2$ displacing CH$_4$
  - CH$_4$ displacing N$_2$

Ternary experiments: CH$_4$ / flue gas (90% N$_2$, 10% CO$_2$) flood
- 2 MPa
  - Flue gas displacing CH$_4$
• Flue gas displacing CH₄ (repeat)

- 10 MPa
  • Flue gas displacing CH₄

3. Core characterisation

Prior to the core floods the adsorption isotherm, swelling characteristics, geomechanical properties and cleat compressibility were measured for the core samples, in a separate triaxial rig to that used for the core flooding, following the procedure described by Pan et al. (2010). The adsorption measurements and Langmuir isotherm best fit relations for the three gases of interest are presented in Figure 3 and the measured sorption strains are presented in Figure 4. These sorption strains are corrected to remove the effects of pore pressure and confining pressure induced compression using the geomechanical properties measured from the core samples and presented in Table 2.

4. Model description

4.1 Coal permeability behaviour

SIMED II represents the various flow processes operating in coal seam reservoirs being two-phase, compositional, and including dual porosity gas exchange and storage from adsorption. It also represents the permeability behaviour important to coal seam reservoirs of sensitivity to effective stress which is counterbalanced by matrix shrinkage due to gas desorption, that is, the sorption strain. SIMED uses various modelling approaches to describing this permeability behaviour including the Shi-Durucan model (Shi and Durucan, 2005). However the Shi-Durucan model is designed for reservoir conditions and is based on assumptions of uniaxial strain and constant vertical stress. The core flooding tests presented here used hydrostatic stress and strain conditions where the sample can undergo tri-axial strain while the confining pressure is uniform around the sample. To allow for these test conditions the hydrostatic permeability model presented by Connell et al. (2010) was implemented in SIMED II. This model differentiates between the bulk and pore strains due to gas desorption and can be written as;

\[
k = k_0 \exp \left\{ -3 \left[ C_{pc}^{(M)} \left( \overline{\sigma}_c - \overline{p}_p \right) + \left( \overline{\sigma}_p^{(S)} - \overline{\sigma}_b^{(S)} \right) \right] \right\}
\]

Where \( k \) is the permeability and \( k_0 \) is the permeability at reference pore and confining pressures, \( C_{pc}^{(M)} \) is the compressibility, \( \overline{\sigma}_c \) and \( \overline{p}_p \) are the change in confining pressure and pore pressure, \( \overline{\sigma}_p^{(S)} \) and \( \overline{\sigma}_b^{(S)} \) are...
the change in pore and bulk sorption strains. As an approximation Connell et al. (2010) proposed that the pore sorption strain could be related to the bulk sorption strain by the following:

$$\varepsilon_{p}^{(S)} = \gamma \varepsilon_{b}^{(S)}$$

(2)

where $\gamma$ is a constant. Substituting Eq. (2) into Eq. (1) leads to,

$$k = k_0 \exp \left\{ -3 \left[ C_p^{(M)} (\bar{p}_c - \bar{p}_p) + (\gamma - 1) \varepsilon_{b}^{(S)} \right] \right\}$$

(3)

The numerical model used in this study applies Eq. (3) to model the behaviour of permeability in the core flooding rig with $\gamma$ determined through history matching. If pore and bulk strains are equal then, under hydrostatic conditions, sorption strain would have no impact on coal permeability. In the core flooding experiments there may be changes in the total gas content as one gas is replaced by another and as a result there could be sorption strain. For example for the same pore pressure methane will have a significantly higher gas content than nitrogen, and so in core floods where nitrogen displaces methane the total gas content will decrease and the core sample will shrink.

### 4.2 Fracture-matrix exchange

Coal is often assumed to have a dual porosity structure where gas diffuses between the cleat macro-porosity and the matrix micro-porosity where it is adsorbed. In SIMED II these diffusion processes are represented using the Warren-Root model (Warren and Root, 1963) which is based on a pseudo-steady state assumption for adsorption within the micro-porosity. This model is applied to each of the individual gas species in a multi-component simulation and can be written as;

$$\frac{d\bar{C}_i}{dt} = \frac{C_i - \bar{C}_i}{\tau_i},$$

(4)

where $\bar{C}_i$ is the average gas content within the matrix block of species $i$, and $C_i$ denotes the adsorbed gas content at the cleat partial pressure of species $i$. The characteristic time for species $i$, $\tau_i$, known as the sorption time, can be written as,
\[
\tau_i = \frac{l}{\sigma D_M^i}
\]  

(5)

in which \( \sigma \) is a shape factor that combines the size and shape of the matrix blocks and \( D_M^i \) is the effective diffusion coefficient of species \( i \). As noted by Lu and Connell (2007a) applying Eq. (4) to mixed gas migration is an approximation since it neglects multi-component diffusion. They went on to present a multi-component diffusion model for dual porosity. Another approximation is that Eq. (4) can be inaccurate at early times (Lu and Connell, 2007b).

