Adsorption Factors in Enhanced Coal Bed Methane Recovery

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Enhanced coal bed methane recovery using gas injection can provide increased methane extraction depending on the characteristics of the coal and the gas that is used. Accurate prediction of the extent of gas adsorption by coal are therefore important. Both experimental methods and modeling have been used to assess gas adsorption and its effects, including volumetric and gravimetric techniques, as well as the Ono–Kondo model and other numerical simulations. Thermodynamic parameters may be used to model adsorption on coal surfaces while adsorption isotherms can be used to predict adsorption on coal pores. In addition, density functional theory and grand canonical Monte Carlo methods may be employed.

ECBM gas adsorption coal

1. Introduction

Coal bed methane (CBM) has been extracted from coal seams for many years [1]. This methane is held in micropores [2] and so numerous methods have been developed based on gas injection techniques to remove the maximum possible amount of methane from these micropores. CBM relied on the natural pressure in the coal bed, but these methods were unable to achieve complete extraction [3]. Since then, other technologies have been developed, including the use of polymers, water injection, and proppant injection [4][5][6][7][8][9][10]. Among the various method, gas injection has been found to provide maximum methane recovery of up to 90% [11]. The gases commonly used in enhanced CBM (ECBM) recovery are carbon dioxide (CO_2) and nitrogen (N_2) or and mixture of the two [12][13][14][15].

The extraction process comprises gas injection into the CBM reservoir, followed by the selective adsorption of the gas on the coal surfaces and in the coal pores, methane desorption from the coal matrix, and methane flow along fractures in the bed based on Darcy's Law [16][17][18][19][20] (**Figure 1**). Both the gas injection and adsorption rates in the coal bed are critical because these factors affect the coal structure and thus the extent of methane recovery [18][21][22][23]. Based on accurate adsorption analyses, including factors such as bed swelling and permeability, the effectiveness of ECBM extraction can be predicted [18][24]. Accurate predictions of gas adsorption must also take into account the possibility of sequestration of the injected gas [17]. Despite that the publication of many research and review articles on the subject of gas adsorption on coal, the adsorption of gases by coal beds based on actual coal pore morphologies and chemical structures poorly understood [25][26].

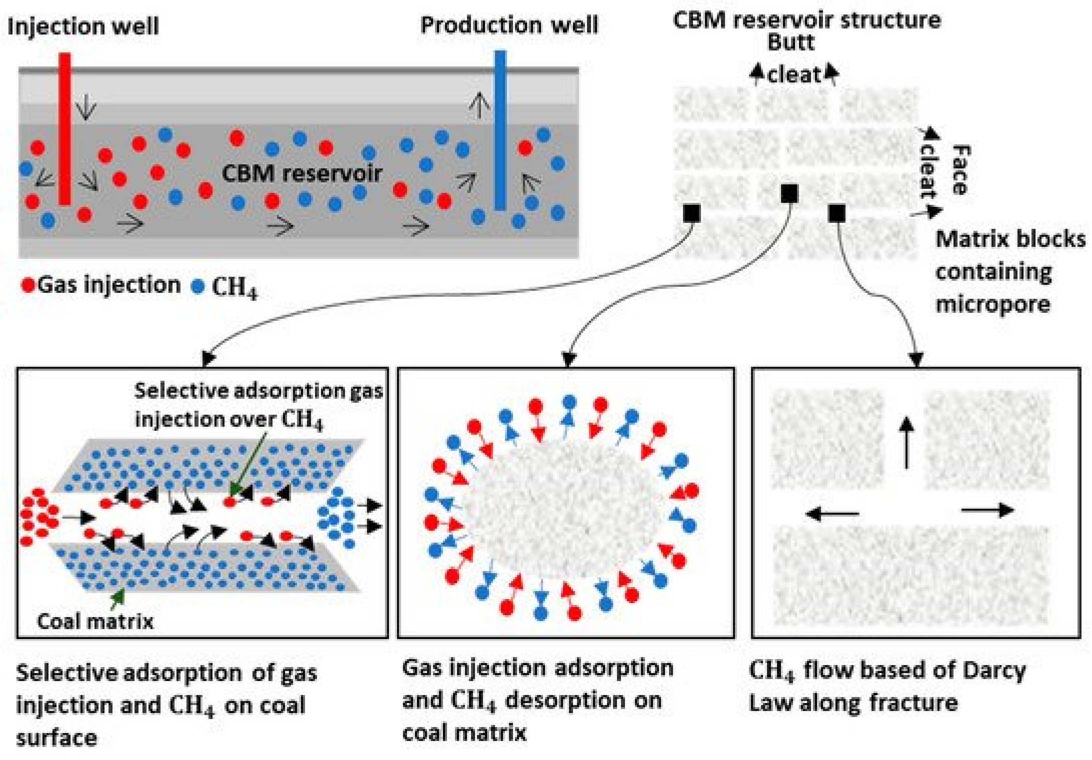

Figure 1. The ECBM extraction process based on gas injection. Adapted from [16][17][18][19][20].

