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3 **Manuscript Title:**

4 A macro-scale view of the influence of effective stress on carbon dioxide flow behaviour in coal:
5 An experimental study

6

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25 **Abstract**

26 Existing studies highlight the uncertainty in the process of CO₂ sequestration in deep coal seams,
27 mainly due to the associated CO₂ adsorption-induced coal matrix rearrangements. This complexity
28 is further increased by the highly heterogeneous nature of the coal mass which causes issues in
29 reproducing field conditions under laboratory conditions. The main objective of this study is
30 therefore to determine the permeability behaviour in coal for CO₂ flow using macro-scale
31 reconstituted coal specimens (203 mm in diameter and 1000 mm in length), particularly under
32 various effective stress conditions. A series of core flooding experiments was conducted on
33 Australian brown coal, using an advanced core flooding apparatus, for a range of CO₂ injection
34 pressures and axial stresses. According to the findings, CO₂ permeability decreases with increasing
35 CO₂ injection pressure, and the reduction is greater for super-critical CO₂ at greater depths. The
36 critical zone of influence for CO₂ injection into a selected coal seam is greater at lower injection
37 pressures and at shallow depths, and it reduces with increasing CO₂ pressure and seam depth. CO₂
38 storage capacity in a selected coal seam is greater at lower depths and higher CO₂ injection
39 pressures. However, the reduction of CO₂ storage capacity with depth was not very significant,
40 which is important for field CO₂ sequestration projects, which normally use deep seams to store
41 CO₂.

42 *Keywords: CO₂ storage; Coal permeability; Core flooding test; Effective stress; Low rank coal*

43 1. Introduction

44 Due to the capability of reducing the amount of anthropogenic carbon dioxide (CO₂) in the
45 environment, CO₂ sequestration in deep un-mineable coal seams has been identified as an effective
46 method to address global warming (Jing et al. 2015; Verma and Sirvaiya 2016; White et al. 2005).
47 Based on the fact that coal mass has higher affinity to CO₂ than other gases like methane (CH₄) and
48 nitrogen (N₂) due to its greater chemical and physical interaction, the sequestration process in return
49 releases the coal bed methane (CBM), which is an environmentally-friendly energy source with
50 higher efficiency (White et al. 2005). However, according to the research literature (Day et al. 2008;
51 Hol and Spiers 2012; Jing et al. 2015; Larsen et al. 1997; Pan and Connell 2007; Wang et al. 2013),
52 the sequestration of CO₂ disrupts the stability of the original coal structure with apparent changes in
53 coal matrix swelling, which decreases the CO₂ injectability into the coal seam and affects its long-
54 term stability. It is therefore important to study the principles of coal mass swelling and to have a
55 better understanding of the factors affecting this process.

56 A number of studies have been conducted on coal seam permeability upon CO₂
57 sequestration and the effects of various factors on this have been highlighted, including injecting
58 CO₂ properties (Jasinge 2010; Day et al. 2008; Pan and Connell 2007; Perera et al. 2011;
59 Siriwardane et al. 2009; Vishal and Singh 2015) and coal mass properties (Merkel et al. 2015;
60 Wang et al. 2013). According to the research, potential coal seams for CO₂ sequestration with
61 higher temperatures and pressures have limited CO₂ flow ability, because CO₂ is in its super-critical
62 state (beyond 7.38 MPa pressure and 31.8⁰C temperature CO₂ exists as super-critical fluid) and has
63 higher sorption capacity in such seams.

64 On the other hand, it is well known that coal is a highly heterogeneous material and it is
65 therefore quite difficult to validate the results obtained from natural coal samples for in-situ coal
66 seams (De Silva 2013). Jasinge (2010) and Liu et al. (2016) found that reconstituted coal (RC)
67 samples with reproducible properties can be used to successfully represent natural coal specimens,

68 to overcome such difficulties. The following section focuses on some previous studies conducted on
 69 RC and natural samples.

70 **1.1 Comparison of fluid flow behaviour in reconstituted and natural coal**

71 To date, many studies have been conducted to identify the flow behaviour in natural coal seams
 72 using reconstituted coal samples and satisfactory results have been obtained. Fig. 1 illustrates a
 73 study that compares N₂ and CO₂ permeability and related swelling in natural and reconstituted
 74 Australian low rank coal (Jasinge, 2010).