Coal seams may have more complex structures than the dual porosity accounted for with Eq. (4). Several studies have found that a bidisperse diffusion model may provide a better representation than the unipore approach of Eq. (4) (Smith and Williams 1984; Clarkson and Bustin, 1999; Shi and Durucan, 2003; Cui et al., 2004; Pan et al., 2010). In the bidisperse model the microporosity is divided into two regions; gas diffuses through these two regions to be adsorbed. Busch et al. (2004) present an approach where the quantity of gas adsorbed is divided into two regions with different rates of diffusion. This was found to provide a more accurate description of the diffusion process than Eq. (4).

In this paper, in addition to Eq. (4), a new simulation approach is introduced to describe the matrix-cleat exchange of gas, based on that presented by Busch et al. (2004). With this it is assumed that the adsorbed gas is within two regions of the matrix which have different sorption times. Thus there are three distinct porosities within the coal; the cleat system and two regions in the microporosity. A simple extension to this triple porosity system can be derived using the Warren-Root model by assuming that the matrix is composed of two separate regions of porosity with differing sorption times. With this the equilibrium adsorbed quantity remains the same but is partitioned between the two micro-porosities. This can be written as,

\[
\frac{d\bar{C}_{1i}}{dt} = \frac{\phi_1 C^e_i - \bar{C}_{1i}}{\tau_{1i}}, \quad \frac{d\bar{C}_{2i}}{dt} = \frac{\phi_2 C^e_i - \bar{C}_{2i}}{\tau_{2i}},
\]

(6)

where \( \phi_1 \) is the fraction of the equilibrium adsorbed gas content associated with sorption time \( \tau_{1i} \) and \( \phi_2 \) with \( \tau_{2i} \). \( \bar{C}_{1i} \) and \( \bar{C}_{2i} \) are the gas contents for each microporosity fraction.
4.3 Model construction

The model grid was constructed to match the core dimensions presented in Table 1. While the core is of cylindrical geometry in the simulations it is represented as a rectangular cuboid. Dead space present within the tubing network of the experimental equipment connecting to the core had to be accounted for in the model since this influenced the measurements of gas migration during the experiments. The core is represented by a grid of 1 x 1 x 90 blocks of which there were regions of 20 blocks each at both ends of the grid used to represent the tubing void volume with the remaining 50 blocks for the core itself (see Figure 2). At the upstream (injection) end of the model grid there was 14.35 ml of dead space and 30 ml on the downstream (production) end.

4.4 Procedure used to simulate the core floods

The gas injection rate and outflow pressure measured during the core floods were used as controlling parameters for the simulation. Gas outflow rates were matched through adjustment of the model properties involved in the diffusive exchange of gas between coal cleat and matrix. Upstream (i.e. injection) pressure was matched through tuning of the initial core permeability and the $\gamma$ factor in Eq. (4) which determines the influence of sorption strain on permeability via Eq. (3).

5. Core Flood Simulation Results

5.1 Binary Displacement - CH$_4$/N$_2$ floods

Comparisons of simulated and experimental outflow rates and upstream pressures for the CH$_4$/N$_2$ core flooding experiments at 2 MPa pore pressure are presented in Figure 5. The mass balance over each core flood was calculated from the cumulative inflow and outflow measurements allowing the quantity of gas within the core sample to be determined. These are presented in Table 3 in the form of gas contents at the start and end of each experiment, along with those derived from the simulations. The optimal properties from the history matching of the simulation model are presented in Table 4. The very close agreement between observations and simulation results indicate that the simulation model adequately describes the flow behaviour in the core. Similar behaviour was found with the 10 MPa experiments which are presented in Figure 6, with the optimal properties from the history match also presented in Table 3. For the N$_2$ flood at 10 MPa there was excellent agreement between measured and simulated outflow rates and with the upstream pressures. The agreement is not as close with the CH$_4$ flood where
the simulation results did not reproduce some of the initial N\textsubscript{2} outflow behaviour however the key feature of CH\textsubscript{4} breakthrough was reliably represented.

Even though the gas injection rates were higher for the CH\textsubscript{4} flood compared to the N\textsubscript{2} flood (0.003 m\textsuperscript{3}/day vs 0.002 m\textsuperscript{3}/day), N\textsubscript{2} breakthrough occurs much earlier than CH\textsubscript{4} breakthrough (approximately 1 day compared to significantly less than a day). This is a result of the higher adsorption capacity of CH\textsubscript{4} compared to N\textsubscript{2} that acts to retard the CH\textsubscript{4} migration through the coal. While nitrogen breaks-through earlier than CH\textsubscript{4} the nitrogen flood takes much longer to displace the methane from the core than methane displacement of nitrogen.

