A Review Summary on Multiple Aspects of Coal Seam Sequestration

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Abstract Presence of natural gas in adsorbed form in coal seams is the primary reason for scientists to attempt CO_2 sequestration in the same. The economic analysis states that the additional methane in case of coupled enhanced coalbed methane recovery (ECBMR) with sequestration partly offsets the cost of the operation. Injected CO_2 reduces the partial pressure of methane and enhances its desorption from the matrix. Furthermore, CO_2 is preferentially adsorbed onto the porous surface of the coal thereby displacing methane from adsorption sites. Apart from estimation of coal gas reserves, several technical parameters related to the adsorption capacity of coals and suitable trapping/sealing mechanism must be ensured before utilizing coal as a CO_2 sink.

Parameters such as geomechanical characteristics, swelling/shrinkage, CO_2 permeability in coal, role of effective stresses at higher confining pressure corresponding to deeper target coal seams etc. should be studied in detail before embarking on such problems in the field scale. Various studies ranging from experimental to analytical and numerical modeling have been conducted in the past. This chapter reviews the literature in CO_2 geosequestration in coal with/without ECBMR covering the physical aspects like fluid flow in coal, fluid storage in the adsorbed form, matrix deformation of the porous media, effect of shrinkage/swelling, flow permeability, existence of fluid in its different phases etc. in context to coals worldwide.

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1 Introduction

Coal is composed of solid matrix blocks bound by a well-defined network of a pore system. Coal is a heterogeneous porous organic rock composed of micropores/primary porosity and macropores/secondary porosity and is otherwise known as a dual porosity porous media. Micropores form the major share of the porous structure and are responsible for storage and concentration gradient based movement of fluid molecules. On the other hand, macropores are the cleats/fractures formed in response to local stresses, and serve as the medium for pressure driven fluid flow. These porous networks represent the domain of fluid existence in the porous media and control the interactions and movement of the fluid in the solid matrix, which in turn depends upon the coal characteristics and fluid properties. The gas flow effectively involves three mechanisms: desorption from the pores, diffusion through micropores to the cleat network, and flow to the outlet by Darcy's laminar flow and Knudsen diffusion.

The fractures/cleats system of coal is complex. There are mutually perpendicular face cleats and butt cleats that comprise the extensively developed fracture network (Fig. 1 from [1]). Compositionally, coal comprises organic and inorganic matter along with volatiles. The organic matter is constituted of "macerals" that are the organic equivalents of minerals in rocks and are broadly classified as vitrinite, exinite and inertinite. The inorganic matter of coal includes the different minerals that are non-combustible. These form the ash that is left after burning of coal. Volatiles in coal mainly include moisture, and other gases. Moisture in coal includes internal, inherent as well as external water. Coal is therefore, a

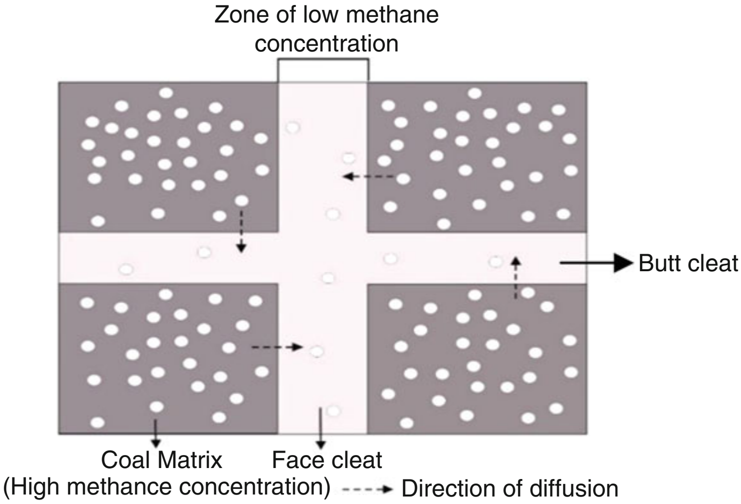


Fig. 1 Cleats orientation and gas desorption in coal [1]

non-volatile, non-crystalline, insoluble, and highly complex mixture of organic molecules of diverse sizes and structures [2].

2 Existence of Gases in Coal

The gases present in coal are formed during the process of coalification and may either be biogenic or thermogenic in origin. Biogenic methane is generated when peat forms as a result of decomposition of organic matter at temperatures below 50 °C. The later stages of coalification witness a high temperature due to an increase in burial depth and influence of magmatic activities because of which, coals of a higher rank are formed. Gases that form in this period of coal formation are referred to as thermogenic methane. Although the gases in coal seams comprise methane dominantly, they also include carbon dioxide, carbon monoxide, and nitrogen from organic decay; in some cases, hydrogen sulphide is generated in trace amounts from the humic source substance. Some higher hydrocarbons are also present in a very small proportion. Methane is dominantly retained in coal seams as adsorbed molecules on the organic micropore surfaces. These pores are mostly inaccessible to formation water and have a diameter <2 nm, qualifying as micropores according to International Union on Pure and Applied Chemistry (IUPAC) classification [3]. It is well understood that more than 95 % of gases in coal beds are stored by the mechanism of physical adsorption or sorption. Minute quantities may be present as free gas in pores and fractures or as dissolved in solutions within the coal beds [4, 5].

