

Review

# A Comprehensive Overview of CO<sub>2</sub> Flow Behaviour in Deep Coal Seams

Mandadige Samintha Anne Perera <sup>1,2</sup> 

<sup>1</sup> Department of Infrastructure Engineering, Room 209B, The University of Melbourne, Building 175, Melbourne, VIC 3010, Australia; samintha.perera@unimelb.edu.au; Tel./Fax: +61-3-9035-8649

<sup>2</sup> Deep Earth Energy Laboratory, Department of Civil Engineering, Monash University, Building 60, Melbourne, VIC 3800, Australia

Received: 15 March 2018; Accepted: 9 April 2018; Published: 12 April 2018



**Abstract:** Although enhanced coal bed methane recovery (ECBM) and CO<sub>2</sub> sequestration are effective approaches for achieving lower and safer CO<sub>2</sub> levels in the atmosphere, the effectiveness of CO<sub>2</sub> storage is greatly influenced by the flow ability of the injected CO<sub>2</sub> through the coal seam. A precious understanding of CO<sub>2</sub> flow behaviour is necessary due to various complexities generated in coal seams upon CO<sub>2</sub> injection. This paper aims to provide a comprehensive overview on the CO<sub>2</sub> flow behaviour in deep coal seams, specifically addressing the permeability alterations associated with different in situ conditions. The low permeability nature of natural coal seams has a significant impact on the CO<sub>2</sub> sequestration process. One of the major causative factors for this low permeability nature is the high effective stresses applying on them, which reduces the pore space available for fluid movement with giving negative impact on the flow capability. Further, deep coal seams are often water saturated where, the moisture behave as barriers for fluid movement and thus reduce the seam permeability. Although the high temperatures existing at deep seams cause thermal expansion in the coal matrix, reducing their permeability, extremely high temperatures may create thermal cracks, resulting permeability enhancements. Deep coal seams preferable for CO<sub>2</sub> sequestration generally are high-rank coal, as they have been subjected to greater pressure and temperature variations over a long period of time, which confirm the low permeability nature of such seams. The resulting extremely low CO<sub>2</sub> permeability nature creates serious issues in large-scale CO<sub>2</sub> sequestration/ECBM projects, as critically high injection pressures are required to achieve sufficient CO<sub>2</sub> injection into the coal seam. The situation becomes worse when CO<sub>2</sub> is injected into such coal seams, because CO<sub>2</sub> movement in the coal seam creates a significant influence on the natural permeability of the seams through CO<sub>2</sub> adsorption-induced swelling and hydrocarbon mobilisation. With regard to the temperature, the combined effects of the generation of thermal cracks, thermal expansion, adsorption behaviour alterations and the associated phase transition must be considered before coming to a final conclusion. A reduction in coal's CO<sub>2</sub> permeability with increasing CO<sub>2</sub> pressure may occur due to swelling and slip-flow effects, both of which are influenced by the phase transition in CO<sub>2</sub> from sub- to super-critical in deep seams. To date, many models have been proposed to simulate CO<sub>2</sub> movement in coal considering various factors, including porosity, effective stress, and swelling/shrinkage. These models have been extremely useful to predict CO<sub>2</sub> injectability into coal seams prior to field projects and have therefore assisted in implementing number of successful CO<sub>2</sub> sequestration/ECBM projects.

**Keywords:** deep coal seams; natural permeability; CO<sub>2</sub> flow; effective factors; flow models

## 1. Introduction

Most of the solar radiation coming towards the Earth's surface is reflected back as shortwave or longwave radiation by the Earth's surface. However, some gases called "greenhouse gases" in the lower

atmosphere behave as a blanket for this longwave radiation, because these gases adsorb and re-emit the solar radiation back towards the Earth's surface within the thermal infrared range. This is commonly known as the "greenhouse gas effect" and results in the increasing global temperature effect called "global warming", one of the most debated topics among researchers. The dangerous consequences of global warming have now been reported worldwide [1]. According to Le-Treut et al. [2], if no greenhouse gas effect existed, the average world temperature would be around 33 °C lower than the present temperature. During the last century, the average world temperature has risen by around 1 °C as a result of global warming [3] and this number is continuously rising with the human activities associated with rapid industrial development. The use of fossil fuels and deforestation are the main causes of increased atmospheric CO<sub>2</sub> levels and to date around 35% increment in CO<sub>2</sub> levels has been recorded in the industrialized areas of the world [2]. Global warming has therefore become a challenging issue which requires the contribution of scientists to overcome it. As a result, a diverse range of greenhouse gas mitigation and global warming control techniques have been tested throughout the world.

The most common types of greenhouse gases are carbon dioxide (CO<sub>2</sub>), methane, and nitrous oxide, and the most abundant greenhouse gas in the atmosphere is CO<sub>2</sub>, as it is a by-product of many industrial applications, such as coal-fired power generation. Therefore, scientists' main concern has been attracted by the mitigation of atmospheric CO<sub>2</sub> levels, as every day the entire world is releasing vast amounts of CO<sub>2</sub> into the world's atmosphere.

Of the suitable CO<sub>2</sub> mitigation initiatives, CO<sub>2</sub> capture and storage (CCS) has been shown to offer a viable path to reduce the CO<sub>2</sub> emissions into the atmosphere. According to scientific predictions, it is necessary to capture and store CO<sub>2</sub> at a 5.1 Gt annual rate by 2050, which is 14% of the total needed for global temperature stabilisation [4]. According to the ETP BLUE Map scenario [5], global CO<sub>2</sub> emissions can be reduced by 50% in 2015 with emission reduction strategies at a cost of up to US \$200 per 1 ton of CO<sub>2</sub>. This includes around 19% CO<sub>2</sub> emission reduction through CCS and if CCS were not employed, the annual cost for the emission reduction in 2050 would be increased by around 71% [6].

In the CCS process, deep brine aquifers are identified as suitable geological structures, in which CO<sub>2</sub> can be stored, since they are widely distributed and have large storage capacity in their pores [7,8]. Also, CO<sub>2</sub> geo-sequestration in subsurface systems has been identified as an effective option, due to its ability to release tightly bonded productive gas, largely methane, from the reservoir. Importantly, CO<sub>2</sub> geo-sequestration in deep coal seams can be considered as economical way of reducing the atmospheric carbon content when consider its ability to enhance the coal seam gas production while storing carbon dioxide and when the two process considered together is called enhanced coal bed methane recover (ECBM). However, CO<sub>2</sub> behaviour in coal seams is quite complicated due to its chemically reactive nature with the coal matrix and the associated modifications of the coal mass chemical and physical structure. These factors have led to unpredictable CO<sub>2</sub> injectivity into deep coal seams after CO<sub>2</sub> injection in field CO<sub>2</sub> sequestration projects [9,10], and the potential phase transition of injected CO<sub>2</sub> in the seam (from liquid or gas to super critical CO<sub>2</sub>) has caused these complications related to CO<sub>2</sub> sequestration/ECBM to become more critical [3]. The aim of this paper is to provide a comprehensive review of current findings on CO<sub>2</sub> flow behaviour in deep coal seams under various in-situ conditions.

## 2. Natural Permeability of Preferable Coal Seams for CO<sub>2</sub> Sequestration

Since permeability quantifies flow ability through any coal seam [11], it is clearly one of the most critical parameters affecting CO<sub>2</sub> sequestration in coal, in terms of CO<sub>2</sub> injectability. Generally, natural coal seams have quite low permeability values, which significantly affect the CO<sub>2</sub> sequestration process in the seam. As a result, precise prediction of permeability is necessary for the effective planning of any CO<sub>2</sub> sequestration process. The permeability of natural coal seams is influenced by a broad range of factors, including the geological stress field of the coal seam, the seam temperature and its moisture content, and the degree of maturity or coal rank.

It is important to understand the permeability measurement techniques used by different researchers for different types of coal seam. Several techniques have been used to precisely measure the permeability values of coal samples under laboratory conditions. Tri-axial experiments on undisturbed core samples is the common technique as it approximately represents the in situ conditions and natural porosity and cleat system of the coal seam. Pulse decay method is an extended technique, which measures the effective permeability and diffusivity of adsorptive gases. Several other techniques such as gas expansion techniques, onsite drill-core desorption tests, mercury intrusion curves have been used for the permeability measurements under different experimental conditions [12,13]. Moreover, stochastic simulations are widely used for coal sample characterization and permeability evaluation. Using advanced techniques like X-ray imaging and modelling, the heterogeneity and the complexity of coal seam can be accurately evaluated and thus the permeability can be predicted [14,15].

The pore space or porosity of the coal mass is the main factor governing its flow ability or permeability, because highly porous media have higher tendency to offer easy flow paths for fluid movement in coal. A direct relationship between coal permeability and porosity has been proposed in existing studies (Equation (1)), and the  $n$  value can be theoretically taken as 3 [16], although it may exceed 3 in actual situations [17]:

$$\frac{k}{k_o} = \left( \frac{\varnothing}{\varnothing_o} \right)^n \quad (1)$$

where  $k$  is the coal permeability,  $k_o$  is the initial permeability,  $\varnothing$  is the seam porosity,  $\varnothing_o$  is the initial porosity and  $n$  is a constant.