76 Fig. 1 (a) N₂ and (b) CO₂ permeability variation with effective stress and the variation of coal mass
 77 swelling with (c) N₂ and (d) CO₂ flow for reconstituted coal (RC) and natural coal specimens
 78 (Jasinge 2010) (The data labels in (c) and (d) denote the respective injection pressures)

According to Fig. 1(a), N₂ flow linearly reduces with increasing effective stress in both natural and RC samples and CO₂ flow (see Fig. 1(b)) follows a negative exponential variation, exhibiting an unfavourable effect of CO₂ exposure to coal permeability. Further, the RC specimens show a higher permeability compared to the natural specimens for both N₂ and CO₂ flow (see Figs. 1(a) and (b)). Similar results were obtained by Liu et al. (2016) who injected CH₄ (~0.69 MPa) to coking coal (from Hexi coal mine, China) at different confining pressures (from around 2-7 MPa) which exhibited higher permeability for RC specimens than natural coal samples. This indicates the difference of fracture system characteristic such as fracture porosity and connectivity of the reconstituted coal sample and the natural coal sample. Further attention to coal mass swelling with N₂ and CO₂ permeation reveals that N₂ injection does not show any significant variation in swelling with effective stress (see Fig. 1(c)) for both natural and RC samples. Conversely, CO₂ flow causes the coal mass swelling to undergo an increasing trend with the increase of injection pressure (see Fig. 1(d)) for both natural and RC specimens. However, RC specimens display a comparatively higher coal matrix swelling compared to natural specimens during CO₂ flow. This is probably caused by the higher amount of CO₂ molecules enter to the coal mass (due to the higher permeability in RC samples compared to natural samples) which will interact with coal mass and create greater swelling. This may also contribute for the observed permeability variations (see Fig. 1(b)) having lower permeability reductions for natural samples than RC samples during CO₂ permeation. However, the correlation between swelling and permeability of coal generally shows a similarity in both natural and RC specimens, allowing the use of RC specimens for experimental purposes (Jasinge 2010, Liu et al. 2016).

1.2 Studies on CO₂ flow behaviour using macro-scale coal specimens

As recorded in the literature (Day et al. 2008; Jasinge 2010; Pan et al. 2010; Perera et al. 2011; Siriwardane et al. 2009; Sun et al. 2016; Vishal and Singh 2015), most coal CO₂ injection studies have been conducted using meso-scale samples (up to around 100 mm in length). This has caused difficulties in confirming the applicability of adsorption theories at a higher scale to the estimation

105 of CO₂ storage capacity (De Silva 2013). Hence, De Silva (2013) conducted some advanced core-
 106 flooding tests using 1 m long and 203 mm diameter (macro-scale) RC low rank coal samples to
 107 effectively estimate the CO₂ flow behaviour in coal to overcome the scale effect. De Silva (2013)
 108 basically focused on the sub-critical CO₂ flow in large coal samples and Ranathunga et al. (2015)
 109 extended this study to super-critical CO₂ to represent actual CO₂ flow behaviour in deep coal seams,
 110 considering the CO₂ flow behaviour along the sample. The following sections summarise some
 111 major findings of the macro-scale core flooding experiments conducted on CO₂ injection.

112 ➤ *Sub-critical CO₂ injection*

113 De Silva (2013) conducted a series of tests of sub-critical CO₂ injection (from 0.4 MPa to 7.4 MPa)
 114 under 1, 2, 4, 6 and 8 MPa axial stresses and observed a reduction in CO₂ permeability with
 115 increasing CO₂ injection pressure and axial stress (Fig. 2). CO₂ adsorption-induced coal matrix
 116 swelling is the main causative fact for this permeability reduction and this was proved by the related
 117 swelling data (Fig. 2). According to Fig. 2, the coal volumetric strain is increased with increasing
 118 injection pressure. Further, according to the figure, the increase of axial stress causes a reduction in
 119 permeability for CO₂ movement, probably due to the greater effective stresses applying on the coal
 120 mass.

121

Fig 2. CO₂ permeability variation with the axial stress and volumetric strain for different CO₂ injection pressures (The first value of the label denotes the CO₂ injection pressure (MPa) and the second value denotes the respective volumetric strain (%)) (De Silva 2013)

➤ *Super-critical CO₂ injection*

As potential coal seams for CO₂ sequestration undergo super-critical CO₂ injection, Ranathunga et al. (2015) conducted a series of tests using super-critical CO₂ to understand the variations of CO₂ phase in large-scale coal specimens. They injected 6 to 10 MPa CO₂ into low rank RC specimens under constant 38 °C temperature (> 31.8 °C, the critical temperature of CO₂) and under 11 MPa axial stress. They injected N₂ before (first N₂ injection) and after the CO₂ injection (second N₂ injection) to quantify the CO₂ flow-induced coal mass changes and the N₂ and CO₂ permeability variations obtained are illustrated in Fig. 3.

Fig 3. Permeability variation with different injection pressures for first N₂ injection, CO₂ injection and second N₂ injection (Ranathunga et al. 2015) (The data labels indicate the coal specimen's volumetric strain (%) under each injection condition. The permeability values have been normalized by dividing each value by the permeability obtained for 6 MPa in each injection condition)

According to Fig. 3, the results are consistent with the results obtained by De Silva (2013) for sub-critical CO₂, showing a reduction of permeability with increasing CO₂ injection pressure.