2. Methane in Coal

Methane is present in coal beds both as an adsorbed gas (accounting for 80–90% of the entire methane content in a coal seam) and a free gas [16]. The latter can be compressed in pore spaces, condensed as a solid or liquid, dissolved in the coal structure or adsorbed on surfaces [2]. Coal contains methane gas because of biogenic and thermogenic processes [27] that occur during coalification with resultant storage of the gas in the coal seams [19].

Biogenic methane produced by bacterial activity at shallow to moderate depths (<500 m) [2][28][29] begins with fragmentation of the coal macromolecules via two main processes; exfoliation and/or anaerobic oxidation [30]. The biogenic processes begin with oxygen consumption after which biologically-generated CO_2 is converted to methane [29]. The anaerobic oxidation reactions are promoted by various bacterial species capable of oxidizing aromatic and aliphatic structures to CO_2 [30]. The majority of the biogenic methane and CO_2 generated in this manner are most likely dissolved in water and removed from the system during compaction and coalification [31].

The thermogenic formation of CBM results from kerogen or the cracking of heavier hydrocarbons and increases with depth [32]. Thermogenic processes that occur in deep coal [33] at higher pressures increase the coverage of the coal surface by the CBM and result in stronger interactions between adsorbate molecules [34]. Although the composition of coal bed gases does not have a strong relationship with either coal rank or depth, thermogenic generation usually begins in highly volatile bituminous rank coal and increases with rank [32]. The thermogenic processes cause coals with higher ranks to have greater holding capacities such that they retain more gas, and also yield micropores that act as methane reservoir [30][35][36][37].

CBM can be produced at almost any time during the coal life cycle based on methanogenic bacterial growth in response to heating if the coal is uplifted and favorable subsurface environmental conditions are restored [30]. Secondary biogenic gases are also generated through bacterial metabolic activity based in the introduction of bacteria by meteoric waters migrating through permeable coal beds [31]. The biological methane in coal be continuously produced, although thermogenic gases tends to result in higher total gas contents in coal beds compared with pure biogenic-derived gases [30].

3. Gas Adsorption Characteristic of Coal

Gas adsorption on coal is influenced by the specific characteristics of the coal. Research has confirmed the effects of the coal condition and the type of coal, as well as the moisture content, ash yield, maceral content and coal pore distribution on the efficiency of ECBM extraction.

3.1. Effects of Sample Condition

The sample aspect that has most frequently been shown to affect coal gas adsorption tests is particle size. Specifically bulk samples adsorb gases more slowly than crushed coal samples [38][39]. The crushed coal used for adsorption analyses is typically in the size range of 100–60 mesh [38][40][41][42] while bulk coal specimen are usually approximately 2 cm cubes [40]. The crushed coal has a higher diffusivity and requires a shorter measurement time to achieve equilibrium compared with coal blocks [39][43][44].

Even so, crushed coal has several detrimental effects on adsorption. As an example, this material will have a damaged pore network in which closed pores have been opened. Therefore, the sample surface area will have been increased so that the adsorption capacity is artificially improved compared with the original state [38]. The crushing of coal also decreases the moisture level and increases the amount of adsorbed gas [40]. When crushed coal is used, it is nearly impossible to observe coal shrinkage or swelling because of adsorption, in contrast to trials using solid coal [45].

3.2. Moisture Effects

Moisture is an important factor in adsorption because water molecules are highly polar [46], and so can modify, the gas adsorption kinetics, mechanisms and capacity [43][46][47][48][49][50]. A comparison of adsorption during ECBM extraction trial using moist and dry coals has shown that dry conditions provide the highest gas adsorption capacity and saturation values [51]. This occurs because the adsorption sites that were originally occupied by moisture become available for methane adsorption [49][52][53][54].

Dry coal has greater coal gas adsorption capacity but can yield a large correction factor because coal in the field contains natural moisture [47][55]. Natural (or inherent) moisture affects the methane adsorption capacity differently for each coal rank. Specifically, low-rank coals exhibit greater capacities for water retention medium-rank coals show pore-blockage and fewer micropores as a result of water adsorption which limits the gas adsorption capacities and high-rank coals contain numerous in micropores that provide sufficient pore space for the exchange of water and methane [49].