The main factor that controls porosity and therefore permeability in a reservoir is the magnitude of the net stress or effective stress acting on the coal mass. Effective stress can be predicted based on the simple force-balance between the confining pressure and the pore pressure (Equation (2)), as pore pressure and confining pressure have opposite effects on the pore volume:

$$P_d = P_c - \alpha P_p \quad (2)$$

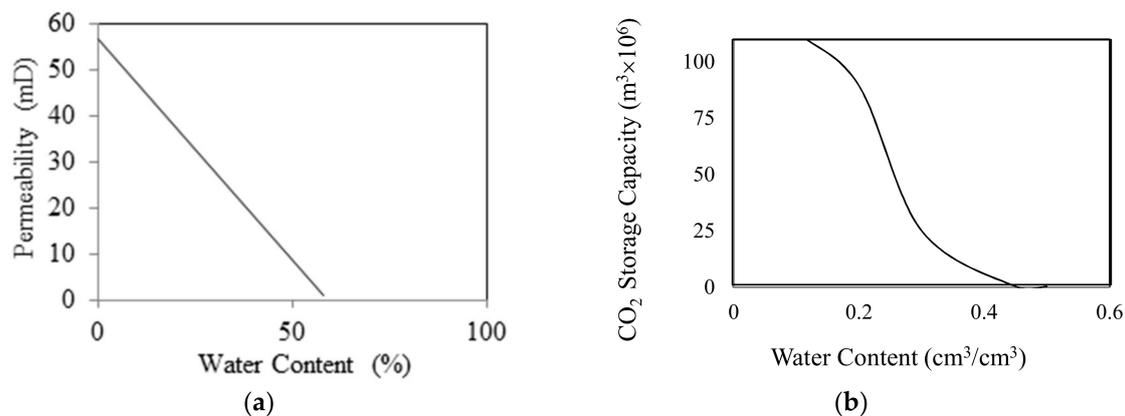
where  $P_d$  is the differential pressure,  $P_c$  is the confining pressure,  $P_p$  is the pore pressure and  $\alpha$  is the effective stress coefficient.

Being a type of relatively weak and compressible rock, coal's pore structure varies greatly with the changing of the applying effective stresses on it. According to Somerton et al. [18], increasing the effective stress acting on any coal mass causes the fissures and pores of that coal body to close, which increases the tortuosity for fluid flow movement inside the coal mass and therefore reduces the permeability. In Somerton's et al. [18] experimental study on a range of coal samples (with 0.1 and 100 mD permeability), increasing the effective stress upon load application during coal failure caused the permeability to be reduced by 1 to 2 orders of magnitude and effective stress was found to have a negative exponential relationship with coal permeability. Interestingly, permeability of high rank coal is more greatly affected by effective stress changes compared to low rank coal, where low rank coal show about an order of permeability reduction with the effective stress increment and high rank coal show two orders of permeability reduction. This negative exponential relationship between coal permeability with effective stress has been confirmed by many researchers, including Durucan and Edwards [19]. According to the experimental results of Durucan and Edwards [19], permeability reduction in coal with effective stress increment exhibits an initial steep gradient followed by a gentle gradient and eventually reaches a residual value. According to these researchers, this initial steep gradient is due to the immediate closure of existing micro-fractures under very low stresses.

It is known that the effective stress applying on any coal seam is increasing with increasing the seam depth, as it causes to generate greater confining stresses on the seam. Deeper coal seams therefore exhibit much lower permeability compared to shallow seams. Most of the deep coal seams normally have high temperatures and a greater amount of water in their pores, thus it is generally expected

to have higher degree of water saturations. Therefore, the combined effects on permeability need to be considered.

In relation to coal permeability variation with moisture content, the coal mass tends to shrink with the reduction of its moisture content, creating permeability enhancement in the coal seam [20]. The moisture in the coal mass pore structure occupies the pore space available for fluid movement and the adsorption of that water in the coal matrix induces significant matrix swelling, both of which obstruct the flow paths available in the coal matrix and therefore cause the reduction of overall coal seam permeability [20]. The variation of coal seam permeability and gas storage capacity with the natural moisture content have been studied by Skawinski et al. [20] and Perera et al. [21] and the results are shown in Figure 1. Regarding the moisture content effect, deeper coal seams generally have water-saturated conditions compared with shallow seams and thus are expected to have low permeability.



**Figure 1.** Variation of coal seam permeability (a) [20], and gas storage capacity (b) [21] with natural moisture content.

In relation to the temperature effect on coal seam permeability, according to De Silva et al. [22], the average thermal gradient available underground is around 25–30 °C/km increment from the ground surface. Therefore, preferable coal seams for CO<sub>2</sub> sequestration or ECBM process (>0.8 km depth) possibly have considerably high temperatures. The high temperatures create thermal expansion in the coal matrix that reduces the available pore space and therefore its permeability [23]. In contrast, high temperature also creates thermal cracks in the coal matrix, thus increasing its pore space and permeability. Therefore, the combined effects of each of these factors must be considered in order to determine the influence of temperature on coal seam permeability.

Further, deeper coal seams normally contain higher-rank coal, as that coal has more opportunities to be subjected to greater pressure and temperature variations [24]. In simple theory, the higher the burial depth, the higher the stress applying on the coal mass. Thus, the coal mass pore structure shrinks with increasing rank, due to the increment of the stress applying on the mass, resulting in a permeability reduction. This was proved both experimentally and numerically by many researchers, in which they have shown that the permeability decreases significantly with the increase of effective stress applied on the coal mass [25–27]. This is because high-rank coal has been subjected to greater stresses and temperature variations and therefore is a tight, low porous medium with a greater proportion of micro-pores [28]. In contrast, low-rank coal has only been subjected to lower stresses and temperatures and is therefore a less compacted highly porous medium with a greater proportion of macro-pores. Therefore, the burial depth or the rank of the coal seam also has a significant influence on its permeability, and a permeability reduction trend can be expected with increasing coal rank.

On the basis of all of these facts, since deeper coal seams have greater effective stresses, higher-rank coals and water saturation condition, deep coal seams generally have very low permeability values.

However, the temperature influence is the opposite, and most available coal seams have less than 0.001 mD extremely low permeability values [29]. The variation of coal seam permeability with depth predicted by Korre et al. [30] is shown in Figure 2.

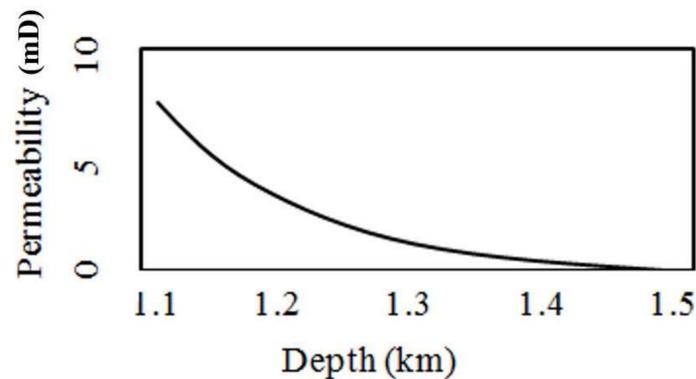


Figure 2. Variation of coal permeability with seam location [30].

### 3. CO<sub>2</sub> Injection-Created Natural Coal Seam Permeability Alterations

The permeability of a coal seam is thought to be independent of the flowing fluid properties. However, the strong dependence of coal mass permeability on the structure of its connected pore spaces causes coal permeability to be dependent on any factor that may contribute to changes in its pore structure. The significant reduction of coal mass pore space with the swelling induced by CO<sub>2</sub> injection and the mobilisation of the hydrocarbons in the seam with CO<sub>2</sub> interaction have been recorded [31]. The corresponding blockage of flow paths by the adsorbed CO<sub>2</sub>-induced swelling and the mobilized hydrocarbons from the coal matrix leads to a significant reduction in coal mass flow ability or permeability, which eventually reduces CO<sub>2</sub> injectability into the coal seam. This has been widely experienced in field CO<sub>2</sub> sequestration projects. For example, the CO<sub>2</sub> injection rate was reduced by around 50% during the first six months of CO<sub>2</sub> injection in the San Juan Basin, USA field CO<sub>2</sub> sequestration project [32], and by around 70% during the first twelve months of CO<sub>2</sub> injection in the Ishikari basin, Japan [33].

Pekot and Reeves [34] have shown how coal permeability is reduced with coal matrix swelling and the results are summarised in Figure 3. A direct relationship between CO<sub>2</sub> adsorption-induced coal matrix swelling and coal permeability has been shown by many researchers [35,36] and one of the results is shown in Figure 4.