2.1 Adsorption/Desorption of Gases in Coal

The recovery of methane from the coals is initiated on creating certain favorable conditions that are different from those applicable in conventional gas reservoirs. Removal of water from the CBM reservoirs generates a pressure gradient due to the depressurization of the coal seam, causing gases to desorb from the pores, diffuse through the matrix and finally catch the pressure gradient in cleat network [6]. This is known as primary recovery of coal bed methane. Other possibilities of extraction of methane include the injection of gases more sorptive than methane, by which the coal automatically releases the methane due to its affinity for the injected gas. The third alternative is injection of gases to reduce the partial pressure of methane that causes methane release from coals. The latter two options of methane recovery are referred to as enhanced coal bed methane recovery. At this point it is worthwhile to understand the gas storage mechanism and the adsorption phenomenon with respect to coal. Various physical models are used to study the adsorption/desorption mechanisms. They are called adsorption isotherms and help to determine the gas storage capacity of coal with respect to gas pressures (or concentrations) at a given temperature. The models are based on various assumptions surrounding the very basic attributes of adsorption.

2.1.1 Models to Predict Gas Adsorption Capacity

Langmuir Model

The most common model used for studying the adsorption mechanism in porous media like coal is the Langmuir equation and it is based on the dynamic equilibrium created between the adsorbent and adsorbate molecules with respect to the pressure and temperature of adsorbate [7]. Langmuir isotherm is produced when the extent of adsorbing molecules coverage is believed to be limited to one molecular layer (Type I). It assumes that each site accommodates one adsorbate molecule and hence, maximum adsorption occurs when a monolayer of adsorbate occupies all the adsorptive sites.

The equation for the Langmuir isotherm for a single gas phase is given as:

$$V = V_L \frac{p}{p + p_L} \tag{1}$$

where, V is the volume of gas adsorbed per unit mass and p, the pressure of the system. V_L and p_L are the Langmuir volume constant and pressure constant respectively. These constants vary from material to material and are obtained from the best fit of the adsorption/desorption information. The isotherm, when modified for coal systems accounting for the ash and moisture content in gas volume estimation, may be written as:

$$V = V_L[1 - (w_a + w_m)] \frac{p}{p + p_L} \tag{2}$$

where, V is gas volume in coal, $\mathrm{V_L}$ is dry, ash-free gas Langmuir volume; w_a is weight fraction of ash content, w_m is weight fraction of equilibrium moisture content, p is pressure and p_L is Langmuir pressure. The utility of adsorption isotherms in CBM exploration lies in the estimation of desorption pressure, rate of release of gas due to pressure decline and the volume of gas remnant in the coals after primary recovery. Figure 2 shows a typical Langmuir adsorption graph where the matrix gas content is plotted against the system pressure.

To account for intermolecular interactions, the equation is modified in the case of gas mixtures. Arri et al. [8] formulated the extended Langmuir equation for gas mixtures based on their experiments on methane-nitrogen and methane-carbon dioxide mixtures:

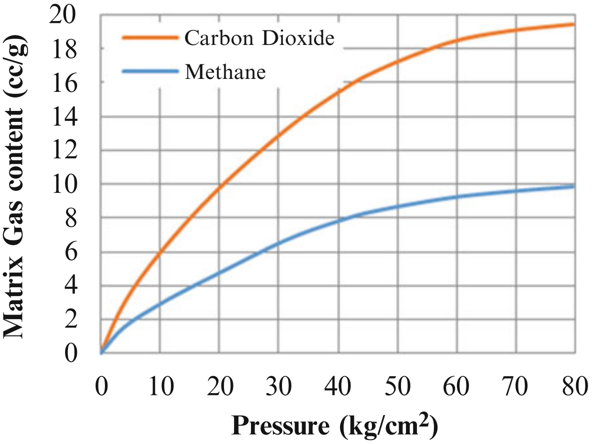


Fig. 2 Langmuir adsorption curves for Raniganj basin coal (After Mendhe et al. [10])

$$v_i = (v_L)_i p \frac{\frac{r_i}{(p_L)_i}}{1 + p\sum_i \frac{r_i}{(p_L)_i}} \tag{3}$$

There are other versions of extended Langmuir isotherm equations [9].

Langmuir's isotherm model is most commonly used for ideal solutions. It is, however, less competent to handle non-idealities in adsorbed phase.

BET Model

The Brunauer, Emmett and Teller (BET) model extends the Langmuir model to multilayers and hence, is applicable to different isotherm types [11]. This accounts well for the enthalpy of sorption and may be used for adsorption of gases at high pressures as well. The heat of adsorption is assumed to be equal to the molar heat of condensation in all the layers. The adsorbed molecules do not interact and the surface of adsorbent is assumed to be energetically uniform. The BET isotherm equation may be defined as:

$$\frac{1}{V\left[(p_o/p) - 1)\right]} = \frac{1}{V_m C} + \frac{(C-1)}{V_m C}\frac{p}{p_o} \tag{4}$$

where, V is the gas volume in coal, V_m is the monolayer volume, C is a constant and p_o is the saturation vapor pressure under experimental temperature conditions. The term p/p_o is known as the relative pressure of the reduced pressure. The rate at which the multilayers form is directly related to the constant C, which is usually greater than unity. BET theory enables experimental determination of the number of molecules required to form a monolayer. At the same time, it establishes that in a multilayer adsorption, the equilibrium of the matrix with gas phase is dynamic and that the actual locations of surface sites covered by one, two or more layers may vary; the number of molecules in every layer, however, will be constant [12]. Although this equation rarely fills to the experimental isotherms for coal matrix, it is still considered to be a useful tool in qualitative study using various isotherm shapes. The model is usually valid between relative pressure values (p/p_o) of 0.05–0.35, when most of the monolayers are completed [13, 14].