With the mass balance summaries presented in Table 3 there is very close agreement between model and observations for the initial methane and nitrogen contents for both 2 MPa and 10 MPa experiments. The largest difference is between the initial N\textsubscript{2} content for the 2 MPa CH\textsubscript{4} flood where there is approximately a 15% difference. Only for the N\textsubscript{2} flood at 10 MPa does the final N\textsubscript{2} content exhibit a notable difference between experimental and numerical results (8.1 m\textsuperscript{3}/t vs. 7.0 m\textsuperscript{3}/t for experimental and numerical data respectively). The discrepancy in N\textsubscript{2} content is carried forward from the N\textsubscript{2} flooding experiment at 2 MPa to the CH\textsubscript{4} flood at 2 MPa that begins with the N\textsubscript{2} saturated core established by the preceeding core flood. Consequently, for this experiment a difference exists in the initial N\textsubscript{2} content between numerical and experimental results (6.9 m\textsuperscript{3}/t vs. 8.1 m\textsuperscript{3}/t).

5.2 Ternary Displacement - CH\textsubscript{4}/flue gas floods

Core floods were also performed were a mixed gas was injected to displace adsorbed methane from the core sample. This mixed gas was formulated to replicate, in an approximate fashion, a flue gas that could be produced from methane combustion and as such was 90% nitrogen and 10% CO\textsubscript{2}. Figure 7 presents the gas rates and upstream and downstream pressures for the 2 MPa experiment and Figure 8 presents the results for 10 MPa. A repeat flue gas flood was performed at 2 MPa but since the results are very similar to those presented in Figure 7 this is not presented however the mass balances and sorption time constants are presented in Table 5 and Table 6 along with the results for the other flue gas core floods. Figure 7 and Figure 8 demonstrate that good agreement could be achieved between simulated and experimental outflow rates for CH\textsubscript{4} and N\textsubscript{2}. However for the 2 MPa case the simulated outflow of CO\textsubscript{2} reaches steady state (that is inflow rates of CO\textsubscript{2} are equivalent to outflow) earlier than that observed and a good match could not be obtained indicating that significantly more adsorption was taking place in the
laboratory experiments than calculated in the simulations using the measured isotherms (in Figure 3) with the extended Langmuir adsorption model. This is also highlighted in the comparison of the CO\textsubscript{2} mass balances presented in Table 5 - the overestimate in CO\textsubscript{2} flow rate is reflected in the underestimate of the CO\textsubscript{2} gas content at the end of the experiment. However for the 10 MPa case the agreement between the observed and simulated CO\textsubscript{2} outflow rates is closer indicating that at this pressure the observed CO\textsubscript{2} adsorption is more closely represented by the simulation approach but there are still significant differences which will be discussed in further detail below.

The good matches of the simulated and the experimental data for CH\textsubscript{4} and N\textsubscript{2} are emphasised in the mass balance comparisons presented in Table 5. There is very close agreement between model and observations with the initial methane contents and the final N\textsubscript{2} contents. The quantity of CO\textsubscript{2} adsorbed, however, is significantly underestimated by the numerical model, resulting in the higher CO\textsubscript{2} outflow rates presented in Figure 7 and Figure 8. For a composition of 90\% N\textsubscript{2} and 10\% CO\textsubscript{2} the numerical model predicts that approximately the same quantities of N\textsubscript{2} and CO\textsubscript{2} are adsorbed, whereas experimental data shows that at 2 MPa approximately 87\% and at 10 MPa approximately 51\% more CO\textsubscript{2} than N\textsubscript{2} is stored. Possible reasons for the underestimate of CO\textsubscript{2} adsorption are a) use of an inaccurate CO\textsubscript{2} isotherm or b) the ELM used to describe multi-component sorption is not capable of predicting sorption behaviour in the ternary system accurately.

The contrasting adsorption behaviour of nitrogen and CO\textsubscript{2} is well demonstrated in the experimental results with nitrogen breaking through early and CO\textsubscript{2} much later. Another aspect is the displacement of methane where the methane outflow rate is high initially and drops rapidly immediately prior to CO\textsubscript{2} breakthrough. The CO\textsubscript{2}, even at relatively low concentrations, acting to efficiently displace the methane within the core sample.