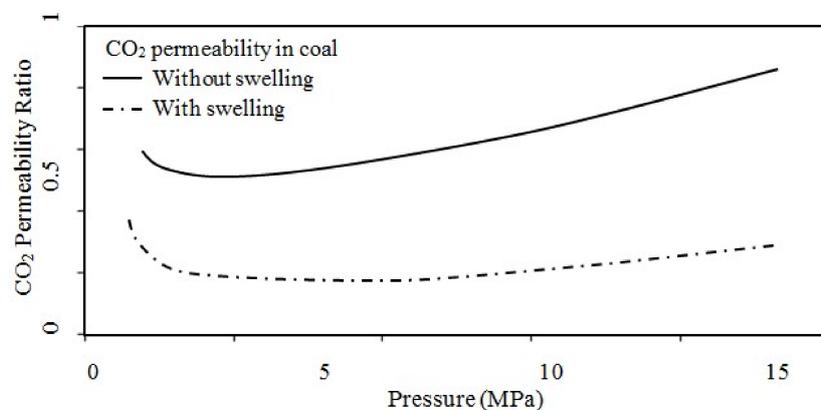
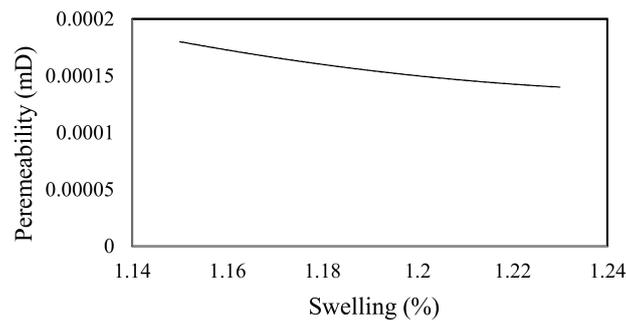
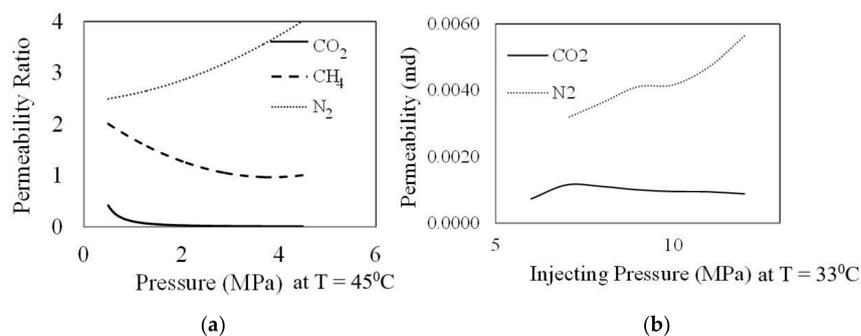


Figure 3. Change of CO<sub>2</sub> permeability in coal with swelling process and the pressure effect on it [34].



**Figure 4.** The direct relationship between coal swelling and permeability [36].

According to the figure, swelling significantly reduces the deep coal seams permeability, and therefore CO<sub>2</sub> flow behaviour in natural coal seams varies significantly from the flow behaviours of other types of gases such as N<sub>2</sub> in them. This unique flow behaviour of CO<sub>2</sub> in coal compared to other types of gases has been shown by many researchers. Cui et al. [37] analyzed the adsorption and transport of CO<sub>2</sub> and N<sub>2</sub> in coal particles with numerical modelling and found that CO<sub>2</sub> can permeate into ultra-micro pores, resulting in a higher diffusivity in coal matrix, than CH<sub>4</sub> or N<sub>2</sub>. This is due to relatively smaller kinetic diameter, linear shape and the high affinity of CO<sub>2</sub>, which allow it to permeate not only to macro pores but also to micro and ultra-micro pores as well [38,39]. Also the significantly reduced coal mass permeability upon CO<sub>2</sub> movement compared to other gases has been clearly shown (Figure 5) and CO<sub>2</sub>-induced coal matrix swelling was found to be the main cause for this reduction [3,35,40,41]. Figure 5a shows that CH<sub>4</sub> has lower permeability values compared to N<sub>2</sub> and for this reason CH<sub>4</sub> causes a considerable swelling effect in coal. However, the swelling-related issues caused by CO<sub>2</sub> are much greater than those caused by CH<sub>4</sub>. According to Larsen [42], the coal mass can swell by up to 4% of its volume due to CO<sub>2</sub> adsorption, which pressures the fractures and cleats in the coal mass, thereby reducing its permeability. For example, the Tahmoor and North Cliff Mines in the Bulli seam exhibited lower gas drainage capabilities due to the available high carbon dioxide concentrations in their coal seam gas [43].



**Figure 5.** Unique flow behaviour of CO<sub>2</sub> in coal observed by different researchers. (a) Durucan and Shi [35]; (b) Perera et al. [3].

#### 4. CO<sub>2</sub> Flow Behaviour in Deep Coal Seams

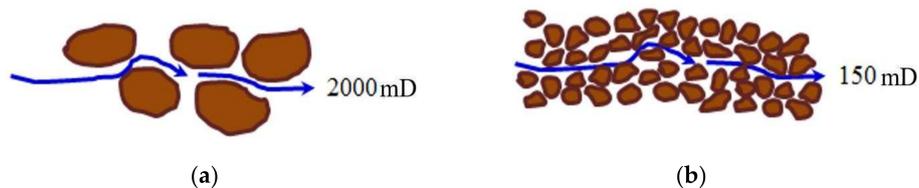
CO<sub>2</sub> has unique flow behaviour in coal due to the hydrocarbon dissolution and matrix swelling occur as results of coal—CO<sub>2</sub> interactions. However, this flow behaviour varies with the physical properties of the seam and the injected CO<sub>2</sub>.

##### 4.1. Effect of Seam Physical Properties on CO<sub>2</sub> Flow Behaviour in Coal

CO<sub>2</sub> flow behaviour in coal seams may vary greatly with seam location-related factors, such as depth, effective stress, temperature and moisture content.

#### 4.1.1. Depth

Flow through any porous medium is greatly dependent on its grain size distribution and packing arrangement, and a medium with tightly-packed fine grains has much poorer flow paths than a medium with loosely-packed large grains, as shown in Figure 6 [44]. This concept is directly related to the depth effect on coal permeability. As mentioned earlier, depending on the location or depth of the seam, the pressure and temperature change, and the grain size, shape and arrangement and pore structure change accordingly, and deeper coal seams generally have tightly-packed finer grains and lower porosities and therefore, are expected to have lower CO<sub>2</sub> flow ability through them [45].



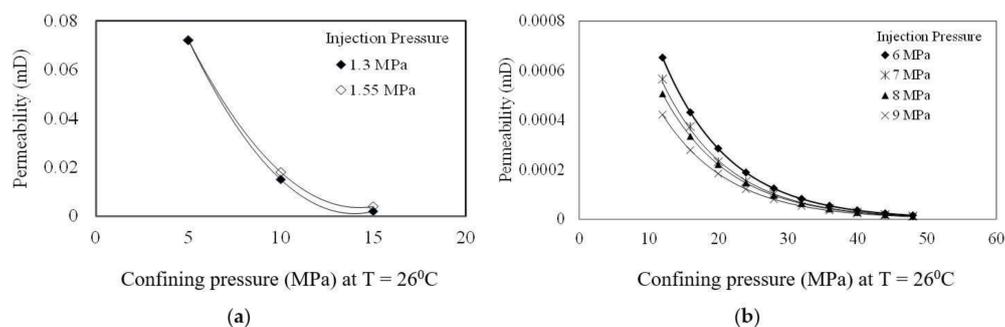
**Figure 6.** Effect of grain size and packing arrangement on permeability in porous media [44]. (a) Round large loosely-packed grains; (b) Irregular shaped small tightly-packed grains.

The effect of depth on permeability has been studied by many researchers and the term “confining stress” has been commonly used to describe the depth effect. The deeper the coal seam, the greater the effective confining stress applied on them, and the following general relation can be used to identify the confining stress at a given depth [11]:

$$P_{con} = D\rho_r g \quad (3)$$

where  $P_{con}$  is the applied confining stress,  $D$  is the depth to the formation,  $\rho_r$  is the average density of the overburden rock mass, and  $g$  is the gravitation acceleration.

The effect of confining stress on coal’s CO<sub>2</sub> permeability has been studied by many researchers. According to Siriwardane et al. [46], who studied a Pittsburgh seam coal by changing the confining stress from 20 MPa to 40 MPa and keeping the pore pressure constant, coal permeability exhibits a steep reduction trend with increasing confining pressure due to the shrinking of internal coal fractures at higher confining pressures, which eventually reduces its permeability. Vishal et al. [40] checked the confining stress effect on CO<sub>2</sub> permeability for Indian bituminous coal samples under two different pore pressure conditions and observed similar reductions in permeability for both pore pressures due to the closing of internal fractures under high confining pressures [47], and increasing the confining stress from 5 to 15 MPa caused permeability to be reduced by around 100% (Figure 7). A similar CO<sub>2</sub> permeability reduction with increasing confinement has been shown by Perera et al. [48] for Australian bituminous coal at greater confinements.



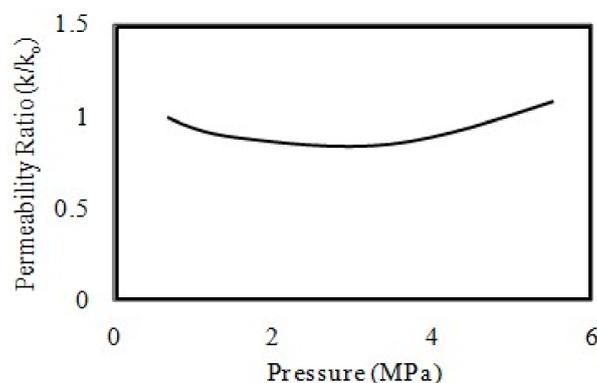
**Figure 7.** Influence of confining stress on CO<sub>2</sub> permeability in (a) Indian [40], and, (b) Australian [48] bituminous coals.