Dubinin-Polanyi's Model

Polanyi's Potential theory assumes the existence of a potential field surrounding the adsorbents and immediately above the solid matrix that contains equipotential lines following the contour of surface potential [15, 16]. The space between each set of equipotential surfaces corresponds to a definite adsorbed volume. The adsorption potential may be described as the isothermal work done per mole of adsorbate in transferring molecules from the gaseous state to adsorbed state. Therefore, the adsorption potential, A, depends solely on the volume of gaseous phase, V, held by the equipotential surface surrounding the adsorbent matrix and is directly proportional to it. The plot of V versus A is known as the characteristic curve as this is characteristic of a gas-solid system. Thus, the potential theory of Polanyi holds that for an adsorbate in adsorbed form, the adsorption potential is given by:

$$A = RT \ln(Po/P) \tag{5}$$

where R is the universal gas constant, P is the adsorption pressure and P_o is the saturation vapour pressure of the adsorbate at adsorption temperature, T.

Dubinin [17] used this concept of potential theory to discuss the adsorption phenomenon and formulated the theory of volume filling of micropores (TVFM) that is based on the concept of pore filling. Adsorption in micropores, according to TVFM, happens when the fluid occupies the pore volume by volume filling and not by forming discrete monolayers on homogeneous adsorbent surface. These molecules fit into the micropores and are pressed further inside on compression until the pressure exceeds the vapor pressure, when the adsorbed phase turns into liquid. The theory was worked upon and a simple mathematical relationship was proposed as:

$$V = Vo \exp - (A/E)^2 \tag{6}$$

where E is the characteristic energy parameter of the adsorption system. It depends on the adsorbate and adsorbent properties and their association [17].

The D–P equations were primarily developed for adsorption of vapors below critical point, although, they have been modified for vapors above critical point as well. The characteristic curve can be derived from a single isotherm at any particular temperature; from the characteristic curve, the isotherm at any other temperature may be obtained. This is a unique advantage of D–P equations adding another dimension of estimation of stored gas volume for coal reservoirs that have high variations in temperature, with the use of limited data.

2.1.2 Validation of Various Adsorption Models Through Different Research Works

For a variety of coals, several researchers reported a close approximation of the Langmuir isotherm with their experimental results. Scientists have found that the experimental results on various American, Canadian and Indian coals correspond to Langmuir's isotherm [10, 18–20].

While considering CO_2 driven ECBM recovery, it is important to understand the adsorption of gas mixtures to coal which have been studied for a number of scenarios. The following research works show the application of the Ideal Adsorbed Solution (IAS) Theory and an extended Langmuir equation to successfully validate the data and use them for estimating the adsorption isotherms of gas mixtures. Stevenson et al. [21] measured the adsorption isotherms for binary and ternary mixtures of CH_4, CO_2 and N_2 on coal at 30 °C and pressures up to 5.2 MPa. They obtained varying proportions of adsorbed gases on the coal surfaces and the total amount of gas mixture adsorbed was strongly dependent on the composition and pressure, validating the utility of the IAS theory for successful estimation of gas adsorption behavior of coal. Arri et al. [8] investigated the adsorption of binary mixtures of methane-nitrogen and methane-carbon dioxide at a temperature of 46.11 °C and pressures up to 10.34 MPa for wet coals. They concluded that instead of independent gas adsorption on to the micropores, the two gases competed for the same sorption sites and Langmuir curves were still valid.

Experimental findings of Busch et al. [22] and Ozdemir et al. [23] further corroborate this theory of competition for adsorption sites. Pariti and Harpalani [24] established the adsorption isotherms for ternary adsorption / desorption data from saturated coals at 319.15 K and their data also fit the extended Langmuir isotherm. Detailed experiments were carried out by De Gance et al. [25] on pure and multi component isotherms of CH_4, N_2 and CO_2 using two dimensional equations of state, IAS theory and extended Langmuir equation and obtained matching adsorption isotherms. For wet coals, they found that the Equation of State models matched the results. For the same gas mixtures and pure forms, Hall et al. [26] studied for adsorption of gas mixtures and established that it is the IAS and 2D Equation of State models that provide a better fit to the data than the Langmuir isotherms, which were only useful for pure gaseous phases; thus, they highlighted the findings of Stevenson et al. [21] who advocated the best fit using the IAS theory. Clarkson and Bustin [27] made a comparative analysis of various model predictions for adsorption/desorption in binary/ternary gas mixtures and found that the IAS model most suitably predicted the experimental results. Some early researchers used the dual sorption mechanism to predict the adsorption of gases on coal partly

