Investigation of the Effect of Carbon Dioxide Sequestration on Coal Seams: A Coupled Hydro-Mechanical behaviour

Abstract
The process of CO$_2$ sequestration in deep coal seam causes both coal seam permeability and strength to be significantly reduced due to CO$_2$ adsorption-induced coal matrix swelling. In deep coal seams CO$_2$ exists in its super-critical state, which has quite different properties compared to sub-critical CO$_2$. The main objective of this study is to understand the effects of sub-critical and super-critical CO$_2$ injections on coal flow and strength properties.

A high pressure rig was first developed to conduct permeability tests and then used to conduct permeability tests for naturally-fractured black coal samples to identify the effect of CO$_2$ injection on coal permeability. According to test results, super-critical CO$_2$ adsorption creates much greater swelling effects and exhibits lower permeability values compared to sub-critical CO$_2$. Interestingly, N$_2$ has the potential to reverse CO$_2$-induced swelling to some extent. Strength tests were then conducted for different rank coals under different saturation conditions. According to the test results, the strength and Young’s modulus of both types of coals are reduced due to CO$_2$ saturation and the reduction is higher for super-critical CO$_2$ compared to sub-critical CO$_2$ saturation. For example, the reductions of UCS strength and Young’s modulus in black coal due to super-critical CO$_2$ adsorption are about 40% and 100% higher than the sub-critical CO$_2$.

Numerical models play an important role in identifying CO$_2$ flow behaviour in coal prior to field-work. Therefore, CO$_2$ movements in coal under laboratory conditions were successfully modelled using the COMET 3 simulator and field-scale models were then developed using the COMET 3 and COMSOL Multiphysics simulators. According to the models, CO$_2$ storage capacity in coal increases with increasing injecting pressure and temperature and decreasing moisture content. Moreover, determination of well arrangements is crucial for optimisation of CO$_2$ injections Furthermore, CO$_2$ injection causes the coal seam cap rock to be significantly deformed in an upward direction and may cause the injected CO$_2$ to leak.

Theoretical and empirical models play an important role in CO$_2$ sequestration in deep coal seams as they speed the identification of coal mass properties. Therefore, this has been considered in the next stage of the study. First, a theoretical equation for coal cleat permeability under non-zero lateral strain triaxial test conditions was developed, and then an empirical relationship for gas adsorption capacity in coal was developed.  

*Keywords: CO$_2$ sequestration, swelling in coal, permeability reduction, strength reduction*
1 Introduction

Global warming is an extremely important challenge for 21st century scientists, and numerous greenhouse gas mitigation and global warming control programs have been initiated during the last few decades. In addition, in order to address the critical human impact on climate change, many countries gathered and established the United Nations Framework Convention on Climate Change (UNFCCC). According to the policies of this gathering, industrial development should occur in a sustainable manner, and the greenhouse gas content in the atmosphere should be maintained such that it does not interfere with natural eco-systems. UNFCCC issued legally-binding warnings to many developed countries including Australia at the 15th UNFCCC conference held in Copenhagen in 2009. At this conference and at the 16th UNFCCC conference held at Cancun in 2010, all the member countries agreed to reduce greenhouse gas emissions in order that the total average world temperature increment will be limited to 2 °C in the future. It is therefore vital for Australian scientists to investigate possible CO\textsubscript{2} mitigation methods. Four main techniques have been identified to minimize atmospheric CO\textsubscript{2} levels: 1) less carbon-intensive fuels; 2) more energy-efficient methods; 3) increased conservation; and 4) carbon sequestration.

After much research on these techniques (Li et al., 2006), CO\textsubscript{2} sequestration has been identified as the most economical and most environmentally attractive method. According to scientists’ estimations (Schrag, 2007), it is necessary to sequestrate trillions of tons of CO\textsubscript{2} by the end of this century to maintain a safe CO\textsubscript{2} level in the atmosphere and therefore, it has been named as one the “21st Century Engineering Grand challenges” (http://www.engineeringchallenges.org/cms/8996.aspx). Up to date various CO\textsubscript{2} sequestration methods are being tested: 1) in depleted oil and gas reservoirs, 2) in saline aquifers, 3) in deep ocean beds, 4) in deep un-mineable coal beds and 5) as mineral carbonates.
Compared with other CO\textsubscript{2} sequestration methods, long-term storage in deep unmineable coal seams has been identified as safe, practical and economically attractive (White et al., 2005). The present study is therefore mainly focused on this particular CO\textsubscript{2} sequestration method. Adsorption is the main gas storage mechanism in coal, and according to existing findings, 98\% of CO\textsubscript{2} is stored in an adsorbed phase and the rest as free gas inside the cleats (Shi and Durucan, 2005). Therefore, CO\textsubscript{2} exists in a more stable form in coal seams, which causes the risk of CO\textsubscript{2} back-migration into the atmosphere to be greatly reduced (Gray, 1987). In addition, coal seams contain large amounts of methane (CH\textsubscript{4}), formed during the coalification process. Therefore, the ability to offset CO\textsubscript{2} sequestration costs using a valuable energy by-product like methane (CH\textsubscript{4}) is a unique advantage of this process. Since coal mass has higher affinity to CO\textsubscript{2} compared to CH\textsubscript{4}, while adsorbing CO\textsubscript{2} into the coal matrix the available CH\textsubscript{4} is released (Perera et al., 2011a). This phenomenon raises the concept of enhanced coal-bed methane recovery (ECBM). Moreover, coal seams have the potential to store a substantial amount of gas in their pore spaces due to the large surface area associated with the micro-pore structure (Stevens et al., 2000).