This reduced CO<sub>2</sub> permeability of coal at greater depths has also been experienced in field CO<sub>2</sub> projects. According to the field data reported by Chatterjee and Pal [49] on seven wells at different depths (52 to 200 m) in the Raniganj coalfield in India, coal permeability reduces with depth due to the steeper stress gradient at greater depths even within the same coal seam, and increasing the depth from 88.3 to 199.8 m caused coal permeability to be reduced from 10.2 mD to 0.5 mD.

According to these laboratory- and field-scale studies, the high confining pressures in deeper target seams cause reduced permeability in them. This creates serious issues for large-scale sequestration projects, as critically high injection pressures are required to achieve sufficient CO<sub>2</sub> injection into them due to the existing low confinements, which significantly increases the project cost and involves some risk associated with CO<sub>2</sub> leakage into surrounding aquifers.

#### 4.1.2. Injecting CO<sub>2</sub> Properties

Injection of CO<sub>2</sub> into a natural coal seam enhances its pore pressure and this pore pressure effect on coal permeability has been examined by many researchers. According to Palmer et al. [50] and Pekot et al. [51], in the primary recovery of coal beds, firstly coal mass permeability reduces with increasing pore pressure and then tends to increase as the adsorbed gas tends to desorb with the opening and expansion of existing cleats/fractures. A similar experimental observation on coal permeability with CO<sub>2</sub> pore pressure was reported by Liu et al. [52]. According to these researchers, increasing the CO<sub>2</sub> pore pressure inside the coal mass reduces its permeability by up to around 3.5 MPa, mainly due to coal matrix swelling, and further increasing the CO<sub>2</sub> pore pressure tends to increase permeability due to the coal matrix shrinkage caused by the high-pressure fluid (Figure 8). However, the CO<sub>2</sub> injection pressures considered in this study were quite low and relate to the sub-critical state of CO<sub>2</sub>.



**Figure 8.** Variation of coal permeability with pore pressure [52].

Depending on the pressure and temperature, CO<sub>2</sub> may exist in three main states: gas, liquid and super-critical, and under ambient conditions CO<sub>2</sub> is a thermodynamically-stable heavy gas, and when the temperature goes beyond 31.8 °C and pressure goes beyond 7.38 MPa (critical point), CO<sub>2</sub> converts to its highly chemically-reactive super-critical state. The flow behaviour of CO<sub>2</sub> is largely dependent on this phase condition. Interestingly, super-critical CO<sub>2</sub> has properties of both gas and liquid CO<sub>2</sub>, and its gas-like compressibility values lead to the easy filling of all the available volume, and its liquid-like density values that vary from 200 to 900 kg/m<sup>3</sup> depending on the pressure and temperature, lead to more stable storage. As shown in Figure 9, super-critical CO<sub>2</sub> has much greater compressibility values than liquid CO<sub>2</sub> and therefore occupies much smaller space deep underground with the potential occurrence of phase transition. As a result, it is possible to store significantly larger amounts of CO<sub>2</sub> in deep coal seams in its super-critical state and deep seams are therefore preferable for CO<sub>2</sub> sequestration.

This super-critical CO<sub>2</sub> flow behaviour under in-situ conditions has been studied by Perera et al. [3], considering a wide range of CO<sub>2</sub> injection pressures covering both the sub-and super-critical regions. According to their experimental results, increasing the CO<sub>2</sub> injection pressure causes coal’s permeability for CO<sub>2</sub> to be reduced due to the associated swelling effect that increases with increasing CO<sub>2</sub> pressure (Figure 10), and this permeability reduction is very significant for super-critical CO<sub>2</sub> movement inside the coal mass.

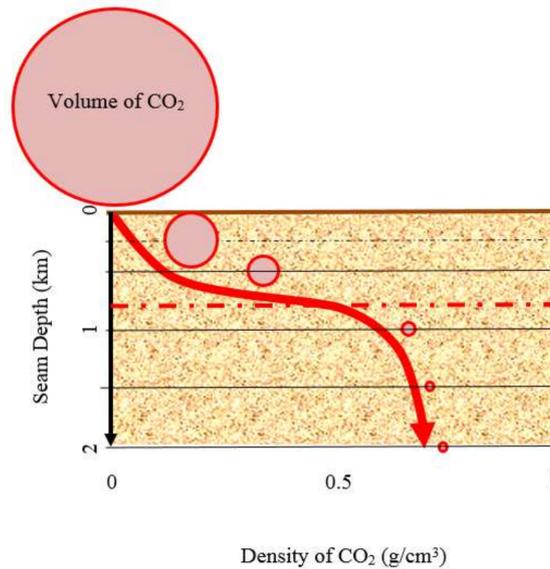


Figure 9. Variation of volume occupied by CO<sub>2</sub> with depth [53].

This greater permeability reduction is related to the greater swelling effect created by super-critical CO<sub>2</sub> in coal. This greater swelling effect on permeability caused by super-critical CO<sub>2</sub> has been shown by Perera et al. [54], and according to these researchers, the permeability reduction with super-critical CO<sub>2</sub> adsorption is about two times higher than that with sub-critical CO<sub>2</sub> adsorption, and this indicates the significantly greater swelling effect created by super-critical CO<sub>2</sub> in coal compared to sub-critical CO<sub>2</sub>. Further, they observed a reduction of the swelling effect with increasing CO<sub>2</sub> pressure, and this observation related to the opening up of coal mass pores and pushing the swelled layers by high-pressure fluid, and the effect increased with increasing pressure. Furthermore, super-critical CO<sub>2</sub> creates much stronger bonding with the coal matrix due to its highly chemically-reactive nature, which was confirmed by the observed lower reduction in the swelling effect with increasing injection pressure in super-critical CO<sub>2</sub> swelled coal samples [54].

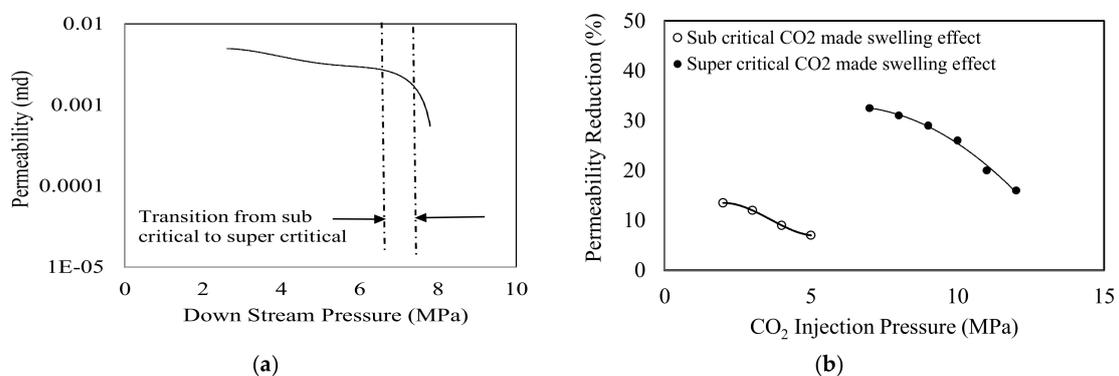
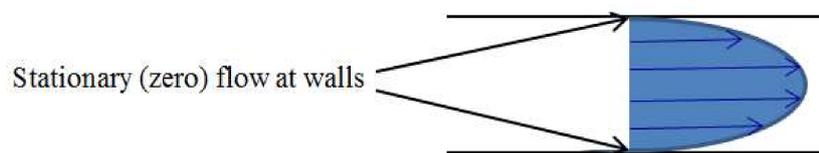


Figure 10. Variation of coal permeability with pore pressure for both sub-critical and super-critical CO<sub>2</sub>. (a) Down Stream Pressure [3]; (b) CO<sub>2</sub> Injection Pressure [54].

According to Perera et al. [55], apart from the swelling effect, the Klinkenberg slip-flow effect also has a considerable influence on CO<sub>2</sub> movement in coal. For example, in Perera's et al. [54] study, sub-critical CO<sub>2</sub> was in the gas state, compared to which super-critical CO<sub>2</sub> has low compressibility potential through coal due to the increased CO<sub>2</sub> molar weight, which reduces the Klinkenberg slip-flow effect and eventually creates comparatively lower permeability for super-critical CO<sub>2</sub>. The influence of the Klinkenberg slip-flow effect on CO<sub>2</sub> permeability has also been shown by Lama and Bodzinoy [56].

In relation to the Klinkenberg effect in porous media, it is known that gas permeability in any porous medium is much greater than liquid permeability. This suggests that the permeability of a particular pores media depends not only on the medium properties, but also on the flowing fluid properties as well. Therefore, the highly permeability nature of gas compared to liquid in porous mediums is inter-related with the medium behavior as well as flowing fluid behaviour. In fact, Klinkenberg [57] found that the reason for high permeability in the case of gas movement is due to the slip flow between gas molecules and solid walls. In ideal fluid conditions, fluid flow through boundaries is as given in the following figure (Figure 11), where flow adjacent to the boundaries has zero velocities (stable) and the center of the fluid flow has maximum velocity.



