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Development of a nutrient recipe for enhancing methane release from coal in the Illinois basin

Ji Zhang[a], Zheting Bi[a], Yanna Liang[a,b,*]

[a] Department of Civil and Environmental Engineering, 1230 Lincoln Drive, Southern Illinois University Carbondale, Carbondale, IL 62901, USA
[b] Department of Environmental and Sustainable Engineering, 1400 Washington Ave, State University of New York at Albany, Albany, NY 12222, USA

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ABSTRACT

Recent literature has shown that in order to enhance methane release from coal through biogasification, suitable nutrient solutions are needed to stimulate microbial activities toward coal depolymerization and conversion to biogas. Specific for bituminous coal in the Illinois basin, a nutrient recipe that can be used to enhance coal biogasification in situ is not available yet. To develop such a recipe, the formation water and the indigenous microbial community were first characterized in detail. Based on these characteristics and our previous experience working with Illinois coal, four parameters, Fe powder, methanol, ethanol, and trace minerals were optimized through using a Box-Behnken design. The optimal condition predicted by the models was: Fe-powder at 74 mM; methanol at 97.9 mM, ethanol at 100 mM, and trace minerals at 100%. Under these conditions, the predicted methane yield and content was 1417.35 ft^3/ton and 80.7%, respectively. These results were then validated by experimental studies. In addition, each added component was evaluated in terms of its contribution to methane generated. Specifically, the role of coal in the biogasification process was studied against two other solid materials. Overall, this study demonstrated that coal can be converted to methane and nutrient solution can definitely enhance methane release from coal. The real effect of this recipe in improving methane release from coal in situ needs to be further evaluated in a field scale.

1. Introduction

Coal Bed Methane (CBM), a naturally occurring methane gas existing in coal seams, is becoming an increasingly important part of US's energy portfolio. In comparison to coal, burning natural gas releases half of the amount of CO_2, 80% less CO, and NOx, which makes CBM a relatively environmentally friendly energy resource. According to data published by the Energy Information Administration (EIA, 2015), CBM represents up to 20% of the world's natural gas reserves. The US was the largest CBM production country in the world with ~1.25 trillion cubic feet (TCF) output in 2015, which was approximately 4.3% of the total US natural gas production and 10% of the proven reserves in the same year. The majority of CBM production in the US has come from three basins: The Black Warrior (Alabama), the San Juan (New Mexico, Utah, Colorado), and the Powder River (primarily Wyoming) (Moore, 2012). Although very limited amount of CBM has been produced in the Illinois basin, this could be changed soon (Eble et al., 2005). The potential for commercial quantities of CBM in this basin is based on the fact that the IL basin has vast coal reserves (Morse and Demir, 2007). The IL basin has over 325 billion tons of remaining coal resource that has been estimated to contain 11 TCF or more of CBM (Demir et al., 2004). Recently, there has been increasing exploration and development activities in this basin, resulting in some, but still insufficient CBM production in this place (Kronkosky, 2009). To aid these efforts and increase methane output from this basin, cost-effective and efficient approaches need to be explored.

Scott (1999) defines the concept of Microbially Enhanced Coalbed Methane (MECoM) as the introduction of microbial consortia and nutrients into coal beds. This not only has the potential to produce fresh methane from coal but could also increase reservoir permeability via microbial consumption of coal. During recent years, MECoM has attracted extensive interest globally among researchers and investors since this approach could be conducted by simply augmenting or stimulating a naturally occurring process within the original environment. To this end, in-situ biogasification of coal to methane eliminates the needs for coal mining, avoids the dangers to miners and environmental degradation that is associated with coal mining and related activities. Although this approach is still at the fundamental research stage, different coal mines have been selected to study microbial CBM production around the world, such as the Powder River Basin in the US (Green

* Corresponding author at: Department of Environmental and Sustainable Engineering, 1400 Washington Ave, State University of New York at Albany, Albany, NY 12222, USA.
E-mail address: yliang3@albany.edu (Y. Liang).

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et al., 2008), the Southern Qinshui Basin in China (Guo et al., 2014), the Alberta Basin in Canada (Penner et al., 2010), and the Surat and Sydney Basins in Australia (Faiz and Hendry, 2006). These studies showed that CBM might be of more recent biogenic origin, and the production could be improved through enhancing biological activities. For commercial applications, rising natural gas prices around the year 2000 led to the formation of several companies, including Luca Technologies, Inc., Ciris Energy, and Next Fuel, Inc. The primary objective of these companies was to stimulate CBM production by adding nutrients to stimulate activities and metabolisms of indigenous microorganisms (Ritter et al., 2015).

As mentioned above, very limited amounts of CBM have been produced in the IL basin (Eble et al., 2005). This is mainly due to low and uneconomic gas content. To improve CBM quality and quantity, we have investigated the approach of biostimulation specially for bituminous coal from this basin. Bioaugmentation is not necessary since the indigenous microbes were found to be capable of converting coal to methane (Pandey et al., 2016; Zhang et al., 2015a, 2015b). To make biostimulation work, specific nutrient solution is generally needed to supplement what is lacking in a specific environment for the purpose of stimulating microbial activities and thus increasing methane release. For the IL basin, although we have successfully developed suitable recipes for coal biogasification ex situ (Zhang and Liang, 2017; Zhang et al., 2016a, 2016b), a nutrient solution dedicated for in situ use has not been attempted before. Thus, in this study, we aimed to optimize critical factors involved in coal biogasification, develop mathematical models for predicting methane production from coal in situ, and understand this process better by evaluating individual parameters. In particular, although numerous studies have been performed on recipe development for coal bioconversion (Bumpus et al., 1998; Formolo et al., 2008; Gao et al., 2013; Jones et al., 2010; Zhang et al., 2015a), all of the developed recipes are tap water- or distilled water-based solutions. Considering the fact that coal seams and the indigenous microbes are often situated in saline environment, whether the freshwater-based nutrient solutions are useful in situ is a pending and interesting question.

