



Changes in gas storage and transport properties of coal as a result of enhanced microbial methane generation



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ABSTRACT

This study was aimed at identifying the changes in coal storage and transport properties affecting gas production from coal-gas reservoirs, when treated with microbial consortia to generate/enhance production of methane. The work expanded on the technology of bio-conversion, first proposed in order to imitate the natural/microbial process of biogenic gasification leading to recharging coalbed methane reservoirs or, setting up natural gas reservoirs in non-producing coalbeds or, to coal waste, typically in the form of fines/ultra-fines. The pressure parameter was considered critical since, with continued production of methane, the produced gas would first diffuse into the coal matrix and get adsorbed with increasing pressure. During production, the pressure would decrease and the process would be reversed, gas diffusing out of the coal matrix and arriving at the cleat system.

The experimental work tested the variation in the sorption and diffusion properties of treated coal, post continued bio-conversion since these are the first two physical phenomena in CBM production. During the first phase, single component sorption–diffusion experiments were carried out using methane and CO₂ on virgin coals retrieved from the Illinois basin. Coals were then treated with nutrient amended microbial consortia for different periods. Gas production was monitored over thirty and sixty day periods of treatment after which, sorption–diffusion experiments were repeated on treated coals, thus establishing a trend over the sixty-day period. The sorption data was characterized using the Langmuir model. The variation trend in the value of diffusion coefficient, *D*, was also established as a function of pore pressure.

The results indicated an increase in the sorption capacity of coal as a result of continued bioconversion. This was attributed to increased pore surface area due to microbial actions resulting in changes in the pore size or, creation of new pores. It was further shown that the rate of diffusion increased, especially for methane, which exhibited rates higher than that for CO₂. These findings clearly support improved gas storage capacity with continued bio-conversion as well as significantly enhanced diffusion rates. As a continuation of this effort, change in permeability, the second gas transport phenomenon in coal-gas production, is currently being evaluated.

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1. Introduction and background

The origin of methane in coal seams can be distinctly related to two processes: thermogenic and biogenic. Thermo-catalytic conversion of coal is initiated at a temperature greater than 70 °C. At such temperatures, when coal attains a rank of approximately 0.5–0.6% vitrinite reflectance (high volatile, bituminous), continued application of heat and overlying pressure over time results in devolatilization of thermogenic gases, like CO₂, H₂O, CH₄, C₂H₆, H₂S and other higher hydrocarbon gases [1,2]. Biogenic methane,

as the name suggests, has its origin in the biosphere of the subsurface, consisting of various forms of microbes, known as methanogens. Primary biogenic methane and CO₂ are formed microbially during the initial stages of peat formation at shallow depths. Biogenic action from microbes is believed to generate in excess of 6% CO₂ in the northwest San Juan Basin [3]. Due to high porosity and lower burial rates, primary biogenic methane is volatilized over time or dissolved in water and expelled during compaction [4]. Late stage biogenic methane, also known as secondary biogenic methane, is formed post-compaction in all ranks of coal due to combination of active groundwater flow recharging underground systems with suitable microbes, along with uplift of the basin helping in meteoric recharge [1,4].

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Taking cue from the widespread application of microbes in the oil industry as a means to increase overall recovery, and the impact that naturally occurring microbes have had in producing methane in coal, Scott in 1995 introduced the concept of microbially enhanced coalbed methane (MECBM) [5]. It aimed at replicating the natural process of secondary biogenic gasification by treating coal with methanogens along with suitable nutrient amendments and trace elements. Studies conducted by Strapoć [6] and Opara [7] further provided substantial evidence of the potential of generating methane by treating coal fines with bacterial inoculations.

Research in the area of MECBM and bio-conversion has accelerated in the last few years for several reasons. First, natural gas is considered a cleaner and, therefore, preferred source of energy. Second, this technology is applicable to unmineable coals and abandoned coal mines with large amounts of coal left behind. Third, the technology is applicable to coal waste in the form of fines or poor quality material. Fourth, several regions with significant coalbed methane (CBM) activity are maturing, thus providing spread out wells along with other supporting infrastructure required for injection of microbes/nutrients as well as gas recovery and utilization. Finally, the technology is applicable to CBM wells that are abandoned due to poor production rates and recovery, like in the Illinois Basin.

The general advance of research has been toward characterizing and improving the microbial solutions, primarily aimed to economically increase methane production. It has been recognized that the process of bio-conversion, in general, is a relatively slow process. Significant production of methane has been observed over a period of thirty to sixty days, or even higher in some instances [7,8]. These studies have been conducted on powdered coal, providing additional surface areas for the microbes. Production rates for in-situ deposits are expected to be even lower. Under such time dependent production characteristics, it is important to identify the properties of the reservoir affecting flow, and also document the changes in these properties. Identifying, and if needed, improving flow characteristics of produced gas, along with engineering optimum nutrient solutions for the microbial population to thrive in would help decide the viability of the process in terms of techno-economic feasibility. The work reported in this paper provides a starting point to identify such changes, specifically in the sorption–diffusion properties of coal as a result of bio-conversion.