**Figure 11.** Velocity profile of fluid flow through solid boundaries under ideal conditions.

This is the condition which exists in liquid flow through the pore walls of the coal matrix, where the liquid molecules adjacent to the pore walls are almost stationary. However, this is not the situation which exists for gas flow in any medium including coal mass, in which the gas molecules adjacent to the boundary are also moving along the flow direction. During this movement, gas molecules collide with one another and with the soil boundary, and the distance a gas molecules travels before a collision with another gas molecule is called the mean free path. Normally, for any fluid movement through boundaries, collisions between fluid molecules and the walls induce friction to the flow and therefore, if there is no molecule adjacent to the boundary that frictional flow is minimal and gas molecules pass through the porous medium more easily. This is what happens when the pore size of the medium approaches the mean free path distance, and in this situation, the gas molecules flow through diffusion through the pores without any pressure differential and without being subjected to wall friction loss, this additional flow being called slip flow. The slip flow enhances the flow rate through the medium and therefore enhances the medium's permeability. This mean free path is greater at low pressures, as gases are more widely spread and shorter at higher pressures than in the condensed situation. Therefore, the slip-flow effect reduces with increasing pressure and therefore, increasing pressure causes gas permeability to be reduced in porous media.

Gas flow through coal occurs along very narrow and complex paths (the pore radius is small) and the corresponding extremely low permeability should in theory create very high sorption capacity in coal. However, this does not happen in reality due to the Klinkenberg slip-flow effect, which causes the gas molecules to leave the pore walls with molecular vibration with the slip-flow effect. Therefore, greater permeability of gas in coal can be expected compared to liquid/super-critical CO<sub>2</sub>.

To account for this Klinkenberg slip-flow effect, the permeability calculated based on the Darcy equation needs to be modified as follows [57]:

$$k_a = k_\alpha \left(1 + \frac{b}{p}\right) \quad (4)$$

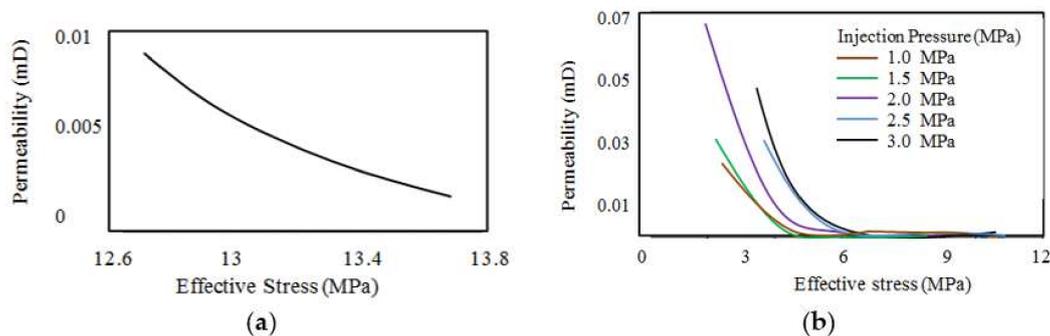
where  $k_a$  is the apparent permeability,  $k_\alpha$  is the Darcy's permeability of incompressible stream line flow,  $b$  is the Klinkenberg slip factor, and  $p$  is the mean gas pressure.

The permeability reduction in gas CO<sub>2</sub> with increasing pressure caused by Klinkenberg slip flow has been reported in coal, as in the laboratory test results on samples obtained from the Bulli seam in Australia by Lama [58].

On the other hand, injection of CO<sub>2</sub> causes the methane present in the coal mass to be desorbed, offering more space for gas movement inside the coal mass and this clearly enhances the coal mass permeability, where the effect is increasing with increasing injection pressure [59]. On the basis of all of these factors, the final CO<sub>2</sub> injection pressure effect on coal permeability is clearly dependent on the combined influence of CO<sub>2</sub> adsorption, coal matrix swelling, CH<sub>4</sub> desorption, matrix shrinkage and the Klinkenberg slip-flow effect.

#### 4.1.3. Effective Stress

Since effective pressure explains the net effect of pore pressure and confining pressure, it more effectively describes the stress effect on coal mass permeability. The coal permeability alteration caused by effective stress during the CO<sub>2</sub> sequestration process has been studied by many researchers. Although, increasing gas pressure during injection reduces gas slip-flow effect, it also decreases the effective stress, which in turn influences the permeability. Yang et al. [60] conducted laboratory experiments to investigate the coupled effect of both slip-flow effect and effective stress and found that the apparent permeability is greatly controlled by these two competing effects. Moreover, according to Vishal et al. [40] and Jasinge et al. [36], CO<sub>2</sub> permeability in coal exhibits an exponentially reducing trend with the effective stress applied on it (Figure 12).



**Figure 12.** Influence of effective stress on CO<sub>2</sub> permeability in (a) high-rank coal [40], and (b) low-rank coal [36].

Vishal et al. [40] proposed Equations (5) and (6) to predict CO<sub>2</sub> permeability in Indian bituminous coal as a function of effective stress applied on the coal mass for low and high CO<sub>2</sub> injection pressures, respectively:

$$k = -0.056 \ln \sigma_e + 0.145 \quad (5)$$

$$k = -0.293 \sigma_e + 0.2139e \quad (6)$$

where  $k$  is the permeability of CO<sub>2</sub> in coal and  $\sigma_e$  is the effective stress applied on coal.

However, Jasinge et al. [36] proposed a different exponential relationship to predict the permeability of Victorian brown coal from effective stress as follows:

$$k = k_0 e^{-1.08(0.05k_0 + 1)\sigma_e} \quad (7)$$

where  $k_0$  is the permeability at initial stress condition and  $\sigma_e$  is the effective stress.

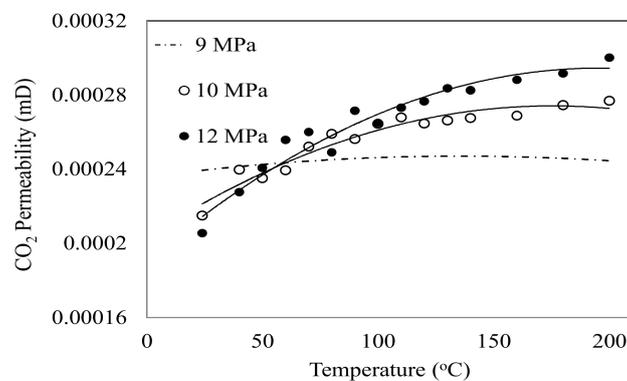
Considering these relations proposed by different researchers, it is clear that although the negative correlation of effective stress with permeability is a common observation, proposing a universal relation

among coal permeability and effective stress is quite difficult, due to the highly inhomogeneous nature of coal. This is because, coal seam gas reservoirs located in deep underground are often subjected to multi-directional stress components like those on any other rock material deposited over the years, and its heterogeneous nature has been formed due to the existence of different constituents in the coal mass. As a result, the overall influence is quite complex and varies significantly from location to location and seam to seam. Such complexities create many uncertainties and complications in both laboratory studies and modelling work. Therefore, the heterogeneous nature of coal has been a major barrier to understanding the effects of some important geomechanical, geochemical and environmental factors on the behaviour of coal [61].

#### 4.1.4. Seam Temperature

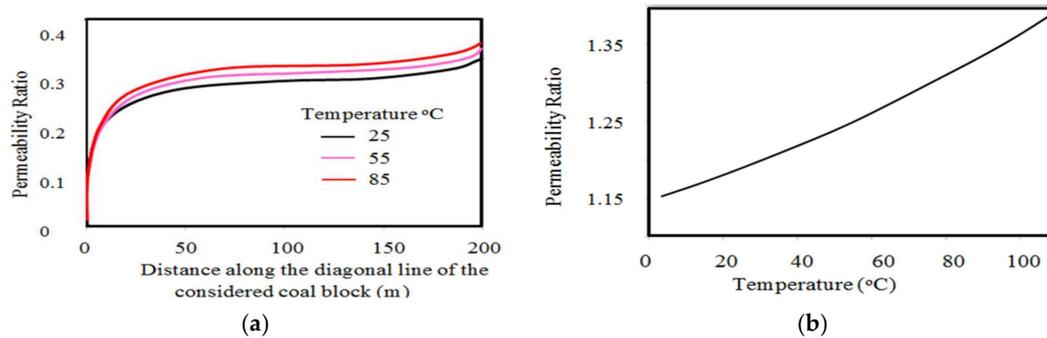
As mentioned earlier, temperature in preferable coal seams is normally considerably higher than the ground condition and this may have a significant influence on the CO<sub>2</sub> sequestration process in deep coal seams. On one hand, the high temperatures underground assist in converting the phase condition of injecting CO<sub>2</sub> to its super-critical condition, reducing its flow ability in coal, as mentioned in previous sections. However, increased temperature also has a positive influence on CO<sub>2</sub> sequestration in coal, with increasing seam porosity through thermal cracks and reducing the swelling effect. Therefore, the combined influence of CO<sub>2</sub> phase transition, thermal expansion and thermal crack formation and changes in adsorption behaviour need to be studied to precisely identify the influence of temperature on coal permeability. However, to date a minor consideration has been given to the identification of the influence of temperature on CO<sub>2</sub> permeability in coal [23,62].