140 However, it can be noted that the reduction is comparatively higher in the super-critical region
141 compared to sub-critical CO₂. For example, there is around a 7% decrease in permeability from 6 to
142 7 MPa sub-critical CO₂ injection and around 21% permeability decrease from 8 to 9 MPa super-
143 critical CO₂ injection. This reduction is three times higher than in sub-critical CO₂ flow. The main
144 reason for this observation is the higher adsorption capacity of super-critical CO₂, as a result of its
145 greater chemically active nature, liquid-like density and viscosity that cause greater coal mass
146 swelling compared to sub-critical CO₂ (Day et al. 2008). This is further evident from the volumetric
147 swelling data for both regions (see Fig. 3). Furthermore, the comparative reduction of the second N₂
148 flow compared to the first N₂ flow clearly illustrates the coal mass changes which occur upon
149 exposure to CO₂. For example, at 6 MPa, the first N₂ to the second N₂ injection shows around 17%
150 permeability reduction, and that for 8 MPa is around 31%. As N₂ is a comparatively less adsorptive
151 gas compared to CO₂ (Perera et al. 2011), this observed flow variation clearly occurs due to coal
152 mass swelling during CO₂ flux.

153 In addition, De Silva (2013) observed a decrease in CO₂ permeability in coal with increasing
154 effective stress (axial stress) for sub-critical CO₂. Hence, it is important to investigate how super-
155 critical CO₂ flow changes with increasing effective stress. Ranathunga et al. (2016) conducted a
156 series of meso-scale experiments to investigate this super-critical CO₂ effect for similar low rank
157 natural coal samples and observed the similar behaviour of CO₂ flow and permeability reduction
158 with increasing effective stress. Table 1, lists some of the results obtained by Ranathunga et al.
159 (2016) for 6 to 7 MPa (sub-critical CO₂), 7 to 8 MPa (the CO₂ phase changing region), and 8 to 9
160 MPa (super-critical CO₂) flow increments under 11, 14 and 17 MPa axial stresses.

161 According to Table 1, reduction of permeability with increasing effective stress is evident
162 and greater permeability reduction in deep coal seams is shown during CO₂ sequestration.
163 Furthermore, this reduction is gradually increased when the CO₂ phase is changed, being
164 comparatively greater in super-critical CO₂ compared to sub-critical CO₂. The opportunity for more
165 adsorptive super-critical CO₂ molecules to interact with the coal matrix, creating increased matrix

rearrangements is the main reason for this observation (Perera et al. 2011). However, this may create complications for CO₂-ECBM in deeper coal seams for CO₂ injectability and productivity. Hence, further research is needed on this to gain better understanding of this phenomenon for application in the field.

Table 1. Variation of CO₂ permeability with effective stress (Ranathunga et al. 2016)

Confining pressure (MPa)	Permeability variation (%)		
	6 to 7 MPa sub-critical CO ₂ flow increment	7 to 8 MPa CO ₂ phase changing region	8 to 9 MPa super-critical CO ₂ flow increment
11 (around 400m depth)	-1.8*	-9.7*	-19.2*
14 (around 500m depth)	-7.6*	-12.4*	-25.1*
17 (around 600m depth)	-10.6*	-23.2*	-30.3*

*A negative sign indicates the permeability reduction

The main objective of this study is therefore to develop knowledge of coal mass behaviour with CO₂ exposure using macro-scale reconstituted low rank coal samples, particularly to identify the influence of axial stress on large-scale samples. The present study can therefore be considered as an extension of the work of De Silva (2013) and Ranathunga et al. (2015), which pays more attention to super-critical CO₂ flow behaviour in coal located at various depth using macro-scale coal samples. An effort was also made to quantify the corresponding CO₂ storage capacity variation with various effective factors (CO₂ phase, pressure and seam depth).

2 Experimental methodology

The advanced core-flooding apparatus available in the Deep Earth Energy Research Laboratory (DEERL) at Monash University (Fig. 4) was used to conduct this study. A detailed description of the apparatus can be found in De Silva (2013) and the experimental procedure is briefly described below.

2.1 Sample preparation

In this experiment RC samples were prepared using powdered low rank brown coal. The coal powder was obtained from coal blocks taken from the Hazelwood open-cut coal mine at Morwell in

187 Gippsland, Victoria. The samples had a natural moisture content of around 55%. The RC specimen
188 construction procedure is shown in Fig. 5, and the detailed methodology adopted to construct the
189 RC samples can be found in De Silva (2013) and Ranathunga et al. (2015).