5.3 Fracture-matrix transfer

There were two approaches considered here in the simulation of the transfer of gas between fracture and matrix; the dual porosity model of Eq. (4) and the triple porosity model where the matrix is partitioned into two regions as represented by Eq. (6). The fracture-matrix transfer influences the shape of the gas outflow behaviour with time, after breakthrough and before steady state. For rapid transfer, represented in the model by a small value of the sorption time property, \( \tau \), there would be a sharp transition to steady state after gas breakthrough.
It was found that the triple porosity model of Eq. (6) provided a better match between simulated and observed gas outflow behaviour than the dual porosity model of Eq. (4). Figure 9 presents a comparison between the simulated and observed methane outflow rates for the N\textsubscript{2} displacement of CH\textsubscript{4} core flood at 2 MPa calculated with dual (Eq. (4)) and triple (Eq. (6)) porosity models. These results demonstrate that the triple porosity model provides a more accurate description of the methane outflow than the standard Warren-Root dual porosity approach for this core sample. The outflow of CH\textsubscript{4} is best described with a porosity where 70\% of the gas is stored in a region with a high sorption time constant and the remaining 30\% in a region of the porosity where gas exchange is much slower. The end result is that the CH\textsubscript{4} outflow has a longer tail with respect to time than that calculated using the dual porosity model.

The triple porosity fractions as well as the sorption time constants used to achieve the production rate history matches are presented in Table 6 for the flue gas floods and Table 4 for the nitrogen and methane floods. The matrix porosity fractions are the same for all the core floods, at 0.3 and 0.7 for \(\phi_1\) and \(\phi_2\) respectively - independent of pressure and injectant gas. For the floods where N\textsubscript{2} displaces methane the sorption time constants are uniform for \(\tau_1\) with 3.5 days for CH\textsubscript{4} and 4 days for N\textsubscript{2} and range from 0.4 to 0.5 days for CH\textsubscript{4} and 0.1 to 0.5 days for N\textsubscript{2} for \(\tau_2\).

There is a distinct contrast in the sorption time constants for the core flooding experiments in which CH\textsubscript{4} displaces N\textsubscript{2} (see Table 4); the \(\tau_1\) value for CH\textsubscript{4} is 0.6 days for the 2 MPa experiment and 0.7 days for the 10 MPa experiment. These are significantly less than the value of 3.5 days for the sorption time constants found for the N\textsubscript{2} floods. The second time constant, \(\tau_2\), was also less being 0.2 days for the 2 MPa and 10 MPa experiments. For the N\textsubscript{2} outflow behaviour the analysis is more complicated where a satisfactory match for the N\textsubscript{2} outflow rates could not be obtained for the CH\textsubscript{4} core flooding experiment at 10 MPa (see Figure 6), meaning that the history matched N\textsubscript{2} sorption time constants were subject to some error. However, there was good agreement for the N\textsubscript{2} outflow rates for the CH\textsubscript{4} flood at 2 MPa which was achieved using N\textsubscript{2} sorption time constants of 0.8 days (\(\tau_1\)) and 0.09 days (\(\tau_2\)). This means that for both CH\textsubscript{4} and N\textsubscript{2} the sorption time constant \(\tau_1\) is significantly lower when the experiment is reversed from N\textsubscript{2} flooding to CH\textsubscript{4} flooding and the higher adsorbing component is displacing the lower adsorbing component. For \(\tau_2\) while the values obtained are smaller for the CH\textsubscript{4} flood compared to the N\textsubscript{2} flood they are much closer. The observed lower sorption times for CH\textsubscript{4} when it is adsorbing compared to the values
when it is desorbing are consistent with the results presented in Pan et al. (2010). In Pan’s work the
diffusivity (the inverse of sorption time) was consistently larger for adsorption than desorption.

The differences between the sorption times for adsorption and desorption core floods for CH$_4$ may be the
result of model inaccuracies or from the physical properties of the coal. In terms of modelling
inaccuracies, the modelling approach used here involves a number of approximations including that
adsorption/desorption is instantaneous and thus adsorption kinetics are ignored. If there are kinetic
effects these would be taken up in the sorption time constant. Another possibility is that the flow path of
gas molecules during adsorption differs from that during desorption, affecting the tortuosity. It is also
possible that coal matrix swelling could influence the flow path through the coal microporosity.

For the flue gas core floods the desorption time constants for N$_2$ are uniform and CH$_4$ are very close for
the 2 and 10 MPa experiments (see Table 6). However the CO$_2$ desorption time constants could not be
determined with confidence due to the significant differences between the simulated and observed
outflow rates for this gas. This indicates that there were other aspects of the model causing the
difference; that is differences in the adsorption isotherm and possibly the procedure used to calculate
mixed gas adsorption. This will be discussed in further detail in the subsequent section.