Coal is generally characterized as having a micro- and macroporosity structure. Micropores occur as part of the coal matrix and serve as a storehouse for over 95% of the gas in adsorbed form [9]. The macropore system consists of a network of closely spaced natural fractures surrounding matrix blocks, which is called the cleat system [10]. Flow of methane in the cleat system is permeability-controlled and is dependent on cleat parameters, like the aperture, continuity and spacing. The movement of gas in the coal matrix, on the other hand, is diffusion-controlled and is dependent on pore size, distribution and connectivity. For under-sized waste coal, microporous characteristics are often intact while the macropore system is non-existent.

Gas is stored in coal primarily by adsorption on organic surfaces. For storage of produced gas, it would first diffuse into the micropores onto the sorption sites, where it would get adsorbed. Since coal has a very large internal surface area and methane is tightly packed in a monomolecular layer, large quantities of gas can be adsorbed in the coal matrix. For a given pressure, the amount of gas sorbed is best described by an isotherm, a plot of the volume of sorbed gas as a function of pressure. The most commonly used adsorption model for coal is the Langmuir isotherm. It is simple to use and works well for coal-methane system. The diffusive movement of methane/CO₂ in coal is best described by Fick's second law of diffusion. Diffusion is characterized by the diffusion coefficient (*D*). Given that bio-conversion of coal is expected to

change the physical structure of coal, two properties that would be potentially impacted are the sorption capacity of coal, that is, the ability of coal to store gases, and diffusion in the microstructure of coal. This study, is therefore aimed at evaluating changes in these two properties with continued bio-conversion.

2. Experimental setup and procedure

2.1. Sample procurement and preparation

Blocks of coal for the experimental work were obtained from the central Illinois basin. Details of geographical location has been previously reported by Zhang et al. [8]. The coal was then ground and sieved to obtain a sample size of 40–100 mesh (0.0425–0.0149 cm). This size has been shown to remove the cracks and macropores present in coal completely and yet preserves the microstructure of coal, ensuring that the movement of gas is purely diffusive in nature [11].

The coal sample was then divided into three parts, ~60 g each. Sorption–diffusion experiments were carried out for all three sets. The first set was untreated, virgin coal. The results from this provided a measure of the baseline characteristics. The other two samples were then treated in microcosms with an adapted microbial consortium initially developed from the formation water retrieved from a CBM well [8]. Briefly, each microcosm contained 10 g coal and 45 ml of sterilized medium [12]. Once 5 ml of the adapted consortium was added, each serum bottle was closed with a butyl rubber stopper and sealed by an aluminum crimp. All bottles were stored at 28 °C in a dark environment. The amounts of methane and CO₂ in the headspace were measured at the end of thirty days for the second sample and sixty days for the third sample using gas chromatography. Replicates of the microcosms were discarded at each time point for analyzing sorption–diffusion behavior of the residual coal. A total of twelve reactors were used, of which six was treated for thirty days and the remaining six for sixty days. Detailed information about the reactor setup can be found in Zhang et al. [8].

Prior to conducting the sorption and diffusion experiment, six grams of sample was used for moisture and ash analysis following the ASTM procedures (ASTM D3173-87, 1987; ASTM D3174-04, 2004). Five grams of the sample was used for the ultimate analysis tests following the ASTM standards (D3176-74) to obtain the mass fractions of C, H, N, S and O.

2.2. Gas chromatograph

The gas chromatograph, with a flame ionizing detector (FID), was used to measure the methane and CO₂ content in the headspace of serum bottles. Briefly, a 50 µl aseptic syringe connected to a sterile 25 gauge needle was used to withdraw the gas sample and inject it in the GC column. The carrier gas (Argon) flow rate was set at 10.1 mL/min with a velocity of 55 cm/s. The isothermal zone temperatures for the injector and detector was set at 75 °C and 310 °C respectively. The retention time for methane was 4.73 min and that for CO₂ was 6.71 min. Calibration curves for methane and CO₂ were established using gas standards.

2.3. Sorption–diffusion

The primary component of the experimental setup was a high pressure vessel assembly, consisting of a sample cylinder and a reference volume connected by a two-way valve and a micro-filter to prevent movement of coal particles with changes of pressure. The setup was placed in a constant temperature bath, set at 88°F and capable of maintaining the temperature to within 0.2 °F of the desired temperature. This is important since the processes of sorp-

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