Fig 4. (a) Advanced core flooding apparatus (Ranathunga et al. 2015) and (b) the schematic diagram of the system

Fig 5. Experimental program for core flooding tests

2.2 Permeability tests

Permeability tests were conducted for CO₂ injections at seven different injecting pressures (6, 7, 8, 9, 10, 12 and 14 MPa) under 17 MPa axial stress, maintaining the system temperature at 38 °C. Since the system temperature was maintained at 38 °C, 6 and 7 MPa CO₂ injections were under sub-critical conditions and the rest of the CO₂ injections were under super-critical conditions (beyond 7.38 MPa pressure and 31.8°C temperature CO₂ exists as a super-critical fluid). The undrained condition was maintained to determine the sample permeability, and the pressure-transient approach was used calculate the permeability (Pan et al. 2010; Perera et al. 2011). CO₂ injection was performed, maintaining the injection pressure at a steady state at each injection pressure (6, 7, 8, 9, 10, 12 and 14 MPa). The corresponding pressure developments at intermediate points along the sample and downstream were monitored and recorded using an advanced data acquisition system.

207 The permeability of the coal sample was then calculated using the pressure decay between
 208 the upstream and downstream vessels through the specimen calculated using Eqs. [1] and [2] (Pan
 209 et al. 2010; Wang et al. 2015):

$$210 \quad \frac{(P_u - P_d)}{\Delta P_0} = e^{-\alpha t} \quad [1]$$

$$211 \quad \alpha = \frac{kA}{\mu\beta L} V_s \left(\frac{1}{V_u} + \frac{1}{V_d} \right) \quad [2]$$

212 where, P_u is upstream vessel pressure, P_d is downstream vessel pressure, ΔP_0 is the step change of
 213 pressure in vessels at time = 0, t is time, k is permeability of the specimen, A is cross-section area of
 214 the sample (0.0324 m²), L is the length of the sample (937 mm for this study after full
 215 consolidation), μ is the viscosity of the injecting fluid, β is the adiabatic compressibility of the
 216 injecting fluid, V_s is the sample volume, and V_u and V_d are the volume of upstream and downstream
 217 pressure vessels, respectively. The viscosity and the adiabatic compressibility were calculated using
 218 the REFPROP database (McLinden et al. 1998) for respective CO₂ pressures and a constant
 219 temperature (38 °C). The pressure decay curves and corresponding permeability values of the
 220 sample for 17 MPa axial stress are shown in Fig. 6.

221

Fig 6. (a) Pressure decay curves (for 20 hours) and (b) CO₂ permeability variation with CO₂ injection pressures for 17 MPa axial stress

2.3 Coal matrix swelling measurements

During each test, the sample length variation was recorded using a linear variable differential transformer (LVDT) at 1s intervals to quantify the CO₂ adsorption-induced coal matrix swelling in the RC specimen. The volumetric swelling was then calculated using the measured length change of

the sample using Eq. [3], because movement in the radial direction was restrained in the tests by the steel casing. When the sample was subjected to swelling, the sample length was accordingly increased and for a shrinkage, the sample length was consequently decreased.

$$\text{Volumetric strain of the coal sample} = \frac{L_t - L_o}{L_o} \times 100\% \quad [3]$$

where, L_t is the sample length at time t and L_o is the sample length at time $t = 0$.

3 Results and discussion

A series of permeability tests was conducted for seven different CO₂ injection pressures under 17 MPa axial stress using a macro-scale RC low rank coal sample, and the results obtained are discussed in the following sections.

3.1 Carbon dioxide flow along the coal sample

3.1.1 Effect of injected carbon dioxide properties on coal mass permeability

According to Fig. 6(b), CO₂ permeability reduces with increasing CO₂ injection pressure at 17 MPa axial stress, and the reduction is relatively higher for super-critical CO₂. For example, increasing the injection pressure from 6 to 7 MPa sub-critical CO₂ flow caused around 11% permeability reduction and increasing the injection pressure from 8 to 9 MPa super-critical CO₂ flow caused a much larger permeability reduction of around 38%. As explained previously, CO₂ adsorption-induced coal matrix swelling is the main causative factor for this observed permeability reduction with increasing injection pressure. Similar results were reported by Ranathunga et al. (2015) for 11 MPa axial stress following similar macro-scale tests using the same type of RC coal samples, and a comparison of CO₂ permeability variations at 11 and 17 MPa axial stresses with respective to volumetric strain is presented in Fig. 7.