5.4 Sweep efficiency

The numerical model has an implied sweep efficiency of 100%, whereas the actual displacement
efficiency is often lower than that due to the heterogeneity of the flow path through the core. This means
the numerical model will predict complete displacement of the component initially in the core if the
simulation is run for long enough, while in fact some gas may remain trapped. However the
experimental estimate of sweep efficiency is dependent on accurate calculation of the mass balance over
the core flood which in turn relates to the accuracy of the measurement of flow rates during the
experiments. These measurements are subject to a degree of error from a number of sources; the flow
measurement itself is a function of the measurement device accuracy. These flow measurements are also
converted from test conditions to standard conditions which involve using pressure and temperature
measurements with an equation of state. As a result the mass balances involve some inaccuracy, some of
which may be cumulative over the experiment. To minimise these errors all experiments involved a
consistency check by running the core flood until steady state and comparison between inflow rates and
concentrations with those observed with outflow. However sweep efficiency, since it based on the
difference between cumulatives over the experiments which are relatively large quantities, is a comparatively small quantity of gas and therefore estimates could have a degree of error.

As a result of the model discrepancy in the calculation of sweep efficiency, the residual gas content of the displaced component can be higher for the experimental data than for the numerical results. This is indicated by the CH$_4$ flooding experiments at 2 and 10 MPa for which the residual N$_2$ content is 1.8 m$^3$/t while the numerical model predicts (almost) complete sweep. However, for the experimental N$_2$ floods almost complete displacement of CH$_4$ is achieved. For these experiments, the mass balance analysis highlights that N$_2$ is a more effective displacing agent than CH$_4$ and can achieve close to perfect sweep.

5.5 **Coal permeability behaviour**

Upstream pressure of core flooding experiments was matched through tuning of the core permeability and, where required, the gamma factor of Eq. (3). The gamma factor relates the role of sorption strain under hydrostatic conditions and given the linear relationship of bulk sorption strain to total gas content used in this work, may only be apparent when there is a change in total gas content during a core flood. This could happen, even under constant pore pressures, as one gas displaces another since the adsorption behaviour and capacity is gas specific. Also, since gamma relates the effect of the difference between bulk and pore strain on permeability, it may be possible that these strains are equal and therefore permeability would not change with gas content under constant pore pressure conditions. Gamma was adjusted based on the behaviour of the difference in upstream and downstream pressures; where there was an increasing or decreasing trend with change in total gas content this was adjusted to attempt to remove this trend as it was assumed that this was derived from sorption strain.

Table 4 presents the permeability used in the simulations at the start and at the end of the CH$_4$/N$_2$ core floods. The permeability did not change significantly during the 2 MPa experiments even though the total gas content changed from ~12 m$^3$/tonne CH$_4$ to ~8 m$^3$/tonne N$_2$ and vice versa in the reverse flood. This change in total gas content translates to a bulk volumetric sorption strain of 0.15% and $\gamma = 1$ was used, meaning bulk and pore sorption strains are equal, as no apparent sorption strain effect could be resolved for these experiments.

However for the 10 MPa CH$_4$/N$_2$ experiments $\gamma$ was used to describe the influence of sorption strain on the calculation of permeability. Figure 10 presents the difference between observed and simulated
upstream pressure calculated using $\gamma = 1$, no sorption strain effects, compared with $\gamma = 122$. The significant trend present with $\gamma = 1$ in the pressure difference reflects permeability behaviour not accounted for in the simulation model. However $\gamma = 122$ largely removes this trend supporting the relationship proposed by Eq. (3) with pore and bulk sorption strains not equivalent. For the 10 MPa displacement of N$_2$ by CH$_4$ a small $\gamma$ was estimated to match the pressure difference.

For the series of flue gas floods the total gas content did not change significantly over the simulations (see Table 5) and thus sorption strain effects on permeability were also not significant.

Coal permeability is sensitive to the antecedent stress conditions and therefore, in the laboratory, the loading history. Because of this issue it is difficult to compare permeability values between experiments, even on the same sample. Therefore even though permeability had been measured in the earlier characterisation work which estimated the properties presented in Table 2 it was not meaningful to compare these to the permeability values determined from the history matching presented here.

### 5.6 Adsorption Isotherms

Figure 11 presents a comparison between the gas contents measured from the core flood mass balances (from Table 3 and Table 5) and the Langmuir adsorption isotherms fitted to the experimental measurements presented in Figure 3. It can be seen from this figure that the CO$_2$ and CH$_4$ gas contents and isotherms are very close; CH$_4$ gas contents are slightly offset from the experimental isotherm but have a high degree of repeatability. However there are significant differences for N$_2$ and also scatter in these results. There are several potential sources for these differences; one could be from inaccuracy in the calculation of the mass balances, as described above. Since the N$_2$ core floods involved significantly larger quantities of gas than CH$_4$ or CO$_2$ as the N$_2$ core floods ran for much longer, any small errors would be magnified in the calculation of the gas content. With the N$_2$ results there appears to be a contrast between the pure gas and the mixed gas core floods. A possible explanation for this behaviour could be the calculation of mixed gas adsorption, the effects of which have not attempted to be represented in these figures since the gas species partial pressure for the gas mixtures is used.
In order to improve the match with the core flooding gas contents the N₂ Langmuir isotherm was also adjusted; this is the isotherm labelled as “Corrected”. The agreement between the CO₂ gas contents and isotherm was considered adequate for the history matching and was not modified.