3.3. Ash Yield Effects

Ash yield is attributed to pore infilling, blockage cleats and fracture systems resulting from extraneous mineral matter (such as clays and carbonates) in coal [56]. The adsorption capacity of coal is decreased with increases in the mineral ash content [57] because this material reduces the storage capacity [53] and blocks gas migration [38]. The presence of mineral matter indicates that the increasing pore volume, especially in open pores and macropores, such that gas adsorption is inhibited at faster desorption rates [2][58][59]. For these reasons, coal having a high ash yield is generally not suitable for ECBM recovery with gas injection because it cannot absorb the injected gas or requires the application of high pressures and temperatures for adsorption [60].

3.4. Maceral Effects

In coal, organic matter is known as the maceral component and this material affects gas adsorption and absorption [41]. Generally, the feasibility of performing ECBM recovery is based on assessing the vitrinite content of coal [17].

Vitrinite is a type of maceral that affects the pore structure of the coals [61], especially the coal micropores and pore distribution [62]. A higher vitrinite content leads to a higher void volume [63], greater specific surface area (SSA) [45], increased adsorption capacity [56], and decreased desorption rate [2][53][64]. Coal that is rich in vitrinite also reacts more effectively to CO_2 injection and undergoes swelling [65][66].

Liptinite is another type of maceral that affects the mesopores in coal [62]. By encouraging surface diffusion, liptinite can promote the adsorption of CO_2 and also act as a medium for gas transport by adsorbing CO_2 while acting as a catalyst [65]. Inertinite differs from vitrinite and liptinite that it contains more macropores and fewer micropores [67]. As a result of the dominance of macropores in this material, liptinite lower the apparent surface area of the coal [62], resulting in a shorter time being required to achieve equilibrium [68], and producing significant swelling upon CO_2 injection [66].

3.5. Coal Pore Effects

Methane in coal is stored on the walls of micropore networks [2] an various methods are used to understand the manner in which gases can be extracted from these micropores. ECBM recovery research has demonstrated that these pores modify the adsorption and flow of gases that are injected into coal or other porous media [69][70].

The pore volume in coal is determined by its thermal maturity [71] such that increasing maturity increases the adsorption capacity [2]. **Figure 2** presents pore size distribution curves for coals from low rank to high rank as obtained from nuclear magnetic resonance (NMR) analyses [72].