269

270 Fig 7. CO₂ permeability vs. coal mass volumetric strain variation under 11 MPa (Ranathunga et al.
 271 2015) and 17 MPa axial stresses (Blue data labels denote sub-critical and green data labels denote
 272 super-critical CO₂ injections)

273 According to Fig. 7, CO₂ permeability and the volumetric swelling of the specimen follows
 274 a negative exponential variation at both axial stresses, confirming the reason for the observed
 275 reduced CO₂ flow-ability with injection pressure. According to Day et al. (2010), higher adsorption
 276 capacity of CO₂ at higher pressures results in greater matrix alterations, causing reduced
 277 permeabilities. Fig. 8 shows the variation of CO₂ properties of viscosity and compressibility with
 278 increasing CO₂ pressure at 38 °C (the temperature used for the current experiment). According to
 279 Fig. 8, CO₂ properties are significantly changed in the super-critical region, where significant
 280 increments in viscosity and reductions in compressibility with increasing pressure can be seen. This
 281 inordinate increment of viscosity (see Figs. 8(a) and (c)) provides higher fluidity for super-critical
 282 CO₂, which increases the adsorption capacity of the coal mass, and the reduction of compressibility
 283 of super-critical CO₂ (see Fig. 8(b)) constrains the amount of CO₂ molecules entering the coal
 284 matrix. These are the reasons for greater coal mass swelling and the lower permeabilities obtained
 285 with increasing CO₂ injection pressures.

Fig 8. Variation of CO₂ properties (a) viscosity and (b) compressibility with pressure (McLinden et al. 1998) (The shaded area is under super-critical conditions)

3.1.2 Effect of coal seam depth on permeability

According to Fig. 7, a reduction of permeability from 11 to 17 MPa axial stress increment can be observed, similar to previous studies using small coal samples (Ranathunga et al. 2016; Vishal and Singh 2015). The disruptions to the flow with increased effective stresses cause these permeability decreases. However, the CO₂ flow reduction is comparatively less for super-critical CO₂ injection than for sub-critical CO₂. For example, a reduction of around 86% for 6 MPa sub-critical CO₂ injection and around 80% for 8 MPa super-critical CO₂ injection can be seen when the axial stress is increased from 11 to 17 MPa. Similar results were obtained by Ranathunga et al. (2016) for small coal samples of similar low rank coal and these researchers observed 82% and 77% decrease of permeability for 6 MPa (sub-critical) and 9 MPa (super-critical) CO₂ injections for 11 to 17 MPa confining pressure increase. This may be due to the already reduced flow with higher coal mass swelling and higher effective stress application in deeper coal seams that lead only to a lower reduction of super-critical CO₂ flow. This suggests that that, regardless of sample size, the effect of seam depth on its permeability is significant.

As discussed in Section 3.1.1, an increase of CO₂ injection pressure from 6 to 7 MPa (sub-critical flow) and 8 to 9 MPa (super-critical flow) cause a permeability reduction of 11 % and 38% respectively for 17 MPa axial stress. For similar CO₂ injection pressure increments during 11 MPa axial stress, Ranathunga et al. (2015) observed around 5% (6 to 7 MPa) and 23% (8 to 9 MPa)

307 permeability reductions for similar macro-scale RC samples made with the same brown coal. This
308 implies that the permeability reduction with increasing injection pressure observed at 17 MPa axial
309 stress is higher than the reduction observed at 11 MPa axial stress (Ranathunga et al. 2015). Similar
310 results have been obtained for a meso-scale study by Ranathunga et al. (2016) for similar rank coal
311 samples, which showed around 2% and 11% permeability reductions for 11 and 17 MPa confining
312 pressures when CO₂ injection pressure was increased from 6 to 7 MPa, and 19% and 30%
313 permeability reductions when CO₂ injection pressure was increased from 8 to 9 MPa at the same
314 confining pressures (see Table 1). The reason is the lower flow ability at higher applied effective
315 stresses, which offers a longer residential time for the CO₂ molecules within the coal mass. This
316 may lead to higher matrix alterations causing lower permeability, especially for super-critical CO₂
317 flow (Vishal and Singh 2015).

318 The other important fact is that the coal matrix swelling is comparatively less at 17 MPa
319 than for 11 MPa axial stress (see Fig. 7). For example, around 0.045% and 0.036% volumetric
320 strains can be seen for 6 MPa (sub-critical) CO₂ flow and 0.053% and 0.046% for 8 MPa (super-
321 critical) CO₂ flow under 11 MPa and 17 MPa axial stresses, respectively (see Fig. 7). This may be
322 due to the obstruction of sample length variation by the higher stresses applying at higher axial
323 stresses (recall Section 2.3). In addition, this volumetric strain increment with injection pressures is
324 higher for 11 MPa axial stress compared to 17 MPa axial stress. For instance, a 6 to 7 MPa sub-
325 critical CO₂ flow increment shows around 7.2% and 4% volumetric strain increases for 11 and 17
326 MPa axial stresses, while that for 8 to 9 MPa super-critical flow is around 16.1% and 12.6%,
327 respectively (see Fig. 7). Similar results have been reported by Jasinge (2010) for RC low rank coal
328 of a similar type. Jasinge (2010) observed around 8.9% and 3.8% swelling increments for 9 to 10
329 MPa confining pressure increments when CO₂ pressure was increased from 2.5 to 3.4 MPa. As
330 explained previously, the higher effective stresses acting on the coal mass at greater axial stresses
331 slow down the CO₂ flow along the coal matrix, offering less opportunity for CO₂ to interact with
332 the coal mass by increasing the matrix rearrangements. Hol et al. (2011) also confirmed this

333 observation after finding a lower CO₂ sorption capacity for bituminous coal due to the higher in situ
334 stresses applied on coal mass which will result in lesser matrix swelling. This shows the lower coal
335 matrix swelling effect expected for CO₂ injection at greater seam depths. This is preferable for CO₂
336 sequestration as most of the potential seams for CO₂ sequestration process are at quite deep depths
337 (> 1 km).