However, injection of CO\textsubscript{2} into a coal seam causes its physical and chemical structure to be significantly changed. According to existing findings (White et al., 2005), adsorption of CO\textsubscript{2} into a coal matrix causes strain to be induced between the adsorbing CO\textsubscript{2} molecules and the coal matrix surface, which is commonly known as coal matrix swelling. Although coal
matrix swelling is small compared to its volume, since coal has very low total porosity values, this process greatly reduces the pore space available for gas movement in the coal mass (Romanov et al., 2005). Under high pressure conditions, this swelling process can create a significant volumetric strain, which greatly reduces the CO₂ injection capacity or permeability of the coal seam. Therefore, the amount of storable CO₂ in a coal seam is highly unpredictable. In addition, potential coal seams for the CO₂ sequestration process exist at extremely deep locations (around 1000m below the surface). Therefore, the pressures and temperatures at these locations are higher than the critical values of gases such as CO₂, and CH₄. For instance, in the case of CO₂, the critical temperature is 31.8°C and the critical pressure is 7.38 MPa (Fig. 2). Since both of these factors normally exceed their critical values in deep coal seams, CO₂ exists in its super-critical state in these locations.

According to Qu et al. (2010), super-critical CO₂ has gas-like compressibility and solubility values and liquid-like density values. Therefore, the density and viscosity of super-critical CO₂ vary greatly with pressure and temperature. In addition, super-critical CO₂ has greater potential to displace existing gases from coal seams (White et al., 2005) and its adsorption capacity is much higher compared to sub-critical CO₂ (Krooss et al., 2002). These facts suggest that super-critical CO₂ behaves quite differently from sub-critical CO₂ and therefore, a completely different behaviour in coal seams can be expected for super-critical CO₂ compared to sub-critical CO₂.

In addition to the great reduction of coal seam permeability, CO₂ sequestration creates a significant influence on coal’s overall strength. The strength reduction associated with CO₂ adsorption is a great threat in terms of long-term safety, because it enhances the risk of CO₂ back-migration into the atmosphere and causes outbursts to occur in coal mining. Therefore, it is most important to address this issue.
As the CO₂ sequestration process in deep coal seams is a highly time- and money-consuming process, numerical models play an important role in identifying CO₂ flow behaviour in coal prior to field work. Many field-scale models have been developed to identify flow phenomena in porous media using different computer software programs including TOUGH 2 (Carneiro, 2009), COMSOL (Liu and Smirnov, 2009), FEMLAB (Holzbecher, 2005), COMET 3 (Pekot and Reeves, 2002), MSFLOW (Wu, 2002) and METSIM2 (Shi et al., 2008). However, less consideration has been given to the application of these software programs in laboratory-scale studies (Hadia et al., 2007). It is important for laboratory-scale studies such as triaxial experiments to analyse the experimental data and extend the findings to field conditions such as extreme pressures and temperatures, which are normally difficult to achieve in laboratories.

In order to inject the optimum amount of CO₂ into a selected coal seam, it is important to identify the effects of coal mass properties (moisture content, temperature), CO₂ pressure and injection and production well operations on its CO₂ storage capacity. Therefore, a comprehensive laboratory-scale study is necessary to understand these effects. In addition, CO₂ sequestration in any deep geological formation is associated with the risk of back-migration of the CO₂ into the atmosphere through its cap rock some time after injection. For instance, around 1700 people died in Cameroon in 1986 due to a leakage of naturally-sequestrated CO₂ from Lake Nyos (Benson et al., 2007). If the CO₂ injection pressure is not properly controlled, it is highly possible to have this kind of leakage in coal seams through their cap rock because high injecting pressures may cause fractures in the cap rock. Therefore, it is important to carry out an adequate field-scale numerical study before starting field work.