The good agreement between the simulated and experimental production rates, mass balances and breakthrough times for the binary CH₄/N₂ core floods indicate that the extended Langmuir model could be used with confidence for these binary gas problems. This is consistent with the findings of Clarkson and Bustin (2000) and Jessen et al. (2007) who also found that the extended Langmuir model could accurately predict binary component mixtures. However, there are significant discrepancies in the representation of the CO₂ sorption behaviour by the simulation model for the ternary gas flow problems involving CH₄/CO₂/N₂. While simulated and observed CH₄ and N₂ flow rates and mass balances were in good agreement.

One explanation for the differences discussed above is that the extended Langmuir model is not accurate for gases at relatively low concentrations in multi-component mixtures, such as CO₂ for the experiments presented here. Similar observations were reported by Jessen et al. (2007) who explained the discrepancy between laboratory and numerical results to have been caused by adsorption hysteresis. In a subsequent study, Jessen et al. (2008) included sorption hysteresis in their displacement calculations and also evaluated the performance of the ideal adsorbate solution (IAS) model compared to the extended Langmuir model. In their study the IAS model yielded results superior to the extended Langmuir model, however, discrepancies with the experimental data were still present, indicating that hysteresis is not (or at least not the only) problem.

6. Conclusions

In the history matching for the 2 MPa core floods the best match between model and observations was obtained where pore and bulk sorption strains were equivalent and the permeability did not change significantly. However for the two 10 MPa binary core floods there was a significant trend in the permeability during the core floods and the best match was obtained where pore and bulk sorption strains were not equivalent in the Connell, Lu and Pan model. For the 10 MPa flue gas flood, since there was no significant net change in gas content, there was no sorption strain effect in the permeability model. These results provide evidence that the pore sorption and bulk strains may not be equivalent, at least at high pressures, even under hydrostatic conditions. Under uniaxial conditions it is possible that
the differences could be even more significant as the impact of sorption strain on cleat porosity is more pronounced.

In this work it was found that the description of the outflow of gas during the core flooding experiments could be improved by using a triple porosity approach when compared to predictions using the dual porosity Warren-Root model. With this triple porosity model the matrix was divided into two regions with different desorption time constants in a similar approach to Busch et al. (2004). In the history matching the best match between simulated and observed gas outflow behaviour was achieved where 30% of the matrix exchanged gas much slower than the remaining 70%. This division of the matrix was found to be constant for different gas species and pressures, indicating that it was representing a physical characteristic of the coal or a common attribute of the gas exchange process not well described by the Warren-Root model.

The simulated and experimental overall mass balances are in close agreement for the binary gas core floods involving N₂/CH₄. However there are significant differences in the CO₂ mass balances and outflow rates for the ternary core floods. The adsorbed amount of CO₂ was significantly underestimated in the simulations with the result that the CO₂ breakthrough occurred earlier in the outflow gas than that observed. One explanation for this is that the description of the multi-component adsorption process with the extended Langmuir model may be inaccurate for gases at low concentrations in a gas mixture; in these experiments the inflow concentration of CO₂ was 10%. While the error in CO₂ was significant, CH₄ and N₂ were accurately represented. Further work is required to substantiate this behaviour experimentally and identify an appropriate approach to represent it in simulation.

7. Acknowledgements

The authors gratefully acknowledge the support of the Australian Coal Association Research Program under project C17055 The feasibility of enhanced gas drainage for coal mine operations.
8. References


### Table 1 Core physical properties.

<table>
<thead>
<tr>
<th>Core Property</th>
<th>Bowen Basin</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volume</td>
<td>0.000329 m³</td>
</tr>
<tr>
<td>Length</td>
<td>0.114 m</td>
</tr>
<tr>
<td>Diameter</td>
<td>0.0606 m</td>
</tr>
<tr>
<td>Core area (circle)</td>
<td>0.002884 m²</td>
</tr>
<tr>
<td>Width for equivalent area (square: x=y) for model</td>
<td>0.05370 m</td>
</tr>
<tr>
<td>Dead space in experiment/model</td>
<td>0.00004435 m³</td>
</tr>
<tr>
<td>Mass</td>
<td>0.4207 kg</td>
</tr>
<tr>
<td>Bulk density</td>
<td>1,278 kg/m³</td>
</tr>
</tbody>
</table>
Table 2: Core sample properties measured in characterisation program.