For example, the detailed combined experimental and numerical study of Perera et al. [23] showed a clear increment in CO<sub>2</sub> permeability in bituminous coal with increasing temperature (Figure 13) and how it varies with CO<sub>2</sub> pressure. According to these researchers, the permeability increment with increasing temperature is only significant at high CO<sub>2</sub> pressures (>10 MPa) and is negligible at low CO<sub>2</sub> injection pressures (<9 MPa). It should be noted that the researchers used tri-axial undrained experimental conditions to calculate the permeability of coal under varying bed temperature conditions, and at high CO<sub>2</sub> injection pressures most of the CO<sub>2</sub> inside the coal sample was in the super-critical state when the temperature was above the critical temperature of CO<sub>2</sub> (their tested temperatures were 25 to 200 °C). As discussed earlier, super-critical CO<sub>2</sub> adsorption in the coal matrix causes it to be subjected to a greater swelling effect and therefore increasing the temperature causes the reduction of this swelling effect. In other words, at high pressure, the swelling effect will be more greatly reduced with increasing temperature and this will result in greatly enhanced coal mass permeability. However, the CO<sub>2</sub> permeability enhancement with increasing temperature in coal may be reduced at very high temperatures (>100 °C, in Perera et al. [23]), due to the associated thermal expansion of the coal matrix.



**Figure 13.** Influence of temperature on CO<sub>2</sub> flow ability in bituminous coal for various CO<sub>2</sub> pressures [23].

Perera et al. [23] used only one type of coal (bituminous) to study the temperature effect on CO<sub>2</sub> flow behaviour in coal. The finding of the greater temperature effect on coal at high CO<sub>2</sub> pressures given in Perera et al. [23]) was later confirmed by the field-scale simulation results of Qu [62], who also showed a greater permeability reduction at 10 MPa CO<sub>2</sub> pressure condition compared to the 7 MPa pressure condition (Figure 14). According to the studies of both Perera et al. [23] and Qu [62], the influence of temperature on coal permeability is much lower than the influence of the other factors, such as effective stress, CO<sub>2</sub> injection pressure and depth. However, in order to obtain more detailed knowledge related to this aspect, it is necessary to find this temperature effect on CO<sub>2</sub> adsorption-induced swelling in varyingly rank coals (low to high rank coals).



**Figure 14.** Influence of temperature on CO<sub>2</sub> flow ability in coal at various CO<sub>2</sub> injection pressures [62]. (a) 7 MPa CO<sub>2</sub> injection pressure; (b) 10 MPa CO<sub>2</sub> injection pressure.

Since CO<sub>2</sub> has unique flow behaviour in coal due to various complexities induced in the coal mass upon CO<sub>2</sub> injection, including adsorption, swelling, and shrinkage and their dependence on various factors, including injecting CO<sub>2</sub> phase, it is essential to have a precise identification of specific flow behaviour prior to the initiation of field work in any given coal seam, in order to achieve successful sequestration.

#### 4.1.5. Effect of Mineralogy

Some coal seams consist of a considerable mineral composition, which triggers the need to evaluate the alteration of mineralogy upon CO<sub>2</sub> injection. Since most coal seams exist water saturated, injected CO<sub>2</sub> dissolves in formation water and the carbonic acid (H<sub>2</sub>CO<sub>3</sub>) forms, resulting in an acidic environment. This results in a pH reduction, which in turn causes mineral dissolution/precipitation and ions releasing from mineral surfaces [63]. Lebus and Bujok [64] analysed the CO<sub>2</sub> mineral sequestration mechanisms and capacity of the upper Silesian coal basin with modelling and experimental verifications. They found that the dissolution of calcite or siderite due to dissociation of carbonic acid is a typical reaction in the particular aquifer, which contains carbonate minerals in its rock matrices. This reaction increases the carbonate concentration in pore water and may enhance the kaolinite dissolution due to increased acidity. Simultaneously, chalcedony and dawsonite minerals are precipitated. This mineral dissolution/precipitation can cause permeability enhancement or reduction, depending on the mineralogy of a particular coal seam. However, it should be noted that the coal seam mineral composition is quite low in most cases, which in turn reduces its effect on coal seam permeability alteration.

## 5. Simulation of CO<sub>2</sub> Flow Behaviour in Coal

To date, many permeability models have been proposed for gas movement in coal and most have been developed by assessing the effect of stress on coal permeability. One of the earliest studies was by Somerton et al. [18], who considered N<sub>2</sub> and CH<sub>4</sub> gas flow injection into three different types of fractured coal samples taken from different locations in USA (Pittsburgh, Virginia Pocahontas, and

Greenwich Collieries) under various axial and radial stress conditions. Their experimental results reveal the importance of stress history on coal permeability and the negligible influences of loading sequence and direction of stress application on it. Based on these observations, they proposed an equation for coal permeability as a function of effective principal stress, as follows (Equation (8)):

$$K_{\sigma} = K_0[\exp\{-(3 \times 10^3 \sigma K_0^{-0.10})\} + (2 \times 10^{-4} \sigma^{1/3} K_0^{1/3})] \quad (8)$$

where  $K_{\sigma}$  is the permeability under stress (mD),  $K_0$  is the permeability under zero stress (mD) and  $\sigma$  is mean stress (psi).

In 1986, Durucan et al. [19] proposed a somewhat different relationship among coal permeability and effective stresses, considering the radial stress applied on the coal mass and the permeability behaviour of fractured coal samples taken from seven different locations (Acilik, Caydamar, Barnsley, Cockhead, Banbury, Dunsil and Deep Hard):

$$K = (1.12 - 0.03\sigma_3)K_i \times \exp\{-(1.12 - 0.03\sigma_3)C\sigma_3\} \quad (9)$$

where  $\sigma_3$  is radial stress (MPa),  $K$  is permeability (mD),  $C$  and  $K_i$  are constants, where  $K_i$  is the relative incidence of excising fissures and fractures of coal and  $C$  is the volatile matter content-dependent compressibility factor of the coal (Figure 15).

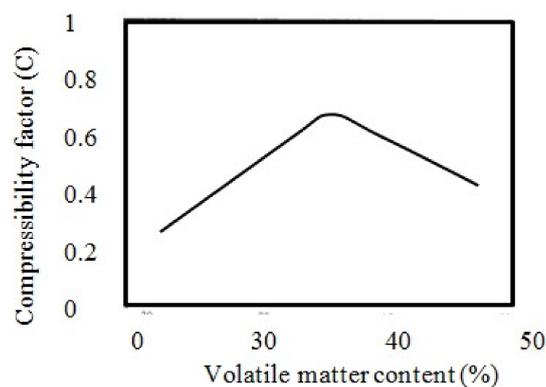


Figure 15. Volatile matter content-dependent compressibility factor of coal [19].

The main limitation in this equation is the finding of  $K_i$ . Although it has been defined as the relative incidence of existing fissures and fractures of coal, insufficient details have been given to find it in practice. Without knowing the way of finding the value of  $K_i$ , it is difficult to use this equation to find the permeability. Further, in this research the researchers maintained the  $\sigma_1 = 3\sigma_3$  relationship throughout each test ( $\sigma_1$  is the major principal stress and  $\sigma_3$  is the minor principal stress acting on the coal sample) and therefore the influence of the major principal stress ( $\sigma_1$ ) on permeability is clear and therefore needs to be incorporated for the precise estimation of permeability.

Another coal permeability relationship with applying stresses has been proposed by Gray [65], who conducted tri-axial experiments on  $\text{CH}_4$  injection into several coal types under various isotropic confined stress levels (Equation (10)):

$$K = 1.013 \times 10^{-0.31\sigma} \quad (10)$$

where  $K$  is permeability (mD) and  $\sigma$  is confining stress (MPa), which is proposed to be estimated using Equation (11) to account for the effect of matrix shrinkage and pore pressure:

$$\sigma - \sigma_o = -\frac{\theta}{1 - \theta}(p - p_o) + \frac{E}{1 - \theta} \frac{\Delta \epsilon_s}{\Delta p_s} \Delta p_s \quad (11)$$

where,  $\Delta p_s$  is the change in equal sorption pressure (MPa),  $\frac{\Delta \epsilon_s}{\Delta p_s}$  is the strain caused by a unit change in equivalent pressure,  $\nu$  is the Poisson's ratio,  $p_o$  is the initial pressure (MPa),  $p$  is the pressure (MPa),  $\sigma_o$  is the initial confining stress (MPa) and  $\sigma$  is the confining stress after the changes caused by pore pressure and matrix shrinkage (MPa). In Equation (11), the first term provides the pore pressure influence and the second term describes the pore pressure reduction creating matrix shrinkage influence on cleat permeability. However, this equation was developed based on the basic assumption that coal matrix shrinkage is proportional to the reduction in equivalent sorption pressure, and cannot predict the initial permeability increment in coal seam gas recovery at the primary stage.

In the same study, the researcher showed the applicability of the well-known Darcy equation for predicting coal permeability (Equation (12)) with assuming a linear laminar behavior of CO<sub>2</sub> in coal:

$$K = (1.013 \times 10^{13}) \frac{2qp_oL\mu}{A(p_i^2 - p_o^2)} \quad (12)$$

where  $q$  is the flow rate (m<sup>3</sup>/s),  $P_o$  is the outlet pressure (MPa),  $L$  is the length of core (m),  $\mu$  is the viscosity of the flow (Pa.s),  $A$  is the area of core sample (cm<sup>2</sup>) and  $P_i$  is the inlet pressure (MPa).

In 2011, Jasinge et al. [36] proposed the following simplified Darcy permeability equation for use in laboratory data analysis:

$$k_{ND} = \frac{\mu L}{A} \frac{Q_1 Q'_1 (Q_1 - Q'_1)}{[Q_1^2 (P'_{out} - P'_{in}) - Q'^2_1 (P_{out} - P_{in})]} \quad (13)$$

where  $Q_1$  and  $Q'_1$  are two known flow rates for known confining pressures and gas injection pressures  $P_{in}$  and  $P'_{in}$ , where  $P_{out}$  and  $P'_{out}$  are the corresponding outlet pressures.