Theoretical and analytical models play an important role in CO₂ sequestration in deep coal seams as they obviate laboratory experimentation and speed the identification of coal mass properties. Coal mass has a natural cleat system and therefore estimation of cleat permeability is of utmost importance for CO₂ sequestration in coal seams. All the existing cleat permeability models have been developed on the basis of the basic assumption that lateral strain is zero. Although this is applicable under the in situ stress conditions in deep coal seams, it is not applicable for laboratory experiments such as triaxial tests as they clearly induce considerable lateral strain (Wang et al., 2007). Since triaxial experiments have been identified as the most suitable for permeability testing, it is important to develop a theoretical model to investigate cleat permeability variation in coal mass under triaxial test conditions. In addition to the permeability, the correct estimation of the gas adsorption capacity is also
important in the CO\textsubscript{2} sequestration process in coal. However, this is a difficult task using the existing gas adsorption models, as it is necessary to provide experimentally-evaluated parameters to use these models. For instance, although the Dubnin-Radushkevich (D-R) model is one of the widely-used models to estimate the gas adsorption capacity in coal, experimental pre-estimation of micro-pore capacity is necessary to use this model. Therefore, the present study has been structured to provide solutions to these issues.

The main objectives of the thesis can be outlined as follows:

1. To conduct a comprehensive experimental study
   a) to identify the sub-critical and super-critical CO\textsubscript{2} adsorption-induced swelling effects on the permeability of coal.
   b) to distinguish the permeability behaviours of coal under sub-critical and super-critical CO\textsubscript{2} flow conditions.
   c) to investigate the sub-critical and super-critical CO\textsubscript{2} adsorption effects on the strength of low rank and high rank coals.

2. To conduct a complete numerical study
   a) to develop a laboratory-scale numerical model using a suitable field-scale simulator to model CO\textsubscript{2} flow behaviour in coal under triaxial experimental conditions.
   b) to develop a field-scale numerical model using a user-friendly simulator to study the effect of coal seam physical properties, CO\textsubscript{2} injecting pressure and injection and production well operations on the CO\textsubscript{2} storage capacity of a selected coal seam, to be used as a base model for field work.
   c) to develop a field-scale numerical model using an appropriate and advanced simulator to study the risk of CO\textsubscript{2} back-migration into the atmosphere from a selected coal seam through the cap rock.

3. To develop the necessary theoretical and analytical studies
   a) to formulate an equation to predict cleat permeability in coal for CO\textsubscript{2} movement under non-zero lateral strain condition.
   b) to develop an appropriate and user-friendly model for gas adsorption capacity in coal.
2. Experimental investigation of CO\textsubscript{2} sequestration effects on coal hydro-mechanical properties

Before starting the CO\textsubscript{2} sequestration study it was necessary to design and construct a new rig to conduct permeability and strength tests for coal under in situ stress and temperature conditions. A high-pressure rig was therefore designed, constructed and certified for the required Australian standards. Since this is the first rig constructed in the Monash Civil Engineering Department for such high gas injection pressure conditions, close attention was given to avoid any accidents, particularly during the initial tests. The set-up is capable of delivering gas (N\textsubscript{2} and CO\textsubscript{2}) to the sample at injection pressures of up to 50 MPa, confining pressures of up to 70 MPa to simulate ground pressures to depths in excess of 2.5 km, and temperatures of up to 50 \(^\circ\)C. In addition, up to 10 tonnes axial load can be applied to the sample. The newly-developed set-up is shown in Fig. 3. It consists of five main units: 1) confining unit, 2) gas injecting unit, 3) water injecting unit, 4) loading unit and 5) temperature control unit.
Figure 3. The newly-developed high pressure rig in the Civil Engineering Laboratory (Ranjith and Perera, 2011)
The developed rig was then used to conduct permeability and strength tests to identify the effects of sub-critical and super-critical CO$_2$ injections on coal permeability and strength. Two types of coal were considered for the flow and strength experiments; 1) Southern Sydney Basin coal (from the Appin coal mine in the Bulli coal seam) and 2) Victorian brown coal (from the Latrobe Valley coal mine in the Yallourn coal seam). However, of these two types, priority has been given to the Southern Sydney Basin coal due to the lack of data available on CO$_2$ sequestration effects on high rank coal flow and strength properties compared to low rank coal. In particular, the effect of CO$_2$ sequestration on coal permeability was examined only for the Southern Sydney basin coal because some studies have already been conducted on this aspect for low rank coal (Victorian brown coal) for low gas injection pressures (Jasinge et al., 2011). Furthermore, it is difficult to conduct permeability testing under high pressures for this particular type of coal due to its low density and strength values. In addition, permeability tests were conducted for naturally-fractured Southern Sydney Basin high rank coal samples as the intact permeability of black coal is quite low (Siriwardane et al., 2009).

First, a permeability study was conducted to compare the sub-critical and super-critical CO$_2$ adsorption-induced swelling effects on the permeability of naturally-fractured black coal. A series of permeability tests was conducted using the newly-developed high pressure rig (Fig. 3) on 38 mm by 76 mm naturally-fractured black coal specimens (Fig.4).