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Coal density</td>
<td>1,280 kg/m$^3$</td>
</tr>
<tr>
<td>Pore compressibility</td>
<td>0.00005 1/kPa</td>
</tr>
<tr>
<td>Max strain (CH$_4$)</td>
<td>0.0125</td>
</tr>
<tr>
<td>Max strain (CO$_2$)</td>
<td>0.017</td>
</tr>
<tr>
<td>Porosity $\phi$</td>
<td>0.015</td>
</tr>
<tr>
<td>Young’s Modulus E $E$</td>
<td>2.6 GPa</td>
</tr>
<tr>
<td>Poisson’s ratio $\nu$</td>
<td>0.35</td>
</tr>
<tr>
<td>Biot coefficient</td>
<td>0.9</td>
</tr>
<tr>
<td>Bulk Modulus K $K$</td>
<td>2.89 GPa</td>
</tr>
<tr>
<td>Isotherm data</td>
<td></td>
</tr>
<tr>
<td>Langmuir volume CH$_4$</td>
<td>28.5 m$^3$/t</td>
</tr>
<tr>
<td>Langmuir pressure CH$_4$</td>
<td>2529 kPa</td>
</tr>
<tr>
<td>Langmuir volume CO$_2$</td>
<td>43.36 m$^3$/t</td>
</tr>
<tr>
<td>Langmuir pressure CO$_2$</td>
<td>1289 kPa</td>
</tr>
<tr>
<td>Langmuir volume N$_2$</td>
<td>25.21 m$^3$/t</td>
</tr>
<tr>
<td>Langmuir pressure N$_2$</td>
<td>5544 kPa</td>
</tr>
</tbody>
</table>
Table 3: Mass balance comparison of experimental and simulation results for CH$_4$/N$_2$ core floods (G$_{c,init}$ is the initial gas content and G$_{c,end}$ is the final gas content).

<table>
<thead>
<tr>
<th>Pressure MPa</th>
<th>Gas in coal</th>
<th>Injectant</th>
<th>Lab G$_{c,init}$</th>
<th>Lab G$_{c,end}$</th>
<th>Lab sweep Efficiency</th>
<th>SIM G$_{c,init}$</th>
<th>SIM G$_{c,end}$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>m$^3$/t</td>
<td>m$^3$/t</td>
<td>%</td>
<td>m$^3$/t</td>
<td>m$^3$/t</td>
</tr>
<tr>
<td>2</td>
<td>CH$_4$</td>
<td>N$_2$</td>
<td>12.6</td>
<td>0.0</td>
<td>8.1</td>
<td>100</td>
<td>13.0</td>
</tr>
<tr>
<td>10</td>
<td>CH$_4$</td>
<td>N$_2$</td>
<td>22.8</td>
<td>0.4</td>
<td>16.2</td>
<td>98</td>
<td>22.8</td>
</tr>
<tr>
<td>2</td>
<td>N$_2$</td>
<td>CH$_4$</td>
<td>8.1</td>
<td>12.9</td>
<td>1.8</td>
<td>78</td>
<td>6.9</td>
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<tr>
<td>10</td>
<td>N$_2$</td>
<td>CH$_4$</td>
<td>16.2</td>
<td>22.5</td>
<td>1.8</td>
<td>89</td>
<td>16.2</td>
</tr>
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</table>
Table 4: Coal properties derived through history matching of CH$_4$/N$_2$ core flooding experiments.

<table>
<thead>
<tr>
<th>Pressure MPa</th>
<th>Gas in coal</th>
<th>Injectant</th>
<th>$\phi_1$</th>
<th>$\phi_2$</th>
<th>$\tau_1$</th>
<th>$\tau_2$</th>
<th>$\kappa_{initial}$ mD</th>
<th>$\kappa_{end}$ mD</th>
<th>$\gamma$ sim</th>
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<tbody>
<tr>
<td>2</td>
<td>CH$_4$</td>
<td>N$_2$</td>
<td>0.3</td>
<td>0.7</td>
<td>3.5</td>
<td>0.5</td>
<td>4.0</td>
<td>0.10</td>
<td>2.0</td>
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<tr>
<td>10</td>
<td>CH$_4$</td>
<td>N$_2$</td>
<td>0.3</td>
<td>0.7</td>
<td>3.5</td>
<td>0.40</td>
<td>4.0</td>
<td>0.50</td>
<td>0.45</td>
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<tr>
<td>10</td>
<td>CH$_4$</td>
<td>N$_2$</td>
<td>0.3</td>
<td>0.7</td>
<td>3.5</td>
<td>0.6</td>
<td>0.8</td>
<td>0.09</td>
<td>2.0</td>
</tr>
<tr>
<td>10</td>
<td>CH$_4$</td>
<td>N$_2$</td>
<td>0.3</td>
<td>0.7</td>
<td>0.7</td>
<td>0.20</td>
<td>0.8</td>
<td>0.10</td>
<td>0.55</td>
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Table 5: Mass balance comparison of experimental and simulation results for CH₄/flue gas core floods (Gₐ,init is the initial gas content and Gₐ,end is the final gas content).

