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Optimization of methane production from bituminous coal through biogasification

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HIGHLIGHTS

- Among 12, coal loading, temperature, particle size and ethanol were statistically significant on methane production.
- The optimal conditions for producing methane from bituminous coal were determined.
- Under optimal conditions with a fed-batch scheme, 2900 ft^3 methane/ton was observed in 55 days.

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ABSTRACT

To optimize methane production from bituminous coal through use of a well-studied microbial community derived from the same Illinois basin in USA, a total of 12 parameters were first evaluated by setting up 64 reactors following a 2-level factorial design. Among the 12 parameters, temperature, coal loading, particle size and ethanol were found to have statistically significant effects on methane content and yield from coal. Following screening, to identify optimal value for each significant factor, a Box-Behnken design necessitating 29 reactors was adopted. Optimal conditions provided by the Design of Expert software for the highest methane yield were: temperature, 32 °C; coal loading, 201.98 g/L; coal particle size, <73.99 μm; and ethanol at 300 mM. Under these optimum conditions, the predicted methane yield and content was 2957.4 ft^3/ton (83.7 mm^3/ton) and 74.2%, respectively. To confirm the predicted results, a verification experiment was conducted, where a methane yield of 2900 ft^3/ton (82.1 mm^3/ton) with a methane content of 70% was observed. Thus, models developed from this study can be used to predict methane content and yield from bituminous coal through biogasification ex situ.

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1. Introduction

The global coal reserve is estimated to be 1,000 Gt [1]. As an abundant and inexpensive resource, coal has been investigated extensively for generating fuels and chemicals through various conversion technologies besides the conventional combustion for power generation. Conversion techniques that attempt to circumvent the negative environmental impacts associated with coal combustion, such as carbon fuel cell [2], coal to synthetic natural gas (SNG) [3], pyrolysis [4] and underground coal gasification [5] typically employ thermal and/or chemical processes under high pressures and/or temperatures with high capital and operating costs [6,7]. To alleviate these problems, coal bioconversion or biogasification has been studied intensively during recent years.

Biogasification transforms solid coal to methane gas through actions of microorganisms. Different from in situ gasification where syngas is produced from controlled combustion of coal [8] or ex situ gasification which is generally performed at temperatures higher than 800 °C [9], biogasification can be conducted under mild environmental conditions. In addition, coal does not need to be cleaned before biogasification like those prepared for power generation [10]. This technology can be used for both in situ (abandoned or unmineable coal seams) and ex situ (coal wastes or mined out coal) scenarios [11]. Considering the U.S. coal resource of 6 trillion tons [12], if a methane content of 200 ft^3/ton could be achieved, then the total methane from coal would be 1,200 trillion cubic feet (Tcf). This volume of methane is much higher than 158.2 Tcf, estimated by the potential gas committee as of year-end 2012 for coalbed gas resources and would be

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53.9% of 2226 Tcf of gas potentially recoverable from traditional reservoirs, such as conventional, tight sands and carbonates and shales [13]. Hence, if methane production from coal through biogasification is successful, it would generate the same volume of methane as that produced from shale gas in 2012, 1,073 Tcf. Considering the fact that coal seams are located at much shallower depth than shales, recovery of methane from charged coalbeds would be relatively inexpensive compared to that from shale.

To harness this natural process and make coal biogasification a commercial reality and a clean coal technology [14,15], huge amounts of efforts have been dedicated to microbially enhanced coal bed methane (MECBM). Specifically, these efforts have spanned from understanding the coal conversion pathways [16,17]; improving methane production rate by investigating different microbial communities [18,19], different nutrient solutions [11], different testing conditions; and conducting pilot scale tests by several companies [20]. Under different testing conditions, evaluating effects from different parameters have been investigated intensively. Factors, such as coal loading, medium pH, coal particle size and temperature [21–23], surfactants [24], solvents [22,25] and salinity [23] have been optimized for different coal samples.

Although excellent studies have been conducted on elucidating the key factors for increasing methane production rate, all researches so far have only evaluated the effect from variation of single parameters. For example, effect of temperature was detected in cultures having the same pH or effect of pH was studied for cultures at one temperature. Combined effects from multiple parameters, though critical, have not been investigated to the best of our knowledge. In addition, no such studies have been carried out for bituminous coal. Thus, for this study, we conducted experiments designed by the use of Design of Expert (DOE) software to: (1) evaluate effects from single and multiple factors simultaneously; and (2) identify the optimal conditions for achieving maximal methane yield. To achieve this purpose, we started with a 2-level factorial design to screen the most important parameters affecting methane productivity. Once significant parameters were identified, we used response surface methodology (RSM) to identify the optimal conditions for obtaining the highest methane yield.