![Cleats](image)

(a) Top surface (Downstream)         (b) Bottom surface (Upstream)

Figure 4. Naturally-fractured coal specimen (Perera et al., 2011b).

These tests were carried out for CO$_2$ and N$_2$ injections at 2-20 MPa injection pressures under 10 to 24 MPa confining pressures at 33 °C. Triaxial undrained tests were conducted for
all the permeability tests as it was necessary to achieve super-critical CO$_2$ conditions inside the coal mass pore space. Here, it should be noted that the downstream pressure was not equal to the upstream pressure, and it was therefore necessary to have very high injection or upstream pressures to create super-critical CO$_2$ conditions in the coal sample. Each coal specimen was then allowed to swell under sub-critical and super-critical CO$_2$ adsorption and the corresponding effects on CO$_2$ and N$_2$ permeabilities were examined. Results indicate that the permeability of naturally-fractured black coal is significantly reduced due to matrix swelling, which is significantly higher for super-critical compared to sub-critical CO$_2$ adsorption (Fig.5).

![Graphs showing permeability reduction vs. CO$_2$ injection pressure for sub-critical and super-critical adsorption](image)

(a) Sub-critical CO$_2$ adsorption effect  
(b) Super-critical CO$_2$ adsorption effect

Figure 5. Reductions of CO$_2$ permeability due to swelling after sub- and super-critical CO$_2$ adsorption-induced swelling (Perera et al., 2011b).

This is due to the fact that the amount of coal matrix swelling due to CO$_2$ adsorption clearly depends on the phase condition of the CO$_2$, and super-critical CO$_2$ adsorption-induced swelling is about two times higher than that induced by sub-critical CO$_2$ adsorption (see Fig.6).
An experimental study was then conducted to identify the effect of CO$_2$ phase condition on CO$_2$ flow behaviour in naturally-fractured black coal, which showed how the permeability of CO$_2$ varies when it converts to the super-critical state from its sub-critical state. Permeability tests were carried out for 15, 20 and 25 MPa confinements at 33.5 °C temperature. Two test scenarios were conducted to investigate: 1) variation of the permeability behavior of coal with CO$_2$ phase condition; and 2) the potential of nitrogen (N$_2$) to recover CO$_2$-induced swelling. According to the test results, the permeability of super-critical CO$_2$ is significantly lower than sub-critical CO$_2$ due to the higher viscosity and swelling associated with super-critical CO$_2$ (Fig.7). Moreover, at super-critical state there is a higher decline of CO$_2$ permeability with increasing injecting pressure due to the higher increments in the associated viscosity and swelling. Although CO$_2$ adsorption-induced swelling causes permeability of both CO$_2$ and N$_2$ to be reduced, it may also cause CO$_2$ permeability to increase for higher injecting pressures, because CO$_2$ flow behavior may transfer from super-critical to sub-critical after the swelling due to the decline of downstream pressure development. Moreover, N$_2$ has the potential to recover some swelling effects due to CO$_2$ adsorption, and this recovery rate is higher at lower injecting pressures and higher confining pressures.
Figure 7. Variation in permeability with varying CO$_2$ and N$_2$ injection pressures for initial CO$_2$ swollen and N$_2$ flooded samples under 15, 20 and 25 MPa confining pressure conditions (Perera et al., 2011c).

After the permeability tests a series of strength studies was conducted to investigate the effect of CO$_2$ sequestration on the strength of coal. First, a strength study was conducted to investigate the effects of different saturation mediums such as CO$_2$, N$_2$ and moisture on the strength of low rank coal. However, super-critical CO$_2$ saturation could not be achieved for the brown coal samples as application of more than 7.38 MPa gas confining pressure may cause the structure of low strength brown coal to change. A series of uniaxial experiments was conducted using the newly-developed high pressure rig on 38 mm diameter by 76 mm high Latrobe Valley brown coal samples with different saturation media (water, N$_2$, CO$_2$) and pressures (1, 2 and 3 MPa). According to the test results, water and CO$_2$ saturations cause the uniaxial compressive strength (UCS) of brown coal to be reduced by about 17% and 10% respectively. In contrast, N$_2$ saturation causes it to increase by about 2%. Moreover, the Young’s modulus of brown coal is reduced by about 8% and 16% due to water and CO$_2$ saturations respectively, and is increased up to 5.5% due to N$_2$ saturation. It can be concluded that CO$_2$ and water saturations cause the strength of brown coal to be reduced while improving its toughness, and N$_2$ saturation causes the strength of brown coal to increase, while reducing its toughness.
Table 1. Saturation effects on the uniaxial compressive strength (UCS) and Young’s modulus (E) values of brown coal samples (Perera et al., 2011d)