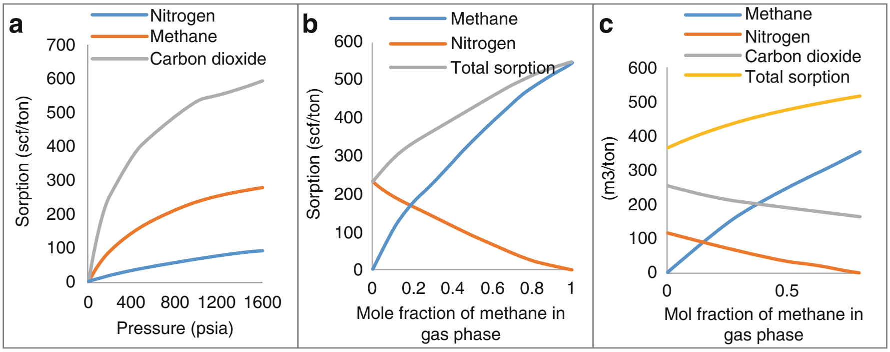


Fig. 3 (**a**) Single component sorption isotherms (dry coal basis) for wet Fruitland coal B at 115 °F, (**b**) $N_2 + CH_4$ mixture sorption isotherms (dry coal basis) for wet Mary Lee coal at 80 °F and 830 psi, (**c**) $N_2 + CH_4 + CO_2$ mixture sorption isotherms (dry coal basis) for wet Fruitland coal C at 115 °F and 526 psi, $y_{CO2} = 0.142$ [31]. It has now been significantly established that there is a preference of adsorption of different gases onto the micro pores of coal—the order from high to low being carbon dioxide, methane, hydrogen, nitrogen—the actual quantities in the proportion varying for different coals [8, 31, 33–36]

based on the solution theory and partly on adsorption [28, 29]. Huddleston et al. [30] studied the adsorption of methane under a temperature of 51.8 °C (125 °F) and pressures up to 15.17 MPa and validated that the Langmuir model could fit the isotherm results at low pressure and a third-order polynomial could fit the whole isotherm. Chaback et al. [31] studied the adsorption/desorption of pure gases and gas mixtures for ECBM recovery process on Fruitland and Mary Lee coals at 46 °C and pressure up to 11 MPa and concluded that the extended Langmuir isotherm was adequate to define the gas adsorption for mixtures as well as for pure forms (Fig. 3). Vishal et al. [1, 32] used the information on Indian coal from previous works and applied the Langmuir models for prediction of CO_2 enhanced coalbed methane extraction and achieved good results on matrix methane saturation, water extraction and volumes of gases released from the chosen coal block.

Although extended Langmuir equations and IAS models fit most of the coal adsorption/desorption data for varying pressures, some data have got a reasonably good fit using the Brunauer, Emmett and Teller (BET) model, Polayni's potential theory and DR and DA models.

3 Coal Matrix Deformation

The gases stored in the coal are adsorbed onto the micropores and desorption is associated with a reduction in the pressure in the coal seam. The natural fractures are widened by this effect enhancing permeability. This occurs in case of sorptive gases when coal swells or expands due to adsorption and shrinks or contracts during desorption. This widening of cleat apertures due to coal shrinkage during desorption results in an increase in macroporosity in coal and vice versa. The phenomenon of adsorption induces swelling in the coal matrix, which is due to the viscoelastic relaxation of the highly crossed macromolecular structure that is strained [37–40]. Upon adsorption of molecules, new bonds that induce swelling are formed between the adsorbate and adsorbent [40, 41]. Increase in gas pressure leads to an increase in swelling and a decrease in the time required to reach the maximum swelling. This confirms that there is a kinetic process involved in the swelling [42]. The other idea maintains that swelling may be an attribute of forces exerted by the adsorbate on to the molecular structure of the adsorbent at high pressures; here, the injected gas behaves like a high density liquid, by which the energy of the system changes. This leads to volumetric changes [43–45].

Several experimental investigations have been conducted to investigate the swelling/shrinkage behavior of coal and to quantify the linear or volumetric strains due to adsorption/desorption of sorptive gases in strained and unstrained conditions. Sorption induced strain have been calculated for pure gases as well as gas mixtures for different types of coals, worldwide. The quantification is important to understand the behavior of coal in the course of methane extraction as well as for gas injection for ECBM recovery. Although most workers have assumed the linear deformation curve for coal to be elastic, it is really not so evident from other key research works [46, 47].

It is important to comprehend the stress-strain relation for coal in order to enable an accurate estimation of matrix deformation at low and high stresses. Researchers have conducted experiments and have presented theoretical models for quantifying the sorption induced strain on coal, using pure gases as well as gas mixtures at varying pressure and temperature conditions. Most of the research have been carried out for coal under unstrained conditions, while a more realistic approach would have been to analyze the rock deformation behavior in strained conditions. The rocks underground are subject to a certain amount of overburden pressure and undergo some compression causing closure of cleats during removal of gas during desorption. However, the two are not independently active and so, may not be the actual case [48] though the effects of overburden stress cannot be totally neglected.