338 3.1.3 *How does carbon dioxide flow behaviour vary along the seam?*

339 As discussed in Sections 3.1.1 and 3.1.2, lower CO₂ injection pressures at shallow depths produce
340 higher permeability through the coal mass, while higher CO₂ pressures at deeper depths cause flow
341 reduction with enhanced coal matrix swelling and higher effective stresses applied on the coal mass.
342 The meso-scale study conducted by Perera et al. (2011) for black coal (high rank), showed a
343 considerable reduction in pressure along the coal mass during CO₂ injection. For example, around 6
344 MPa downstream pressure was observed for 10 MPa CO₂ injection under 15 MPa confinement and
345 around 3 MPa for 20 MPa confinement for the same injection pressure. Similar lower pressure
346 gradients were observed by Ranathunga et al. (2015) for a low rank macro-scale coal sample, and
347 non-linear pressure variation along the specimen could be seen. Therefore, it was interesting to
348 study how this CO₂ flow varies through the coal sample length (937 mm for the current study)
349 during each injection conditions under this greater axial stress condition. Fig. 9 illustrates how the
350 CO₂ pressure varied along the length of the sample during 6 MPa (sub-critical) and 8 MPa (super-
351 critical) CO₂ injections at both 11 and 17 MPa axial stresses at the end of injection period measured
352 using intermediate pressure transducers (see Fig. 4). The injection period was the duration of time
353 taken to reach to the steady state at downstream and the additional 10 days (approximately) allowed
354 for swelling-induced matrix changes.

355 As shown in Fig. 9, the respective injection pressures dropped gradually from upstream to
356 downstream, regardless of injection pressure or depth (axial stress). The important thing is that this
357 decrease is greater at higher injection pressures (super-critical) and higher depths compared to
358 lower pressures and shallow depths. For example, 6 MPa sub-critical CO₂ flow under 11 MPa axial

359 stress caused a decrease of 21.4% from upstream to downstream, while the same injection under 17
360 MPa axial stress caused a greater reduction of 32.9%. In addition, 8 MPa super-critical CO₂ flow
361 under 11 MPa axial stress produced a 29.8% drop in pressure, and 17 MPa axial stress caused a
362 greater reduction of 43.6%. These lower pressure gradients caused lower permeabilities through the
363 sample (Fig. 7). Interestingly, the injected pressure remained constant up to around 500 m, 200 m,
364 400 m and 50 m during 6 MPa and 8 MPa CO₂ injection under 11 MPa and 17 MPa axial stress
365 respectively. Nevertheless, this length reduced with increasing injection pressure and axial stress.
366 Hence, it can be deduced that the critical zone of influence (area of CO₂ injection pressure kept
367 constant) during CO₂ injection into a coal seam is larger at lower injection pressures at shallow
368 depths, while it is reduced when higher CO₂ pressures are injected into deeper coal seams. This is
369 important for the CO₂ storage process in deep coal seams.

370 Furthermore, only around 50% of the sample is under super-critical conditions under 11
371 MPa axial stress for 8 MPa CO₂ injection, and this proportion become less (around 25% of the
372 sample) under 17 MPa axial stress (see Figs. 9(c) and (d)). This observation clearly indicates the
373 lower volumetric strain increments at higher effective stresses (refer to Section 3.1.2). The reason is
374 that the coal matrix alterations due to more adsorptive super-critical CO₂ occur over a smaller area
375 of the sample due to the lower permeabilities at higher effective stresses. Hence, the volumetric
376 strain increment is lesser at greater depths than shallow depths.

Fig 9. Pressure variation along the length of the sample after injection period under 11 MPa (Ranathunga et al. 2015) and 17 MPa axial stresses (Here, P_i is injection pressure and σ_a is axial stress. The colour scale of each figure depicts the CO_2 pressure (in MPa) variation along the coal sample)

382 3.1.4 An empirical relationship for carbon dioxide flow variation along a low rank coal seam

383 As CO₂ permeability along the coal sample varies with both axial stress and CO₂ injection pressure,
 384 a multi-variable regression equation was developed for permeability variations using the
 385 experimental data from Ranathunga et al. (2015) and the present study. Refer to Eq. [4]:

$$386 \quad k = e^{(-0.2944\sigma_a - 0.1105P_i - 7.9127)} \quad R^2 = 0.936 \quad [4]$$

387 where, σ_a is axial stress (MPa), P_i is CO₂ injection pressure (MPa) and k is CO₂ permeability (mD).
 388 The predicted permeability variations with the axial stresses and CO₂ injection pressures are shown
 389 in Fig. 10. Fig. 10 clearly indicates that CO₂ permeability is higher at lower axial stresses and lower
 390 CO₂ injection pressures. In addition, it is reduced gradually with the increment of both axial stress
 391 and CO₂ injection pressure, confirming the experimental observations.