<table>
<thead>
<tr>
<th>Specimen</th>
<th>UCS</th>
<th>△UCS%</th>
<th>E (MPa)</th>
<th>△E%</th>
</tr>
</thead>
<tbody>
<tr>
<td>Natural</td>
<td>2.40</td>
<td></td>
<td>41.60</td>
<td></td>
</tr>
<tr>
<td>Water Saturated</td>
<td>2.00</td>
<td>-16.80</td>
<td>38.42</td>
<td>-7.63</td>
</tr>
<tr>
<td>CO₂ Saturated at 1 MPa</td>
<td>2.34</td>
<td>-2.53</td>
<td>40.32</td>
<td>-3.07</td>
</tr>
<tr>
<td>CO₂ Saturated at 2 MPa</td>
<td>2.25</td>
<td>-6.49</td>
<td>33.78</td>
<td>-18.78</td>
</tr>
<tr>
<td>CO₂ Saturated at 3 MPa</td>
<td>2.17</td>
<td>-9.60</td>
<td>35.08</td>
<td>-15.67</td>
</tr>
<tr>
<td>N₂ Saturated at 1 MPa</td>
<td>2.41</td>
<td>0.42</td>
<td>41.94</td>
<td>0.83</td>
</tr>
<tr>
<td>N₂ Saturated at 2 MPa</td>
<td>2.42</td>
<td>0.91</td>
<td>42.52</td>
<td>2.21</td>
</tr>
<tr>
<td>N₂ Saturated at 3 MPa</td>
<td>2.46</td>
<td>2.23</td>
<td>43.92</td>
<td>5.59</td>
</tr>
</tbody>
</table>

The final experimental study was then conducted to investigate the effects of sub-critical and super-critical CO₂ adsorption on the strength of high rank coals. A series of UCS tests was conducted on natural intact black coal samples under both sub-critical and super-critical CO₂ saturation conditions. In addition, N₂ saturated samples were used to compare the results with CO₂-saturated samples. According to the results, sub-critical CO₂ adsorption causes the UCS and Young’s modulus of the bituminous coal to be reduced by up to 53 % and 36%, respectively (Fig.8). Super-critical CO₂ adsorption causes more significant modifications to the mechanical properties of the bituminous coal, resulting in 40 % greater UCS strength reduction and 100 % greater Young’s modulus reduction compared to sub-critical CO₂ adsorption. The greater influence of super-critical CO₂ on the UCS of the bituminous coal is thought to be related to the greater adsorptive potential (and coal swelling produced) for super-critical CO₂. The more significant influence of super-critical CO₂ on the Young’s modulus of the bituminous coal is thought to relate to the greater dissolution (and thus coal plasticization) potential of the super-critical CO₂. N₂ saturation was not observed to have any significant effect on the mechanical properties of the bituminous coal.
3. Numerical investigation of CO\textsubscript{2} sequestration effects on coal hydro-mechanical properties

As the CO\textsubscript{2} sequestration process in deep coal seams is a highly time- and money- consuming process, numerical models play an important role in identifying CO\textsubscript{2} flow behaviour in coal prior to field work. It is particularly important for laboratory-scale studies such as triaxial experiments to analyse the experimental data and extend the experimental findings for field conditions such as extreme pressures and temperatures, which are normally difficult to achieve in laboratories. Therefore, a comprehensive study was conducted to develop an appropriate laboratory-scale model to simulate gas permeability in coal under triaxial test conditions.

A numerical model was developed using the COMET 3 field-scale simulator to examine and predict the CO\textsubscript{2} permeability values in coal under triaxial drained and undrained conditions. A basic model for gas movement in porous media under triaxial drained test condition was first developed for the movement of a non-reactive gas in a non-reactive medium (air flow in sandstone), which is the simplest triaxial test condition and therefore, needs fewer model parameters, resulting in a highly accurate model.

Figure 8. Effect of CO\textsubscript{2} adsorption on black coal mechanical properties (Perera et al., Under Review-a)
Figure 9. (a) Vertical pore pressure distributions in sandstone sample and, (b) horizontal pore pressure distribution at the bottom layer of the sandstone sample at the steady state condition for 180 kPa gas injecting pressure and 220 kPa confining pressure condition (Perera et al., 2011e).