3.1 Reviewing Development of Understanding on Coal Matrix Deformation

Briggs and Sinha [49] determined the sorption-related changes in coal at 2.07 MPa in CH_4 and CO_2 and found that the axial strain for methane ranged from 0.06 % to 0.30 %. Though the values for CO_2 induced swelling were higher, ranging from 0.34 % to 0.58 %, they showed that the induced strain is recoverable at ambient pressure. Reucroft and Patel [50] conducted dialatometric studies on elongate coal samples due to adsorption of N_2, He, Xe and CO_2. Volume increase of up to 1.3 % were obtained on exposure to CO_2, while negligible effects were obtained on exposure to N_2, He, and Xe. Reucroft and Sethuraman [42] obtained significant swelling from 0.75 % under 0.5 MPa to 4.18 % under 1.5 MPa for Kentucky coals of varying ranks. The magnitude of swelling decreased with increasing ranks and increasing moisture content of coals. Walker et al. [51] studied the deformational behavior of coals of varying ranks when subjected to CO_2 at different pressures and found that the maximum coal swelling increased from nearly 1 % at 0.68 MPa to around 4 % at 4.8 MPa. They observed an interesting phenomenon: expansion of coals at high pressures was not fully reversible when the pressure was reduced, due to permanent structural deformation as a result of CO_2 dissolution. The percentage of swelling increased with increasing pressure and decreased with high ranks. Although similar observations were made by earlier workers too when Moffat and Weale [52] reported that coal matrix swelling using methane recorded a maximum strain of 1.75 % with increase in pressure while the strain associated with desorption of methane was 1.49 % resulting in residual volumetric strain of 0.27 % for Cannock Wood Coal.

Harpalani and Schraufnagel [53] used methane gas for their study and obtained a linear increase in volume by 0.48 % due to adsorption, with maximum CH_4 pressure as 6.2 MPa. The decrease in the matrix volume was nonlinear as the pressure was completely reduced, resulting in a residual expansion. Ceglarska-Stefanska and Czaplinski [54] used CO_2 on coking coal as well as an anthracite and obtained differential swelling in directions parallel (maximum linear strain = 0.65 %) to and perpendicular (maximum linear strain = 0.92 %) to the bedding at pressures around 4.8 MPa. Ceglarska-Stefanska [55] compared the rates of adsorption/desorption with the rates of swelling/shrinkage using CH_4 at pressures reaching up to 4 MPa, keeping the temperature constant at 25 ° C and found that gas adsorption/desorption occurred faster than the matrix swelling/shrinkage. Differential swelling was found in directions parallel (maximum linear strain = 0.134 %) to and perpendicular (maximum linear strain = 0.175 %) to the bedding at a gas pressure of 3.04 MPa and the shrinkage was not the same as swelling leading to some residual expansion. The observation on differential rates of sorption and swelling was also made when the sorption of carbon dioxide was faster than the development of swelling strain, but at higher pressures (>4 MPa) the two occur simultaneously [56]. No change in sample dimensions were observed upto 60 % of gas adsorption in both the studies at low pressure levels for which it was hypothesized that "the delay in coal dilation at the initial low pressure levels causes the gas to enter the coal macro-pores, causing a minimum of volume change; increased swelling of coal takes place when the gas, at higher pressures, is forced into the micro-pores" [56].

Harpalani and Chen [57] attempted to eliminate the effects of overburden in matrix compression in experiments to study shrinkage of coal due to desorption of CH_4. They obtained a strain of 0.21 % due to desorption when pressure was reduced from 10.3 MPa to 0 MPa and found a linear relation between matrix shrinkage and adsorbed volume. Levine [58] used CH_4 and CO_2 for determination of matrix shrinkage/swelling in coal and found that swelling was greater in the plane perpendicular to the bedding in both the cases. The data also showed that sorption strain is not linear with pressure, but exhibits a curvilinear form that is steeper at a low pressure, becoming flatter at a higher pressure, resembling the sorption isotherm in shape. Ceglarska-Stefanska and Holda [59] also conducted studies to understand the sorption of various gases by the coal matrix and obtained a maximum swelling for CH_4 as 0.36 %. The other gases like H_2, N_2 and Ar induced swelling of 0.05 %, 0.15 % and 0.18 % respectively. There was almost negligible interaction between He and coal substrate. George and Barakat [48] used gas-saturated coals and found that the swelling due to adsorption was 2.16 % with CO_2, 0.38 % with CH_4 and 0.17 % with N_2 while there was a negligible compression of coal using He. The volume shrinkage in coals were less during desorption resulting in a permanent strain. Ceglarska-Stefanska et al. [60, 61] used a mixture of CH_4 and CO_2 and found that the matrix swelling perpendicular to bedding increased with pressure and reached upto 0.249 % at 3.70 MPa gas-mixture pressure while the same set of samples when exposed to pure methane pressure of 2.83 MPa manifested a strain of 0.16 %.