However, there are some limitations when applying this simplified Darcy equation for actual reservoir conditions. The empirical equation was developed by testing natural and reconstituted brown coal samples in laboratory conditions. The equation does not address the effect of natural cleat system in high rank coals and the influence of coal matrix swelling. Importantly, it is not possible to expect the linear laminar condition of the flow in deep coal seams during the CO<sub>2</sub> sequestration process, particularly close to the injection point, and flow through the seam may be in a turbulent, non-Darcy state. In such situations it is necessary to use a more accurate approach as shown below to predict permeability [66]:

$$\frac{dP}{dx} = \frac{\mu V}{k_{ND}} + \rho \beta V^2 \quad (14)$$

where  $\frac{dP}{dx}$  is the pressure gradient,  $V$  is the velocity,  $\rho$  is the flowing fluid density,  $\mu$  is the flowing fluid viscosity,  $k_{ND}$  is the non-Darcy permeability and  $\beta$  is the non-Darcy coefficient of the porous medium.

The pore pressure effect on permeability has also been studied by Harpalani and Zhao [46], who proposed a fully empirical relation for coal permeability with pore pressure as follows:

$$K = \frac{A}{P} + B + CP \quad (15)$$

where  $A$ ,  $B$  and  $C$  are constants and  $P$  is the pore pressure. Although this model is easy to use due to its simplicity, it does not precisely describe the pore pressure effect on coal permeability, as the first and third terms in the right-hand side of the equation describe opposite pore pressure influences on coal permeability.

In 1992, Seidle [67] proposed a new relationship among coal cleat permeability with effective stress and pore pressure, using two types of coal taken from the Black Warrior and San Juan Basins (Equations (16) and (17)):

$$K_{f2} = K_{f1} [\exp\{-3C_f(\sigma_{h2} - \sigma_{h1})\}] \quad (16)$$

$$K_{f2} = K_{f1} [\exp\{-1.91E - 3(p_{p1} - p_{p2})\}] \quad (17)$$

where  $K_f$  is the cleat permeability (mD),  $C_f$  is the cleat volume compressibility ( $\text{kPa}^{-1}$ ),  $\sigma_{h2}$  is the hydrostatic stress (kPa),  $E$  is the elastic modulus of coal and  $p_p$  is the pore pressure of the coal mass. However, since these relations are based on the data of only two different types of coal samples, Seidle [67] suggested more studies using a variety of coal samples taken from different locations to check the accuracy of this model as the universal model for coal.

In 1995, Seidle and Huitt [68] proposed a descriptive model for coal porosity considering the effect of coal matrix shrinkage on gas desorption (matrix shrinkage expands the pore space in coal and therefore enhances coal mass permeability), based on the experimental data of highly volatile bituminous coal samples taken from the San Juan Basin. This relationship more precisely describes the influence of coal matrix deformations (which occur through sorption and desorption during  $\text{CH}_4$  and  $\text{CO}_2$  gas flows) on coal mass pore space:

$$\varnothing - \varnothing_o = 1 + \left(1 + \frac{2}{\varnothing_o}\right)\varepsilon_1 \left(\frac{bp_o}{1+bp_o} - \frac{bp}{1+bp}\right) \quad (18)$$

where  $\varnothing$  is the coal bed porosity after sorption/desorption of gases,  $\varnothing_o$  is the initial coalbed porosity,  $\varepsilon_1$  is the strain and  $b$  is a constant. This equation can be used to predict coal permeability with the assistance of the basic porosity-permeability cubic law for porous media:

$$\frac{k}{k_o} = \left(\frac{\varnothing}{\varnothing_o}\right)^3 \quad (19)$$

However, this equation concerns only the effect of matrix shrinkage on permeability and does not consider the influence of effective stress.

In 1990, Sawyer et al. [69] developed the following widely-used equation for coal permeability (commonly called the ARI model):

$$\varnothing = \varnothing_o \left[1 + C_p(p - p_o) - C_m(1 - \varnothing_o) \frac{\Delta P_i}{\Delta C_i} (C - C_o)\right] \quad (20)$$

where  $C_p$  is the pore volume compressibility,  $C_m$  is the matrix shrinkage compressibility ( $C_m = \varnothing C_p$  according to McKee et al. [16]),  $\frac{\Delta P_i}{\Delta C_i}$  is the pressure change per  $\Delta C_i$  concentration variation and  $C_o$  is the initial gas concentration. Here, the first term describes the porosity changes due to pore pressure and the second term describes the porosity changes due to matrix shrinkage.

Later, Palmer and Mansoori [50] proposed the following equation for coal permeability based on the theoretical evaluation of the effects of matrix shrinkage and effective stress on coal permeability, and proposed a new model for coal permeability as a function of both matrix shrinkage and effective stress (commonly known as the P & M model):

$$d\varnothing = C_m dp + \varepsilon_l \left(\frac{K}{M} - 1\right) \frac{d}{dp} \left(\frac{bp}{1+bp}\right) dp \quad (21)$$

where:

$$C_m = \frac{1}{M} - \left[\frac{K}{M} + f - 1\right]\beta \quad (22)$$

where  $d\varnothing$  is the porosity difference,  $dp$  is the pore pressure difference (md),  $\varepsilon_l$  is the Langmuir volume,  $b$  is the Langmuir constant (describes the effect of grain compression),  $K$  and  $M$  are bulk and the constrained axial modulus, respectively and can be given as functions of Poisson's ratio as follows:

$$M = \frac{E(1 - \vartheta)}{(1 + \vartheta)(1 - 2\vartheta)} \quad (23)$$

$$K = \frac{E}{3(1 - 2\vartheta)} \quad (24)$$

Integration of Equation (21) gives:

$$\frac{\varnothing}{\varnothing_o} = 1 + \frac{C_m}{\varnothing_o}(p - p_o) + \frac{\varepsilon_l}{\varnothing_o} \left( \frac{K}{M} - 1 \right) \left( \frac{bp}{1 + pb} - \frac{bp_o}{1 + bp_o} \right) \quad (25)$$

where  $\varnothing_o$ ,  $p_o$  and  $C_m$  and are the original reservoir porosity, the original pressure (MPa) and the matrix shrinkage compressibility, respectively. This equation also can be used to predict permeability by combining it with Equation (19). Here, the first terms describe the effective stress influence and the last term describes the matrix shrinkage influence. According to the researchers, this equation can be reduced to the following simpler model if the compressibility factor,  $C_m$  is constant, although it will not be constant practically as it varies with medium porosity and Young's modulus:

$$\frac{k}{k_o} = \exp[3C_p(p - p_o)] \quad (26)$$

However, it should be noted that Equation (25) can only be used under constant applied pressure conditions (only the flow effect is considered) as the effect of stress variation is not considered. To date, this model has been widely used in coal bed methane simulators. However, all the coal permeability models described above have been developed for methane production and CO<sub>2</sub> injection has been ignored.

Therefore, Pekot et al. [51] studied both the ARI and the P & M models and showed the importance of adding a new term to account for the effect of differential shrinkage of coal mass due to CO<sub>2</sub> movement. This is because, based on their experimental study, they found that greater strain (swelling) was developed by CO<sub>2</sub> in coal compared to CH<sub>4</sub>, which causes greater porosity and permeability reductions and is called "differential swelling". The proposed new equation for coal porosity after adding the differential swelling term is shown below:

$$\varnothing = \varnothing_o[1 + C_p(p - p_o)] - C_m(1 - \varnothing_o) \frac{\Delta P_i}{\Delta C_i} [(C - C_o) + C_K(C_o - C)] \quad (27)$$

where  $C_K$  is the differential swelling coefficient. The effect of differential swelling on coal permeability reported by Pekot et al. [51] is shown in Figure 16.

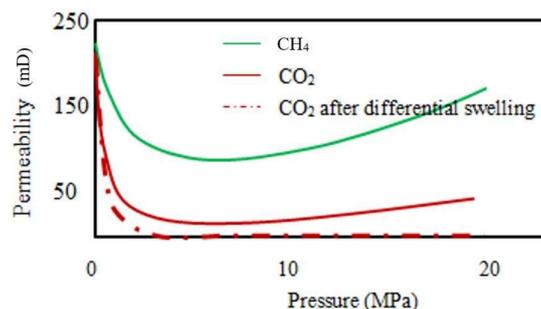


Figure 16. Differential swelling during CO<sub>2</sub> flow in coal [51].