2. Materials and methods

2.1. Coal, sand, and graphite samples

For this study, the coal sample used was the same as what has been investigated and reported for ex-situ application before (Zhang et al., 2016a). Briefly, coal blocks were collected from the Herrin Seam, # 6 in the Illinois basin. This coal contained 70.1% of carbon, 1.4% of nitrogen, 5.2% of hydrogen, 0.6% of sulfur, 15.4% of oxygen, and 7.5% ash (dry weight basis). Contents of volatile matter and fixed carbon were 49.9% and 42.6% (dry weight basis), respectively. Immediately before use, a block of coal was broken into lumps approximately 1.3 cm in size. The coal lumps were subsequently ground and sieved to obtain coal samples < 200 mesh (74 μm). This particle size was chosen based upon our previous observation that among different particle sizes, biogasification of coal < 74 μm led to the highest methane yield for this Illinois coal (Zhang et al., 2016a). Ground coal samples were stored in re-sealable zip-loc bags at room temperature in order to prevent moisture loss and oxidation.

Sand (CAT#S23-3, Fisher Scientific, USA) and graphite (CAT# G67500, Fisher Scientific, USA) were also used in this study. These two solid materials served as controls to those with coal. The sand was composed almost entirely of naturally rounded grains of nearly pure quartz. For the graphite powder, the content of graphite was higher than 99%. The particle sizes for both control samples were also < 200 mesh.

2.2. Formation water collection

Formation water used in this study was collected from an established coal-bed methane (CBM) well as described in our reported study (Zhang et al., 2015a). At the sampling site, the formation water was retrieved from a depth of around 850 ft. The in situ temperature was measured immediately after the formation water came to the surface. Fresh formation water was handled differently depending on final use. For those dedicated to the development of suitable nutrient solutions and experimental setup, the water samples in half-gallon containers were supplemented with sodium sulfide (Na_2S) at 0.25 g/L and resazurin at 1 mg/L to maintain anaerobic conditions. Once sealed tightly, these containers were brought back to our laboratory where they were immediately stored in a − 20 °C freezer for later use. No external compounds were added to water samples used for chemical compositions analysis. The only exception was those for analyzing content of total organic carbon (TOC). For this purpose, water samples were added to glass vials containing hydrochloric acid. All water samples for chemical analysis were kept on ice on the road to our laboratory. They were delivered to the Carbondale Central Laboratory (CCL, Carbondale, IL, USA) immediately once they reached the destination.

2.3. Formation water chemical characterization

Chemical analysis of the formation water was conducted for several groups of chemicals. First, for dissolved metals, such as Na, K, Ca, Mg, Fe, Al, Co, Mn, Zn, W, Cu, Ni, Se, B, and Mo, EPA method 220.8 through the use of Inductively Coupled Plasma – Mass Spectrometry (ICP – MS) was adopted. Second, for anions, such as Cl^-, SO_4^{2}, PO_4^{3-}, and NO_3^-, EPA method 300.0 through using of Ion Chromatography (IC) was employed. Third, HCO_3^- concentration was determined following standard method (SM) 320B. Fourth, TOC content was measured according to SM5310B. Fifth, regarding nitrogen species, the ammonia-nitrogen concentration was determined by using an ion selective ammonia electrode following EPA method 350.3. Total nitrogen concentration was measured by using a Hach Kit TNT827 (Hach, Inc. USA). Sixth, the content of dissolved H_2S was measured according to EPA 376.2.

2.4. The microbial community

The microbial community used in this study was that initially present in the formation water described above. Upon arrival in our laboratory, the formation water was concentrated 80 times through high-speed centrifugation. The resulting slurry was used to make glycerol frozen stocks and for DNA extraction. All inoculum used in the experiments detailed below was from these frozen stocks. The resulting DNA was subject to next generation 16S rDNA sequencing according to procedures reported by our laboratory (Zhang et al., 2015a).

2.5. Development of a nutrient recipe

2.5.1. The Box-Behnken design

In order to determine a suitable nutrient solution for maximizing methane yield, a three-level factorial design (Box-Behnken design) through using Design of Expert (DOE, Stat-Ease, Inc. Minneapolis, MN) was adopted. In this study, four parameters: Fe-powder (< 10 μm), ethanol, methanol, and trace minerals, were evaluated (Table 1). A total of 29 reactors was established according to this design. The upper and lower limits were 100 mM and 0 mM for Fe-powder, ethanol, and methanol; 100% and 0% supplement for trace minerals. Regarding the latter, a 100% trace metal supplement meant that the difference of trace mineral contents between the formation water used and a MS medium (Bonin and Boone, 2006) was provided. Specifically, a mixture of 1310.3 μg/L $CoCl_2.6H_2O$, 763.39 μg/L $ZnCl_2$, 264.84 μg/L $Na_2Wo_4.2H_2O$ and 100.00 μg/L H_2BO_3 was added to microcosms with 100% trace minerals. Glass serum bottles (120-mL) were used to establish the microcosms. Each microcosm contained 45 mL formation water, 2 g/L of yeast extract and trypticase peptone, 200 g/L of ground