A total of 12 parameters were evaluated in this study. These 12 factors were chosen based on their reported effects on methane production from coal. It needs to be noted that these parameters are strongly tied to ex situ coal bioconversion although valuable information can be applied to in situ scenarios. The selected parameters were: (1) particle size (<420 μm, mesh size 40); (2) pH (6.0–8.0). As demonstrated by our previous study [26], in our enriched microbial consortium, the order of Methanomicrobiales was 90.4% of the methanogenic population. For this order, the optimal pH ranges from 6.0 to 8.0 [27]; (3) temperature (20–40 °C). With two exceptions that can tolerate temperatures up to 60 °C, the majority of the Methanomicrobiales are mesophiles and have optimal temperatures from 20 to 40 °C; (4) mixing (0–75 rpm). As almost all studies on coal biogasification are conducted under static conditions, effect from mixing is unknown. On one hand, mixing can enhance the contact and interaction between coal and microorganisms. On the other hand, mixing may damage the attachment of cells to coal; (5) inoculum size (10–20% of final liquid volume in each reactor). An inoculum size of 10% has been commonly used. But it is unknown whether increased initial cell numbers will enhance methane release from coal; (6) coal loading (200–700 g/L). Different studies have reported different coal loadings ranging from 25 to 800 g/L [28]. Low coal loading requires large consumption of nutrient solutions while high loading may render some coal un-accessed by medium and microbes; (7) mercaptoethanesulfonic acid (coenzyme M, CoM, 0–0.25 g/L). This compound is generally included in nutrient media for anaerobic cultures. It is a reducing agent and also required by an enzyme: methyl-coenzyme M reductase in the final step of methane formation from various substrates in anaerobic environments [11,29]. This chemical accounts for approximately 75% of the total medium cost when used at 0.5 g/L; (8) two surfactants (30–50% of critical micelle concentration (CMC)). Triton X-100 is nonionic and was reported to exert no effect on enhancing methane production from subbituminous coal while its effect on bituminous coals are unclear [24]. Sodium dodecyl sulfate (SDS), is anionic and has not been evaluated in terms of impact on methane production. These two surfactants were chosen due to their low toxicity and low cost. Cationic surfactants were not selected since their solutions can interact with coal and result in decreased pH [30]; and (9) three carbon sources (each at 100 mM). Members of the order of Methanomicrobiales grow by reducing CO_2 with H_2 and some strains can use formate and alcohols as electron donors. Since H_2 is a cleaner fuel than methane, it does not make sense to add large amount of H_2 (80% of headspace gas) for the purpose of producing methane, especially considering large scale applications. Thus, in the reported study here, we did not attempt to supply H_2 in the headspace. To reduce CO_2, we investigated effects from sodium formate, 2-propanol and ethanol. It needs to be noted that: (a) like H_2, the two alcohols are also biofuel molecules. But their concentrations in this study were only 100 mM; (b) the two alcohols either being miscible with or having high solubility in water might also serve as solvents to increase coal solubility.

2. Materials and methods

2.1. Coal samples

For the current study, the coal samples used were the same as what were studied and reported before [26]. 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% of ash (dry weight basis). Contents of volatile matter and fixed carbon were 49.9% and 42.6% (dry weight basis), respectively. The high content of volatile matter and a heating value of 12,548 BTU/lb put this coal in the category of high volatile B Bituminous. 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 at different particle sizes as discussed below. Ground coal samples were stored in re-sealable plastic bags at room temperature in order to prevent moisture loss.

2.2. A microbial community and nutrient solution

The microbial community used in this study was developed from microorganisms initially present in the formation water collected from an on-going coal-bed methane operation in southern Illinois, USA. Through cultivating on ground bituminous coal detailed above in a MS medium [31] and after four transfers, the final enriched community comprised a total of 185 bacterial species and nine species of archaea. The abundant bacterial species were: *Clostridium bifermentans* (15.1%), *Massilia spp.* (11.1%), *Pseudomonas putida* (11.1%), *Proteiniphilum spp.* (6.5%), and *Pseudomonas stutzeri* (6.4%). The majority of archaea belonged to the *Methanocalculus* genus and the *Methanomicrobiales* order [26]. From this enriched consortium, frozen stocks were made and stored at −80 °C for later use. All inocula used in this study were developed from the same frozen stocks.

The MS medium contained (per L of distilled and deionized water (DDW)) 0.1 mol of $NaHCO_3$, 2.0 g of yeast extract, 2.0 g of trypticase peptones, 0.5 g of mercaptoethanesulfonic acid (Coenzyme M, CoM), 0.25 g of $Na_2S{\cdot}9H_2O$, 1.0 g of NH_4Cl, 0.4 g of K_2-