In 2000, Gilman and Beckie [70] developed a theoretical approach to coal permeability for methane movement, considering the regular cleat system in coal, adsorptive gas storage, the extremely slow mechanism of methane release from the coal matrix to cleats and the significant permeability changes caused by this desorption. Here, they considered two different types of methane gas flow mechanisms inside the coal mass: (a) in fractures, modelled using Darcy's law and, (b) in coal mass micro-pores,

modelled using the Knudsen diffusion formula (coupled with ideal gas flow), to derive the following equation for coal seam permeability:

$$\frac{k}{k_0} = \exp\left[-\frac{3\Delta\sigma_x^{(e)}}{E_F}\right] \quad (28)$$

where  $\sigma_x^{(e)}$  is the change in effective stress and  $E_F$  is the fracture's Young's modulus. This equation was then converted into a more comprehensive form by adding the matrix shrinkage/swelling effects as follows, showing both effective stress and the matrix shrinkage effects. Here two basic assumptions have been made; (1) the coal mass is a homogeneous isotropic elastic medium and (2) zero lateral strain occurs in it:

$$k = k_0 \exp\left(\frac{3\vartheta}{1-\vartheta} \frac{\Delta p_F}{E_f}\right) \exp\left(\frac{3\alpha E}{1-\vartheta} \frac{\Delta S}{E_f}\right) \quad (29)$$

where  $\Delta S$  is the change of adsorbed mass,  $\alpha$  is the volumetric swelling coefficient,  $\vartheta$  is the Poisson's ratio,  $p_F$  is the pore pressure and  $E$  is the coal mass Young's modulus.

A similar but more advanced and precise theoretical approach to simulate the CO<sub>2</sub> flow behaviour in coal has been proposed by Wang et al. [71], based on some basic assumptions: (1) non-water flow in the fluid phase, (2) the adsorbed phase is in equilibrium and (3) the system is isothermal, (4) zero lateral strain occurs in it:

$$k(S) = k(S_0) \exp\left(\int_{S_0}^S \frac{3\alpha(S)E}{(1-\vartheta)\tau_f E_f(S)} dS\right) \quad (30)$$

where

$$\frac{1}{E_t} = \frac{1}{E_f} + \frac{1}{E} \quad (31)$$

where  $E_t$  is the coal mass Young's modulus,  $E$  is the Young's modulus of the coal matrix,  $\tau_f$  is the tortuosity,  $S_0$  and  $S$  are the initial and final adsorbed mass, and  $k(s_0)$  and  $k(s)$  are the initial and final permeability values.

However, all the models described above have been developed based on one common assumption: that lateral strain is zero. Although this is applicable for field coal seams with hydrostatic stress, the use of this assumption for laboratory-scale coal sample testing such as tri-axial testing is not correct, as there is considerable lateral strain in the coal sample under such conditions. Therefore, in 2013, Perera et al. [48] derived the following more compressive equation to predict coal seam permeability in coal under tri-axial, non-zero lateral strain conditions:

$$k = k_0 \left\{ \exp\left[\Delta\sigma_{CO_2} \left(\frac{3\frac{E}{E_f}(1-\vartheta_f)}{2(1-2\vartheta)k'_{bulk}} - \frac{3(1+\vartheta_f)}{2E_f}\right)\right] \right. \\ \times \exp\left[\Delta\sigma_A \left(\frac{3(1+\vartheta_f)}{2E_f} + \frac{1\frac{E}{E_f}(1-\vartheta_f)}{2(1-2\vartheta)k'_{bulk}}\right)\right] \\ \left. \times \exp\left[-\Delta\sigma_c \left(\frac{\frac{E}{E_f}(1-\vartheta_f)}{(1-2\vartheta)k'_{bulk}}\right)\right] \times \exp\left[-\Delta S \left(\frac{9\frac{E}{E_f}(1-\vartheta_f)\alpha_s}{2(1-2\vartheta)}\right)\right] \right\} \quad (32)$$

where  $k$  is the permeability of the coal mass,  $k_0$  is the initial permeability of the coal mass,  $E$  is the coal mass Young's modulus,  $E_f$  is the fracture Young's modulus,  $\vartheta$  is the Poisson's ratio of the coal mass,  $\vartheta_f$  is the Poisson's ratio of fractures,  $k'_{bulk}$  is the bulk modulus of the coal mass,  $\sigma_{CO_2}$  is the CO<sub>2</sub> injection pressure,  $\sigma_A$  is the axial stress,  $\sigma_c$  is the confining pressure,  $\varepsilon_v$  is the volumetric strain,  $\alpha_s$  is the volumetric swelling coefficient for coal, and  $\Delta S$  is the change of adsorption mass.

## 6. Potential Hazards Associated with CO<sub>2</sub> Infectivity

It is important to consider some of the potential hazards associated with the CO<sub>2</sub> sequestration process in coal seams, as it can create several environmental and social issues. This paper has comprehensively discussed the possibility of permeability reduction during CO<sub>2</sub> injection. As discussed, the CO<sub>2</sub> injection significantly alters the coal pore-structure resulting in a global permeability reduction due to cleat healing that is caused by matrix swelling during the CO<sub>2</sub> adsorption in the micro-pores. This can possibly affect the sequestration projects' cost factors, as the swollen matrix can reduce or completely stops well injectivity in the long run. Furthermore, it is important to rigorously study the coal matrix swelling process as it can possibly fail the cap rock due to the significant stress applied on it. This failure can cause back-migration of CO<sub>2</sub> in to the atmosphere, causing number of environmental hazards. Sudden CO<sub>2</sub> outbursts can even be lethal for people living near the project areas. Furthermore, the higher adsorption capacity, the higher potential as a solvent and acidic chemical interactions with CO<sub>2</sub> can significantly influence coal mass mechanical properties, resulting a large reduction of coal seam strength. This weakening of coal mass can cause irreversible damage to the reservoir and the CO<sub>2</sub> and methane can be leaked into the adjacent aquifers, creating dramatic environmental hazards. Thus the CO<sub>2</sub> sequestration projects should be carefully designed and implemented with considering all these factors, in order to maintain an economical process, while preventing the so called social and environmental hazards.

## 7. Conclusions

The permeability of natural coal seams is influenced by the geological stress field of the coal seam, its temperature and moisture content, and the coal maturity or rank. Coal permeability is basically dependent on the porosity of the coal mass, as highly porous media have higher capability to offer easy flow paths. Therefore, the factors affecting coal mass porosity have a direct impact on its flow ability or permeability. In this respect, effective stress plays a major role, because being a type of relatively weak and compressible rock, coal's pore structure is greatly reduced with increasing effective stresses applied on it, because it causes the fissures/pores of the coal mass to close, increasing the tortuosity. For this reason, deep coal seams generally have much lower permeability values than shallow seams.

Coal tends to become more permeable with the reduction of its moisture content, because moisture occupies the pore space available in the coal mass for fluid movement and adsorption of that water into the coal matrix induces significant matrix swelling, both of which obstruct the flow paths in the coal matrix. Preferable coal seams for CO<sub>2</sub> sequestration/ECBM have considerably high temperatures due to their deep depths. Although high temperatures cause thermal expansion in the coal matrix with resulting its permeability reduction, extremely high temperatures may create thermal cracks in the coal matrix, thus increasing permeability. Deeper coal seams normally contain higher ranked coal as that coal had more opportunities to be subjected to greater pressure and temperature variations and much lower permeability can be expected in deeper coal seams. Based on all of these facts, since deeper coal seams have greater effective stresses, higher ranked coals and potential water saturation conditions, deep coal seams generally have very low permeability values, although the effect of temperature is the opposite.

Flowing CO<sub>2</sub> inside a coal seam during CO<sub>2</sub> sequestration/ECBM has a significant influence on its natural existing permeability through CO<sub>2</sub> adsorption-induced swelling and hydrocarbon mobilisation in the seam, which have been widely experienced in field CO<sub>2</sub> sequestration projects, and the CO<sub>2</sub> injection rate has been greatly reduced after the initial stage of CO<sub>2</sub> injection in many field projects.

However, CO<sub>2</sub> flow behaviour can vary from seam to seam, and deeper coal seams generally have lower CO<sub>2</sub> flow ability due to the shrunken pore structure due to the available greater pressures and temperatures. This low CO<sub>2</sub> permeability existing at greater depths has created serious issues in large-scale sequestration projects, as critically high injection pressures are required to achieve sufficient CO<sub>2</sub> injection into them, which is an expensive task and also involves some risks associated with CO<sub>2</sub> leakage. Therefore, the combined influences of high temperatures (thermal cracks and

thermal expansion), changes in adsorption behaviour and therefore swelling and the associated phase transition need to be studied to precisely identify the influence of temperature on coal permeability. Coal permeability for CO<sub>2</sub> reduces with increasing CO<sub>2</sub> pressure due to the swelling effect and the slip-flow effect. The change of CO<sub>2</sub> phase condition from gas/liquid state to super-critical state in deep seams has a significant influence on swelling. Since super-critical CO<sub>2</sub> occupies much smaller space in the seam, generally it is planned to target deep seams for CCS projects. However, super-critical CO<sub>2</sub> is more chemically reactive and creates a significantly higher swelling effect in coal and has much reduced flow ability. Gas CO<sub>2</sub> flow through coal occurs along very narrow and complex paths and in theory extremely low permeability creates very high sorption capacity in coal. However, this does not happen due to the Klinkenberg slip-flow effect, which causes greater permeability for gas in coal compared to liquid/super-critical CO<sub>2</sub>.

Theoretical and empirical models play very important roles in predicting CO<sub>2</sub> flow behaviour in order to identify the coal mass properties in deep coal seams, considering the factors affecting it, including porosity, effective stress, and swelling /shrinkage.

**Author Contributions:** Mandadige Samintha Anne Perera, the sole author of the manuscript, carried out all the manuscript preparation steps including, reviewing of literature, drafting the manuscript and the proof checking of it.

**Conflicts of Interest:** The author declares no conflict of interest.

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