The model was then extended to simulate the more advanced condition of CO₂ movement in naturally-fractured black coal (movement of a reactive gas in a reactive medium). The triaxial un-drained test condition was used for this case, as the experiments required for model verification were conducted under the un-drained condition.
The model calibration and verification were carried out using measured triaxial test data (newly developed high pressure rig was used) for CO₂ movement in naturally-fractured black coal at the temperature range of 25 to 70 °C. This model was then used to predict permeability under higher temperature conditions (80 to 200 °C) to understand the effect of temperature on the CO₂ permeability of deep coal seams (Fig.11). According to the developed laboratory-scale model, there is a clear increase in CO₂ permeability with increasing temperature for any confining pressure at high injecting pressures (more than 10 MPa). However, for low injecting pressures (less than 9 MPa) the temperature effect is less. With increasing injecting pressure, CO₂ permeability decreases at low temperatures (less than around 40 °C), and increases at high temperatures (more than 50 °C). Interestingly, the temperature effect on permeability is significant only up to around 90 °C within the 25 to 200 °C temperature limit. These observations are related to the sorption behaviour of the adsorbing CO₂ during the injection.
After conducting a comprehensive study on the development of a laboratory-scale model for the CO\textsubscript{2} sequestration process in coal under triaxial test conditions, an appropriate field-scale model was developed using the COMET 3 software to identify the field CO\textsubscript{2} sequestration process.

Figure 11. Model-predicted temperature effects on CO\textsubscript{2} permeability at two different depths (Perera et al., 2012a).

Figure 12. Selected geometry of the model grid blocks and the CO\textsubscript{2} distribution with time for 15 MPa CO\textsubscript{2} injection pressure at 30 °C (Perera et al., 2011f).
The model was then used to investigate the effects of coal mass physical properties such as moisture content, temperature, injecting gas pressure and production and injection well operations on the CO$_2$ storage capacity in a selected deep coal seam. This kind of numerical modelling provides important information about how these parameters should be controlled in the field to enable the injection of the optimum amount of CO$_2$ into a coal seam. According to the model, the temperature and moisture content of the coal seam and CO$_2$ injection pressure have significant effects on CO$_2$ storage capacity. According to the model results, CO$_2$ storage capacity decreases with increasing coal seam water saturation and increases with increasing temperature, and this observation relates to the adsorption behaviour of the injected CO$_2$. If the effect of CO$_2$ injection pressure is considered, during the initial injection period, the increase of CO$_2$ injection pressure causes the CO$_2$ injection rate to be significantly increased. However, this injection rate decreases with time due to coal matrix swelling and may again increase suddenly due to the formation of fractures.

![Graphs](image)

Figure 13. Variation of coal seam CO$_2$ storage capacity with (a) injection pressure, (b) temperature and, (c) moisture content (Perera et al., 2011f).

The developed model was then extended to investigate the effect of injecting and production well operations on CO$_2$ storage capacity in deep coal seams. According to the
model results, the maximum storage capacity can be obtained by having two injection wells for the selected coal seam (Fig 14). However, a further increase in the number of wells (up to four) causes the storage capacity to be reduced due to the coincidence of injecting wells creating pressure contours (Fig 15). This is confirmed by the fact that, for the two injecting wells condition, when the distance between the injecting wells increases from 150m to around 570m, the CO₂ storage capacity increases by about 270%. Considering all of these observations, it can be concluded that, not only the number of injecting wells but also the distance between the injecting wells should be calculated using an appropriate numerical model, before starting any field CO₂ injection to enable the injection of the optimum amount of CO₂ into a selected coal seam.

![Figure 14. Variation CO₂ storage capacity with number of injection wells (Perera et al., 2012b).](image)

![Figure 15. CO₂ pressure developed after 10 years of injection for two to four injection wells (Perera et al., 2012b).](image)
Moreover, CO₂ storage capacity can be significantly increased by inserting a water production well, because it causes the pore pressure inside the coal seam to be reduced. However, this storage capacity increment will reduce with time due to the reduction of mobile water inside the coal seam. Therefore, the effectiveness of the production well will reduce and disappear with time. Furthermore, when the distance between the production and injecting wells was reduced from 700m to 400m, the CO₂ storage capacity was increased by around 30% for the selected coal seam (Fig.16), because the pore pressure reduction effect from the production well will more quickly affect the CO₂ injecting area. However, further reduction of distance among the wells causes the storage capacity to be reduced due to the mixing of injecting CO₂ with desorbing CH₄ from the coal seam. Therefore, it is also important to calculate the distance between the production and injecting wells.

![Figure 16. Variation of CO₂ storage capacity with distance between the injection and production wells (Perera et al., 2012b).](image)