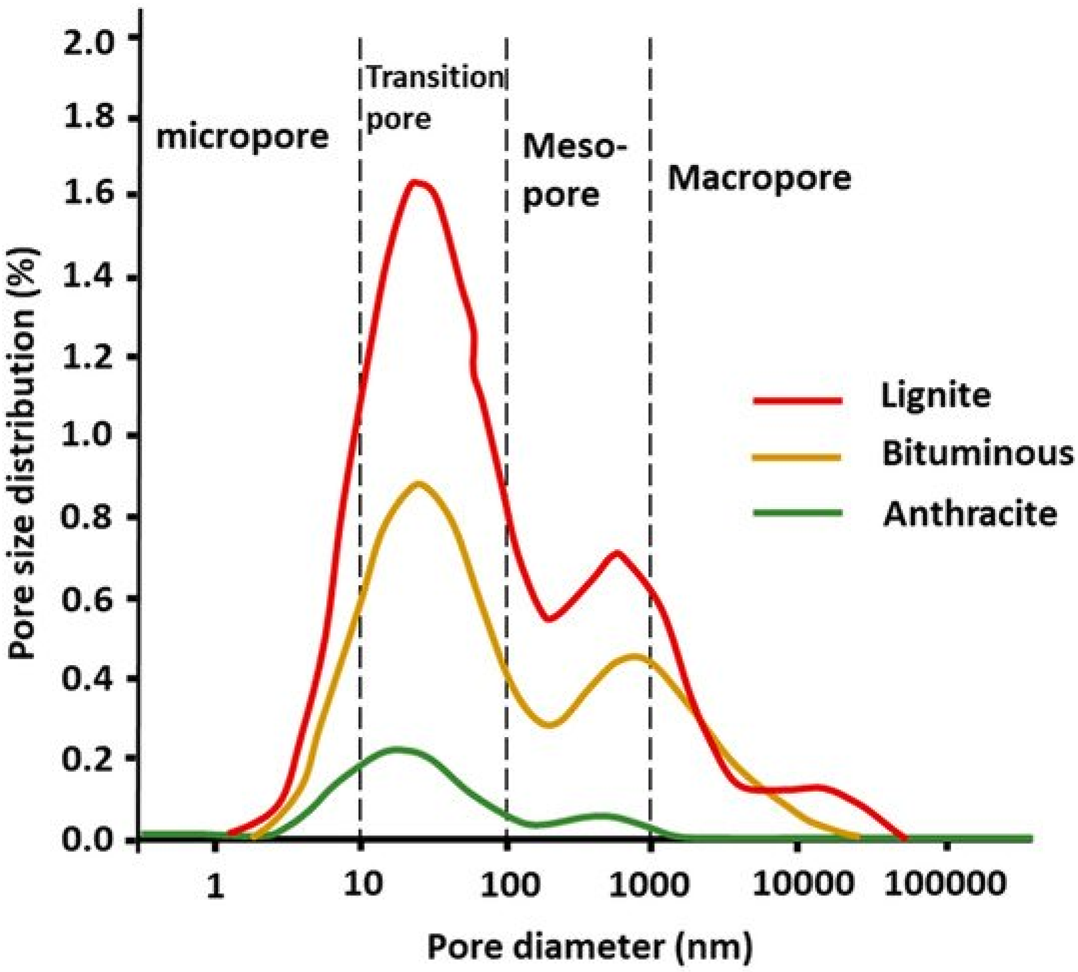

Figure 2. Pore distributions in different coal ranks as determined using an NMR method. Modified from [72].

Low-rank coal contains primary epigenetic pores having irregular shapes and poor connectivity. Although the dehydration of lignite to low-rank coal reduces the moisture and oxygen-to-carbon ratio of the material [36], low-rank coal exhibit a high degree of porosity and low pore compressibility [14].

Metamorphism changes pores into circular, oval or slit morphologies [73][74] and medium-rank coal contains pore sizes ranging from macropores to micropores [75]. Medium-rank coal with a high proportion of micropores is the most suitable for industrial methane production [73].

High-rank coals contain primarily micropores with limited pore connectivity as a result the coalification process [49][62]. Coalification also leads to smaller coal pore, larger surface areas, a greater number of micropores and a higher methane content [52][76][77]. Although increases in coal rank are associated with increases in the methane content [78], the pores gradually close and form flattened structure that make gas absorption impossible [35][37][62]. As a result of the small pore surfaces, gas injection into high-rank coal must be performed at high pressures [79].

4. Gas Injection for ECBM Recovery

As noted, CO_2, N_2 and their mixtures are commonly used for ECBM extraction, and the injection of pure or mixed gas will lead to different adsorption effect, as explained in this section.

4.1. CO_2 Injection

CO_2 is an acidic gas [80] that is widely for ECBM recovery because it can extract methane with significant efficiency [12]. Coal has a high adsorption affinity for CO_2 and so this gas is adsorbed rapidly, whereupon it seeps into micropores [47][64][65][81][82][83][84]. The CO_2 molecule also has a small kinetic diameter and so can replace methane originally present in the micropores [12][14][85]. However, the CO_2 storage capacity is affected by temperature and pressure, both of which can change the coal structure and permeability [35][43][86][87][88][89][90].

4.2. N_2 Injection

N_2 is used for ECBM extraction because N_2 promotes methane desorption from the coal matrix [12][14][91][92]. N_2 reaches equilibrium quickly, leading to a more rapid response [91][93]. N_2 adsorption increases with increases in pressure, although, N_2 undergoes weak interactions with adsorbents [94][95]. N_2 injection also alters the coal pore structure and increases the transition pore volume such that the pore volume, pore size distribution, and connectivity are all increased [96].

4.3. Mixed Gas (CO_2-N_2) Injection

ECBM recovery experiments using gas mixtures have been carried out, Such mixtures have been found to be applicable to low-permeability coal [13][97] because N_2 prevents expansion of the coal matrix and increases the diffusion coefficient to provide faster methane extraction [14]. However, such mixtures are not suitable for carbon sequestration [98].

The desorption of methane is enhanced in the case that the mixed gas has a CO_2 concentration of less than 10%. Increasing the proportion of CO_2 increases the probability of adsorption while decreasing the desorption of methane [99]. The adsorption selectivity obtainable from a mixed gas injection may be calculated as [99]

$$S_{\mathrm{CO_2/N_2}} = \frac{(x_{\mathrm{CO_2}}/x_{\mathrm{N_2}})_{adsorbed}}{(y_{\mathrm{CO_2}}/y_{\mathrm{N_2}})_{bulk}}, \quad (1)$$

where $S_{CO2/N2}$ is the adsorption selectivity and, x and y represent the mole fractions of each gas in the adsorbed and bulk phase, respectively. An adsorption selectivity of 1 indicates that N_2 is adsorbed more strongly than CO_2 while a value greater than 1 indicates the opposite.

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