392
 393 Fig 10. Predicted permeability variation with axial stress and CO₂ injection pressure

394 3.2 CO₂ storage capacity variation

395 One of the main purposes of CO₂-ECBM process is to store anthropogenic CO₂ in deep un-
 396 mineable coal seams. It is therefore vital to investigate the amount of CO₂ that can be stored in the
 397 coal mass under various conditions, including various seam depths (axial stress) and injecting fluid
 398 properties (CO₂ phase and pressure). The next stage of analysis was therefore to quantify the CO₂

399 storage capacity in the coal mass under the various test conditions. This was accurately monitored
 400 using a digital platform scale at the CO₂ injection point. The amount of gas injected was monitored
 401 over time after changing the injection pressure and maintaining it for more than 10 days, to allow
 402 sufficient time for swelling. Fig. 11 shows the variation in amount of injected CO₂ into the coal
 403 mass (after around 10 hours) with effective stress under 11 MPa and 17 MPa axial stress conditions
 404 at each injection pressure. The effective stress was calculated as the difference between the applied
 405 axial stress and the mean gas pressure (the average of the pressures applied at upstream and
 406 downstream).

407
 408 Fig 11. Variation of injected CO₂ weight (after 10 days) and CO₂ permeability with effective stress
 409 for 11 and 17 MPa axial stresses (The blue data labels denote the sub-critical and the green data
 410 labels denote the super-critical CO₂ injections).

411 According to Fig. 11, the amount of injected CO₂ is increased with increasing effective
 412 stress for both 11 and 17 MPa axial stresses. This increment is greater when the CO₂ injection
 413 pressure is greater than 7 MPa (super-critical flow) and lower for 6 to 7 MPa (sub-critical flow)
 414 CO₂ injections. For example, around 4.9% increment in injected CO₂ weight was observed from 6
 415 to 7 MPa pressure increment, and that for 8 to 9 MPa was 14.9% at 11 MPa axial stress. Similarly,
 416 for 17 MPa axial stress, the injected CO₂ weight was around 1.9% for an increase from 6 to 7 MPa
 417 pressure and 9.8% for 8 to 9 MPa pressure increment. The main reason for this observation is the
 418 reduction of CO₂ flow with increasing injection pressure (see Fig. 7) due to the coal matrix

rearrangements which require less CO₂ injection into the coal sample. Furthermore, the higher the axial stress, the greater the effective stress application, which causes greater reductions in CO₂ injectivity (see Fig. 10).

However, this CO₂ storage process is time-dependent, and longer time offers more opportunity for CO₂ adsorption and storage. Fig.12 shows the injected CO₂ weight variation with time at 6 MPa (sub-critical) and 8 MPa (super-critical) injection pressures under both 11 and 17 MPa axial stresses. According to Fig. 12, the cumulative weight of injected CO₂ increases over time, and the rate of increase is lower at higher CO₂ injection pressures and at greater axial stresses. For example, the average increases are around 0.0031 and 0.0025 at 6 and 8 MPa CO₂ injection pressures under 11 MPa axial stress, while they are around 0.0008 and 0.0005 for the same injection pressures (6 and 8 MPa) under 17 MPa axial stress. This observation clearly explains the CO₂ storage behaviour observed in Fig. 11, which shows greater CO₂ storage capacity at lower depths and lower CO₂ injection pressures.

Fig 12. Cumulative weight of injected CO₂ variation with time for 6 MPa and 8 MPa injection pressures at 11 MPa and 17 MPa axial stresses

Interestingly, in relation to the weight of CO₂ injected at the end of the injection period, a contrasting behaviour compared to that shown in Fig. 11 was obtained. Fig. 13 illustrates a comparison of the weight of CO₂ injected after 10 days of injection and at the end of the injection

438 period under both 11 and 17 MPa axial stresses. According to Fig. 13, it can be noted that, although
 439 flow ability reduces at higher CO₂ injection pressures, a comparatively larger amount of CO₂ can be
 440 stored if a long time is offered for CO₂ sequestration at greater depths. The reason is, as stated in
 441 Fig. 8, the inherent liquid-like properties of super-critical CO₂ permit greater adsorption potential
 442 with the coal matrix, which requires some time to complete.