Chikatamarla et al. [62] examined the matrix deformation behavior of West Canadian sedimentary basin samples with different ranks—from sub-bituminous to medium volatile coals—using various gases. They showed that the volumetric strains are proportionate to the volume of the adsorbed gas. Maximum volumetric strains obtained were 9.33 % for H_2S, nearly 14 times greater than CO_2 (0.66 %), 20 times more than CH_4 (0.30 %), while nitrogen induced strain was almost negligible (0.03 %). Mazumder et al. [63] conducted experiments to replicate the underground in-situ conditions at total gas (CH_4 and CO_2) pressures ranging from about 4 MPa to nearly 23 MPa and obtained linear strains on the coal equal to 0.6 %. Siemons and Busch [64] obtained the swelling of coal at high gas pressures up to 20 MPa using an indirect approach and found that it ranged from 3 to 13 % depending on the type of coal. Day et al. [65] experimented on three Australian high volatile bituminous coals with gas pressures up to 15 MPa and obtained a maximum volumetric swelling ranging from 1.7 to 1.9 % with indications of a relationship between the percentage of swelling and the volume of CO_2 adsorbed. Zarebska and Ceglarska-Stefanska [66] experimentally studied the linear strains of coal for varying mixtures of CO_2 and CH_4, with maximum values ranging from 0.45 % to 0.8 % and 0.4 % for pure CO_2 and CH_4 sorption, respectively, for gas mixtures. Pone et al. [67] reported a three dimensional strain distribution due to interaction of CO_2 when injected in bituminous coal in which the positive strain due to swelling was 0.93 %, 0.94 % and 0.30 % along X, Y, and Z axes, respectively. However, the average volumetric strain was reported to be negative, indicating an overall volumetric reduction as an influence of stresses. Majewska et al. [68] conducted binary gas sorption experiments and found that swelling strain at 4.0 MPa was equal to 1.2 %. The maximum volumetric strain varied from 0.9 to 1.4 % for pure CO_2 and 0.25 to 0.35 % for pure CH_4. The volumetric strain consistently decreased due to increasing CH_4/CO_2 binary ratio. Pini et al. [69] investigated the role of adsorption and swelling on the dynamics of gas injection in coal; they obtained the swelling isotherms as a function of different gas pressures, using CO_2 and N_2 with maximum strain corresponding to maximum gas pressure. Detailed studies were carried out by van Bergen et al. [70] on the development of strain in unconfined coals for different gases and they established that the maximum equilibrated strain due to CO_2 was 2.24 % at 8.2 MPa while CH_4 and Ar showed maximum strain of approximately 0.65 % and 0.50 %, respectively. Swelling and sorption experiments by Battistutta et al. [71] revealed a fully reversible swelling in case of CO_2 with a maximum swelling nearly equal to 1.44 %. They obtained the swelling ratios between the maximum value in excess sorption as 1:1.5:2.6 for N_2:CH_4:CO_2 at 318 K.

Day et al. [73] experimented on the moist coals with pressures up to 16 MPa and found that the maximum volumetric swelling occurred from 2 to 5 % under dry conditions depending on the rank of coals. Day et al. [74] conducted swelling measurements in Australian coals for CO_2 and CH_4 and mixtures of both in fixed compositions. Helium was seen to completely displace an already swelled coal with CO_2 at 15 MPa. In another experiment, it was observed that the CO_2 completely displaced an already saturated coal with CH_4; swelling of coal was higher than before, supporting the fact that CO_2 has higher affinity for adsorption. Syed et al. [75] conducted swelling strain measurements and the results suggested that the sample pore size distribution has higher role in determining the swelling induced by CO_2 adsorption compared to adsorption of other gas molecules. Majewska et al. [76] simultaneously measured the induced strain, stress and acoustic emissions in coal upon sorption of CO_2 and observed that the swelling reduced by about 60 % upon application of axial stress. Vishal et al. [72] conducted triaxial experiments on Indian coal and measured coal swelling using a radial strain gauge put over coal core sample in triaxial conditions; they reported a deformation ranging 0.041–0.062 % due to the initial CO_2 flow (Fig. 4). Anggara et al. [77] experimented on low rank coals with supercritical CO_2. They observed that moisture was the deciding factor for swelling extent whereas the swelling behavior with

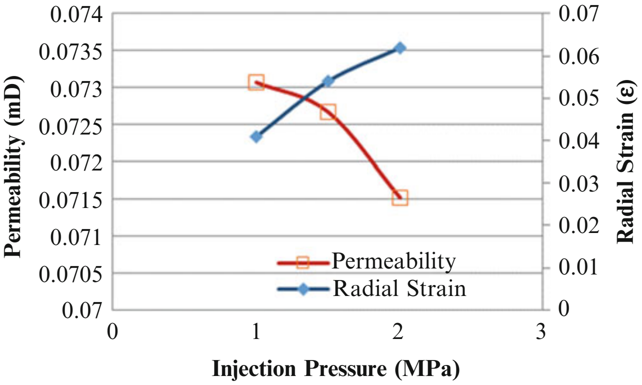


Fig. 4 Coal matrix swelling corresponds to the reduction in permeability with injection pressure during initial gas flow [72]

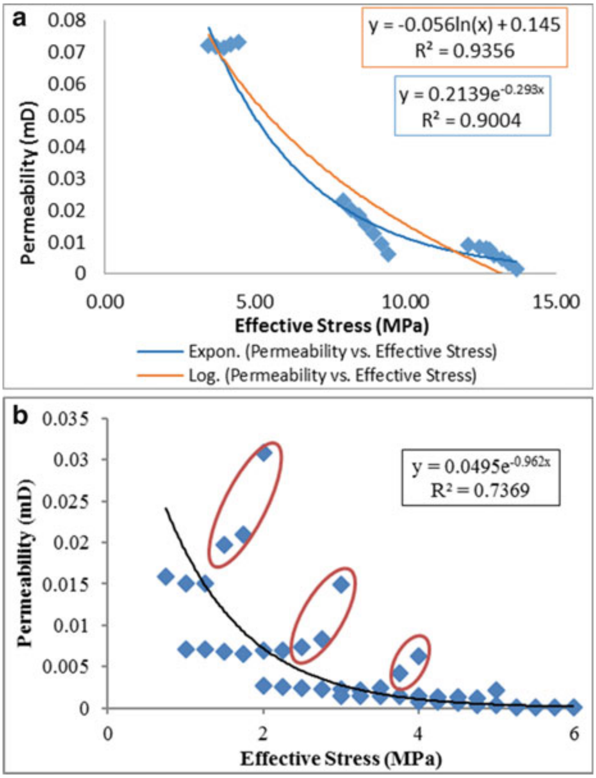
respect to bedding orientation was dependent upon megascopic texture. Works continue to establish this phenomenon and characterize coal from different basins from across the globe and this appears to be a significant challenge not only for the operations of CO_2 injection in coal but also the overall safety and stability of the system.