<table>
<thead>
<tr>
<th>Pressure MPa</th>
<th>Gas in coal</th>
<th>Injectant</th>
<th>Lab G_c,init m^3/t</th>
<th>Lab G_c,end m^3/t</th>
<th>SIM G_c,init m^3/t</th>
<th>SIM G_c,end m^3/t</th>
</tr>
</thead>
<tbody>
<tr>
<td>2</td>
<td>CH₄ 90% N₂, 10% CO₂</td>
<td>12.9</td>
<td>1.1</td>
<td>9.3</td>
<td>5.6</td>
<td>13.0</td>
</tr>
<tr>
<td>2</td>
<td>CH₄ 90% N₂, 10% CO₂</td>
<td>12.9</td>
<td>0.0</td>
<td>9.2</td>
<td>4.7</td>
<td>12.9</td>
</tr>
<tr>
<td>10</td>
<td>CH₄ 90% N₂, 10% CO₂</td>
<td>21.6</td>
<td>0.0</td>
<td>18.0</td>
<td>11.0</td>
<td>22.8</td>
</tr>
</tbody>
</table>
Table 6: Coal properties derived through simulation of CH4/flue gas core flooding experiments (measured permeability values are presented in bold).

<table>
<thead>
<tr>
<th>Pressure MPa</th>
<th>Gas in coal</th>
<th>Injectant</th>
<th>Matrix porosity φ1</th>
<th>CH4 τ1</th>
<th>CO2 τ1</th>
<th>N2 τ1</th>
<th>k_initial mD</th>
<th>k_end mD</th>
<th>γ</th>
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<tbody>
<tr>
<td>2</td>
<td>CH4</td>
<td>90% N2, 10% CO2</td>
<td>0.3 0.7</td>
<td>3.0 0.6</td>
<td>6.0 0.3</td>
<td>4.0 0.5</td>
<td>2.0</td>
<td>2.0</td>
<td>1</td>
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<tr>
<td>2</td>
<td>CH4</td>
<td>90% N2, 10% CO2</td>
<td>0.3 0.7</td>
<td>3.5 0.5</td>
<td>6.0 0.3</td>
<td>4.0 0.5</td>
<td>0.63</td>
<td>0.63</td>
<td>1</td>
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<tr>
<td>10</td>
<td>CH4</td>
<td>90% N2, 10% CO2</td>
<td>0.3 0.7</td>
<td>3.5 0.9</td>
<td>0.5 0.5</td>
<td>4.0 0.5</td>
<td>0.60</td>
<td>0.58</td>
<td>1</td>
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</table>
Figure 1. Schematic of the experimental equipment used in the core flooding experiments.
Figure 2. Illustration of the core sample and the model grid used to represent the core floods.
Figure 3. Adsorption measurements (symbols) and best fit Langmuir adsorption isotherm (solid lines) for the Bowen Basin coal sample. For the measurements, the filled symbols signify those made as the pressure is progressively increased whereas unfilled indicate decreasing pore pressure.
Figure 4. Measured coal swelling with gas adsorption with respect to the gas content for the three gases used in the core flood experiments for the Bowen Basin coal sample. Coal swelling is calculated from measured apparent swelling after allowing for pore pressure and confining pressure compression using the geomechanical properties in Table 2.
Figure 5. Simulated and measured gas rates (left) and upstream pressure, $P_{up}$, and downstream pressure, $P_{down}$, (right) for the binary core floods at 2 MPa: the top figures are for $N_2$ displacement of $CH_4$; the lower figures for the subsequent core flood where $CH_4$ displaced $N_2$. 
Figure 6. Simulated and measured outflow rates (left) and upstream pressure, Pup, (right) for the binary core floods at 10 MPa: the top figures are for N₂ displacement of CH₄; the lower figures for the subsequent core flood where CH₄ displaced N₂.
Figure 7. History match of flue gas displacement of CH₄ @ 2 MPa showing comparisons between gas rates (right) and upstream pressure (left).
Figure 8. History match of flue gas displacement of CH$_4$ at 10 MPa showing comparisons between gas rates (right) and upstream pressure (left).
Figure 9. A comparison of simulations of CH₄ outflow made with dual (Eq. (4)) and triple (Eq. (6)) porosity models for the N₂ displacement of CH₄ core flood presented in Figure 5; the methane outflow rate is presented in the top figure and the difference between observations and the simulated methane rates in the lower figure.
Figure 10. Difference between observed and simulated upstream pressure for 10 MPa coreflood of CH$_4$ by N$_2$ for two values of gamma in Eq. (3).
Figure 11. Comparisons of the Langmuir adsorption isotherms determined from measurements presented in Figure 3 and the gas contents measured in the core flooding (presented in Table 3 and Table 5).