Conducting a safe CO₂ sequestration process is as important as studying the CO₂ sequestration process in coal and the ways to optimize it. Therefore, a field-scale model was then developed to investigate the effect of CO₂ injection pressure on cap rock deformation. In this case, the COMSOL Multiphysics simulator was used instead of the previously used COMET 3 simulator as it was necessary to model coupled flow and strength behaviour to identify the cap rock and coal mass deformations upon CO₂ injection.
In addition to being caused by the injecting gas pressure, cap rock deformation also occurs with CO$_2$ adsorption-induced coal mass swelling. This factor was also considered in the model development to find the total deformation of the cap rock on CO$_2$ injection. The developed model shows that during the CO$_2$ injecting period, the cap rock is vertically deformed by CO$_2$ pressure (Fig.18b). Moreover, the maximum vertical deformation of the cap rock and the CO$_2$ spread rate along the interface increase with the CO$_2$ injecting pressure (Fig 18a). This vertical deformation of the cap rock increases from around 0.2 mm to 15 mm when the gas injecting pressure increases from 10.2 MPa to 30 MPa.
This model was then used to determine the optimum CO$_2$ injection pressure into a selected coal seam to avoid any hydraulic fracture or shear failure in the cap rock, which cause the injected CO$_2$ to back-migrate into the atmosphere some time after CO$_2$ injection. According to the model, as a result of gas injection, the cap rock may fail at around 21 MPa due to hydraulic fracture, and at around 19 MPa due to shear failure (Fig.19). Considering the lower value and a 1.2 safety factor, the maximum gas injecting pressure should not exceed 16 MPa to avoid any failure in the cap rock of the selected coal seam.
Figure 19. Optimum CO$_2$ injection pressure to avoid (a) hydraulic fracture and, (b) shear failure in the cap rock (Perera et al., Under review-b).

4. Development of a theoretical model for coal cleat permeability under CO$_2$ movement

Theoretical and empirical models play an important role in the CO$_2$ sequestration process in deep coal seams as they obviate laboratory experimentation and speed the identification of coal mass properties. Cleat permeability is the most important parameter which can be used to determine the CO$_2$ sequestration potential of any coal seam, as it represents the balance between effective stress and matrix shrinkage and swelling. To date, many studies have been conducted on this aspect and various theoretical and empirical relationships have been proposed for cleat permeability in coal. However, these models have been developed on the basic assumption that lateral strain is zero, which is not applicable under laboratory experimental conditions with triaxial testing. Therefore, by applying the theory of elasticity to the constitutive behaviour of fractured rocks, a theoretical relationship between permeability and gas injecting pressure, confining pressure, axial load and gas adsorption in triaxial tests has been developed.
The developed model is shown below:

\[
\begin{align*}
k &= k_0 \exp \left\lbrace \Delta \sigma_{ch} \left[ \frac{3E}{E_f} \frac{(1 - \vartheta_f)}{2(1 - 2\vartheta_f)k_i} - \frac{3(1 + \vartheta_f)}{2E_f} \right] \right\} \times \exp \left\lbrace \Delta \sigma_{c} \left[ \frac{E}{E_f} \frac{(1 - \vartheta_f)}{2(1 - 2\vartheta_f)k_i} \right] \right\} \times \exp \left\lbrace \Delta \sigma_{A} \left[ \frac{E}{E_f} \frac{(1 - \vartheta_f)}{2(1 - 2\vartheta_f)k_i} \right] \right\} \times \exp \left\lbrace \Delta \sigma_{\text{CO}_2} \left[ \frac{E}{E_f} \frac{(1 - \vartheta_f)}{2(1 - 2\vartheta_f)k_i} \right] \right\}
\end{align*}
\]  

where, \( k \) is the cleat permeability, \( k_0 \) is the initial cleat permeability, \( \sigma_c \) is the confining stress, \( \sigma_A \) is the axial stress and \( \sigma_{\text{CO}_2} \) is the CO\(_2\) injecting pressure into the coal sample under triaxial test condition, \( \vartheta \) is the matrix Poisson’s ratio, \( \vartheta_f \) is the fracture Poisson’s ratio, \( E \) is the sample matrix Young’s modulus, \( E_f \) is the fracture Young’s modulus, \( S \) is the adsorption mass, \( \alpha \) is the volumetric swelling coefficient, \( k_i^\text{bulk} \) is the bulk modulus.

The new model was then verified using experimentally-determined permeability data (newly developed high pressure rig was used) of two coal samples. Results indicate that the new model can quite accurately predict the combined effects of effective stress and coal matrix swelling on cleat permeability for both CO\(_2\) and N\(_2\) injections at various injection pressures. The model also provides quite accurate prediction of the effect of confining
pressure on cleat permeability for both CO$_2$ and N$_2$ injections. The model includes parameters for fractured rock properties namely Poisson’s ratio and Young’s modulus. The model can be applied to predict cleat permeability, regardless of cleat size.

Figure 21. Variation of model-predicted and experimentally-evaluated CO$_2$ permeability values (a) with injecting pressure at 5 MPa confining pressure, (b) with injecting pressure at 10 MPa confining pressure, (c) with injecting pressure at 15 MPa confining pressure, (d) with confining pressure at 2 MPa injecting pressure, (b) with confining pressure at 2 MPa injecting pressure, (c) with confining pressure at 4 MPa injecting pressure conditions (Perera et al., Under review-c).