4 Permeability of Gas in Coal

Permeability in coal is a critical parameter that is affected by several inter-related phenomena such as shrinkage/swelling of coal matrix, gas slippage, geomechanical effects, cleat anisotropy and effective stresses. The coal matrix undergoes volumetric deformation, typically, swelling and shrinkage with gas adsorption and desorption, respectively; this alters the cleat apertures and therefore, significantly influences the in situ reservoir permeability. Further, permeability in coal is dependent on the effective stress. Several permeability models have been developed for coal seam gas production in the past few years [78–84]. Permeability in coal is commonly estimated using the Darcy's Law for interpretation of experimental results, provided the volumetric flow rate varies linearly with pressure gradient across the ends of the sample [85]. For higher flow rates, the pressure gradient may exceed that which is predicted by Darcy's law; such behavior is known as non-Darcy flow. In recent times, studies have been done in permeability evolution upon variation of different parameters [1, 32]. As discussed earlier, coal contains dual porosity. Similarly, on the basis of disposition and arrangement of cleats, coal exhibits flow anisotropy. Permeability of coal is at its maximum in the direction of the face cleats. From several studies [86–88], the importance of permeability anisotropy calculation for the coal seam/basin for correct estimation of gas flow behavior during coal bed methane production and/or carbon dioxide sequestration is established.

Coal exhibits differential sorption affinity to different gases and hence, depending on the sorptive and non-sorptive gas type, the permeability of coal varies. Early research works established that coal permeability is significantly lower for methane as compared to nitrogen primarily because of the high sorption affinity of methane in coal [89, 90]. Patching [89] however, concluded that the molecular diameter of gas type inversely affected the permeability attributes of coal. Skawinski [91] showed that CO_2 resulted in still higher reduction in coal permeability as compared to CH_4 and N_2 due to the high adsorption affinity of the coal mass towards CO_2. Cui [82] found that the molecule size of the flowing medium along with the pore structure of coal influenced the selective adsorption and gas diffusion in coal. Results showed that the gas desorption and diffusion rate was inversely related to its kinetic diameter, while it varied directly with sorption affinity. This established that the pore structure and molecular geometries of the phases play a more dominant role over mere sorption affinity. Robertson [92] also demonstrated a decreasing order of coal permeability with N_2,

Fig. 5 Variation of permeability of coal in intact (**a**) and fractured (**b**) coal specimens [1]



CH_4 and CO_2, respectively. The major application of this phenomenon would be in enhanced recovery of CBM using CO_2 sequestration.

Al-hawaree [93] tested the change of permeability of coal samples from Alberta, Canada using CO_2 and CH_4 and found that at a constant effective stress in coal, permeability to CO_2 reduced upto 84 %, while that in CH_4 reduced upto 50 % in a comparable range of increasing pore pressure. Li et al. [94] used Powder River Basin coal and utilized pure N_2, CH_4, CO_2 and mixtures of N_2 and CO_2 under a constant effective stress. They established that coal permeability decreased with an increase in the gas sorption pressure, while an increase in the CO_2 component in the flowing mixtures reduced permeability to a greater extent. For pure gases, CO_2 caused the highest permeability reduction followed by CH_4 and finally N_2. Vishal et al. [1] estimated the changes in permeability of intact and fractured coal respectively with effective stresses (Fig. 5a, b).

Several field based studies have also demonstrated reduction in coal permeability with different phases of gas. Reeves [95] detailed the observations from first field scale pilot of enhanced CBM recovery using CO_2 in San Juan Basin, USA. Reduction in injection rate of CO_2 took place with time, from 5mscf/day to 3mscf/day due to loss in injectivity, which shows influence of CO_2 on permeability of coal. Mavor et al. [96] reported a reduction of nearly four times in CO_2 permeability in coal in ECBM pilot tests in Alberta, Canada. Shi et al. [97] showed that injection of pure N_2 over some days could reverse the permeability reduction due to CO_2 injection in coal beds of Yubari pilot project, Japan. Botnen et al. [98] showed that a reduction by nearly ten times in CO_2 permeability took place in Williston Basin (North Dakota) for lignite.

Mazumder et al. [99] observed multifold increase in CO_2 permeability with respect to reservoir pressure owing to matrix shrinkage. Qu et al. [100] developed models to see the evolution of permeability with CO_2 injection at different temperatures. They observed a maximum permeability reduction of 95 % at 278.15 K. Sander et al. [101] performed core-flood experiments of CO_2-ECBM on two different samples of Australian coal and found the results to be similar to that observed in past studies. No permeability changes were observed in reverse core flood with CH_4 displacing CO_2, thus agreeing with the fact that CO_2 has greater affinity to coal.