443
 444 Fig 13. Comparison of weight of CO₂ injected after completing the injection period and after 10
 445 days during (a) 11 MPa axial stress and (b) 17 MPa axial stress (The data labels denote the injection
 446 period (in hours) for each injection pressure)

447 However, in the case of CO₂ storage capacity variation with axial stress, the injected CO₂
 448 weight reduction with axial stress is not much lower compared to the permeability reduction
 449 observed with axial stress (see Fig. 7). A comparison of the observed permeability reductions and
 450 the CO₂ storage capacity reductions under 11 to 17 MPa axial stress increment is shown in Table 2.

451 Table 2. Comparison of permeability and CO₂ injectivity reductions with 11 MPa to 17 MPa axial

	CO ₂ injection pressure (MPa)	Permeability reduction (%)	Injected CO ₂ weight reduction (%)	452 stress
				453 es
Sub-critical region	6	86.72	27.08	454
	7	85.96	25.01	
Super-critical region	8	83.98	22.99	455
	9	80.31	19.75	
	10	77.06	14.67	

456

457

458 According to Table 2, similar to permeability reduction, the injected CO₂ or stored CO₂
459 weight is greatly reduced in the super-critical region, and the permeability reduction is around
460 60~70% higher than the injected CO₂ weight reductions (see Table 2). The higher residential time
461 for CO₂ molecules with higher effective stresses may offer greater adsorption possibility for CO₂
462 molecules with the coal matrix (De Silva 2013) with time and the increased amount of CO₂
463 molecules enter to the coal mass at higher CO₂ pressures, warranting lower reductions than in flow
464 ability.

465 **4 Suggestions for CO₂-ECBM field applications**

466 Potential coal seams for CO₂-ECBM are located deep underground, where the injected CO₂ is in its
467 super-critical state (beyond 7.38 MPa and 31.8 °C). The liquid-like viscosities (see Fig. 8) of super-
468 critical CO₂ provide more potential for stable CO₂ storage within the coal matrix. Nevertheless, the
469 observed decrease in CO₂ flow ability along the coal matrix, especially during super-critical CO₂
470 injection, raises concerns concerning the productivity of the ECBM process. On the other hand,
471 according to the findings, regardless of the permeability reductions, CO₂ storage capacity in coal
472 increases with increasing injection pressure upon offering sufficient time for sequestration.
473 Furthermore, a comparatively lower reduction of CO₂ storage capacity was witnessed with
474 increasing axial stress from 11 MPa (representing an approximately 400 m deep coal seam) to 17
475 MPa (representing an approximately 600 m deep coal seam). This information is very useful for
476 CO₂ sequestration field projects, because there is current interest in utilising extremely deep
477 underground coal seams for CO₂ sequestration. According to current findings, this depth effect does
478 not have such a great influence on CO₂ storage potential.

479 However, attention should be paid to the flow reductions which occur with the coal matrix
480 swelling, because this might affect long-term CO₂ sequestration processes by allowing more CO₂

adsorption, which has been witnessed in several field-scale projects (White et al. 2005). Therefore, the implementation of flow-enhancement techniques (e.g. hydro fracturing, alternative injection of N₂) (White et al. 2005) will provide more opportunities to store greater amounts of CO₂ in deeper coal seams.

Hence, resolving complications when using super-critical CO₂ for CO₂-ECBM recovery in terms of CO₂ storage capacity and CH₄ production enhancement using flow-enhancement techniques is essential prior to field-scale projects.

5 Conclusions

Following a series of CO₂ permeability tests using macro-scale low rank coal specimens, the following conclusions can be drawn:

- CO₂ permeability in coal is greater at lower depths and lower CO₂ injection pressures, while it gradually reduces with increasing depth and injection pressure, and the associated effective stress variation and the coal matrix swelling caused by CO₂ adsorption are the main causative factors, respectively.
- Super-critical CO₂ causes greater swelling in the coal mass compared to sub-critical CO₂, and the swelling increases with increasing injection pressure, regardless of depth. This swelling increment is however reduced with increasing seam depth, probably due to the associated greater effective stresses effect that obstructs CO₂ flow along the coal matrix, offering less opportunity for CO₂ to interact with the coal mass.
- The permeability variation along the coal sample under the tested coal seam and injecting CO₂ conditions can be effectively represented using a simple multivariable regression model, and such models play an important role in field projects to predict CO₂ flow migration along the seam.
- The observed CO₂ permeability along the tested coal specimen indicated that the critical zone of influence for CO₂ injection into a selected coal seam is greater at lower injection pressures and at shallow depths, and it this reduces with increasing CO₂ pressure and seam depth.

➤ Finally, the observed CO₂ storage capacity of the tested coal under various conditions revealed that CO₂ storage capacity in a coal seam is greater at lower depths and higher CO₂ injection pressures. However, the reduction of CO₂ storage capacity with depth was not significant, which is important for field CO₂ sequestration projects, which normally use deep seams to store CO₂.

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