5. Development of an analytical model for gas adsorption capacity in coal

Adsorption plays an important role in the carbon dioxide sequestration process in deep coal seams as it is the main gas storage mechanism inside the coal mass. Therefore, correct estimation of gas adsorption capacity is most important for the related field-work to estimate the CO$_2$ storage capacity in a selected coal seam. Although the D-R equation is one of the most widely-used gas adsorption models, experimental evaluation of micropore capacity is
necessary to use this equation. Therefore, a new descriptive model for the gas sorption
capacity of coal as a function of various factors is proposed. The new model is based on the
existing D-R equation, which has been modified by inserting a new expression for the term
for micro-pore capacity.

If $T > T_c$ (T is the temperature and $T_c$ is the critical temperature),

$$
\log \left( \frac{V}{1 - \frac{\rho}{\rho_s}} \right) = \log \left\{ \frac{1}{T} \left[ a(V_{\text{co}} \%) + b(V_{\text{mic}} \%) + c(W_c \%) \right] + g \right\} - \frac{BT^2}{\beta_a^2} \log \left( \frac{\rho_s}{\rho} \right) \right\} \tag{2}
$$

If $T < T_c$,

$$
\log (V) = \log \left\{ \frac{1}{T} \left[ a(V_{\text{co}} \%) + b(V_{\text{mic}} \%) + c(W_c \%) \right] + g \right\} - \frac{BT^2}{\beta_a^2} \log \left( \frac{\rho_s}{\rho} \right) \right\} \tag{3}
$$

where, $\rho$ is the adsorbing agent density (kg/m$^3$) and $\rho_s$ is the density of adsorbed phase. $p$ is
the pressure (MPa) and $p_s$ is the saturated vapour pressure of the adsorbed gas (MPa), $\beta_a$ is
the affinity coefficient of the adsorbate, and $d_e$ is the effective molecular diameter of
adsorbate (nm).

Two types (CO$_2$ and N$_2$) of gas adsorption data for coal from five different locations
(British Colombia and Alberta in Canada and Victoria, Sydney and Bowen in Australia) at
three different temperatures (273, 296.5 and 318 K) were considered for the model
development. According to the model, the new gas adsorption equations can quite accurately
predict the adsorption capacity of coal.
6 Conclusions

A comprehensive study has been conducted using experimental, numerical, theoretical and analytical approaches and the main conclusions drawn from the study can be listed as follows:

- The permeability of coal is significantly reduced due to swelling, which starts as quickly as within 1 hour of CO$_2$ injection.
- The amount of coal matrix swelling due to CO$_2$ adsorption clearly depends on the phase condition of the adsorbing CO$_2$, where super-critical CO$_2$ adsorption-induced swelling is up to two times greater and therefore permeability is significantly lower than sub-critical CO$_2$.
- N$_2$ has the potential to reverse some swelling areas created by CO$_2$ adsorption.
- CO$_2$ adsorption has a more significant influence on the mechanical properties of black coal than brown coal due to the well-developed cleat system in black coal. Sub-critical CO$_2$ adsorption causes the UCS and Young’s modulus of brown coal to be reduced by about 10% and 16% respectively, and of black coal to be reduced by 53% and 36%, respectively.
Super-critical CO₂ adsorption has a much greater influence on coal’s mechanical properties compared to sub-critical CO₂, and it causes the UCS and Young’s modulus of black coal to be reduced by up to 78% and 71%, respectively.

CO₂ movement in naturally-fractured black coal under triaxial test conditions can be successfully modelled using the COMET 3 field-scale numerical simulator.

According to the developed laboratory-scale model, there is a clear increment in CO₂ permeability with increasing temperature under any confinement for high injecting pressures (more than 10 MPa), while for low injecting pressures (less than 9 MPa) temperature has little effect on permeability.

According to the developed field-scale models, CO₂ storage capacity increases with decreasing bed moisture content and increasing temperature and CO₂ injection pressure. However, the injection pressure effect will be gradually reduced by the process of coal matrix swelling. It is also important to have an adequate distance between the injection wells to inject the optimum amount of CO₂ into any coal seam, as reduced distance may cause the storage capacity to be reduced due to the coincidence of the pressure contours created by each injecting well.

Cap rock deforms considerably in an upward direction due to the CO₂ injection pressure and the amount of deformation depends to a great extent on the injecting gas pressure and duration. Therefore, high injection pressures may cause back-migration of the injected CO₂ into the atmosphere.

An accurate theoretical relationship for coal cleat permeability under triaxial, non-zero lateral strain conditions can be obtained by using basic geotechnical engineering fundamentals. This model is expected to be highly useful in future, laboratory-scale CO₂ sequestration studies to establish the required triaxial test conditions.

An accurate descriptive model for CO₂ adsorption capacity in coal can be developed by applying a multi-linear regression approach to the micro-pore capacity term in the existing D-R equation.

7 References


*Papers resulting from this study*