5 Adsorptive Weakening of Coal

Previous studies conducted on coal from different basins around the globe have established the adsorptive weakening of coal. Ettinger and Lamba [102] mechanically crushed the coal samples and used the amount of 0.5-mm sieve dust residue as an indicator of coal strength. The samples were first evacuated and then subjected to air and CO_2 saturation at a pressure of 4.0 MPa. The results indicated that the "disturbed" coal samples showed strength reduction by a factor of 0.75 in CO_2 environment as compared to air. Similarly, Tankard [103], measured the changes in surface area of crushed coal due to gas adsorption to understand the influence of sorptive and non sorptive gases on coal. Czaplinski and Holda [104] experimented on the crushing strength of coal in normal air at 0.1 MPa and in CO_2 environment at 2.0 MPa and found that the amount of coal extracted by crushing was much higher when saturated with CO_2 than in air. Holda [105] used the same setup and introduced methane along with CO_2 and air in the scope of investigation. The results revealed that major reduction in coal strength was observed in samples saturated with CO_2 than the ones with CH_4. In contrast to the findings of earlier researchers, Ates and Barron [106] conducted Brazilian tests on Canadian and Australian coals and indicated that no significant reduction in coal strength occurred due to CO_2 saturation upto gas pressure less than 20 atm. Later, Aziz and Ming-Li [107] investigated the effects of gas sorption induced changes in coal strength in terms of coal drillability characteristics. They used pure CO_2 and CH_4 along with their mixtures at variable pressures. The results indicated that CO_2 caused maximum reduction in coal strength as a higher drilling rate and coarser drilling particles were observed in this case. They also showed that an increase in gas pressure increased the drilling rate. This implies that gas type as well as gas pressure influence the strength characteristics of coal.

Table 1 Comparative chart on saturation effects on the uniaxial compressive strength (UCS) and Young's modulus (E) of bituminous coal, Jharia [112]

Condition	UCS (MPa)	ΔUCS (%)	E (GPa)	ΔE (%)
Untreated	15.29 ± 0.32	–	5.34 ± 0.19	–
CO_2	12.62 ± 0.44	17.6	3.94 ± 0.25	26.2
Water	11.39 ± 0.59	25.5	3.32 ± 0.27	37.8
Water + CO_2	$10.95 + 0.41$	28.4	2.79 ± 0.24	47.8

The explanation to this was found in Viete and Ranjith [108] who explained that with an increase in confinement on coal, there was reduction in lowering of adsorptive surface energy and hence, not much weakening was observed. For reduction in the strength of coal under triaxial conditions, the confinement on the coal must be less than the tensile stress of the coal. Karacan [109] explained that coal matrix swelling due to gas adsorption led to an increase in the distance between the atoms in the coal, leading to reduction in its strength. The increase in vitrinite content and increasing CO_2 pressure lead to higher strain and therefore higher reduction in the strength of the coal [109]. These studies are limited to only certain coal and the role of gas injection in coal strength under varied conditions of confinement, coal types and gas phases are still ambiguous and need further investigation. Different ranks of coal, coal with different maceral content are expected to undergo differential reduction in strength due to gas injection. Another study by Pan et al. [110] on Australian black coal indicated no direct evidence of the sorptive weakening character of coal and they suggested that the effect might vary from one coal type to the other.

Hol and Spiers [111] from the laboratory experiments, emphasized on the effects of plasticization that is believed to weaken the internal bonds in coal. They observed that most of the microfractures developed parallel to the bedding plane. A detailed investigation was conducted by Vishal et al. [112] on behavior of coal under saturation with both CO_2 and moisture. They found that the CO_2 treatment of moisture saturated coal reduced the strength of coal by almost 28 % and the Young's modulus by nearly 48 % (Table 1). The acoustic emission results also showed major difference in the pattern of failure of these samples in uniaxial loading. These findings highlight the sorption induced weakening in coal and that it should be addressed before any CO_2 storage operation in coal seams.

6 Conclusions

This chapter reviews the physical attributes associated with the coal-fluid interactions during CO_2 sequestration with/without simultaneous CBM recovery. It is evident that adsorption of CO_2 in coal causes coal matrix swelling which in turn leads to reduction in the strength of coal. Different adsorption models are applied to understand the adsorption phenomenon in coal. Permeability is one of the most important parameters in methane extraction or CO_2 injection. It depends on several factors such as confining pressure, effective stresses, pore pressure, coal deformation, gas type etc.

CO_2 storage in coal seams has been tested in the past for feasibility at all levels ranging from laborious laboratory scale experiments to pilot scale demonstrations and there is still scope for more research at these levels of testing. But the present scenario demands urgent deployment of such techniques to be able to allow mankind to continue using fossil fuels for a time, until sustainable energy can take charge, without disrupting the climate change predictions. The major advantage of sequestration in coal is that due to the widespread scenario of coal worldwide, technology transfer is the easiest in contrast to geological storage in aquifers and basalt. Geosequestration may be the fastest mode to effectively tune the anthropogenic CO_2 cycle thus making it a forerunner in climate change mitigation initiatives and a buffer directed towards any delay in sustainable energy commercialization.

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