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Microbial enhancing coal-bed methane generation potential, constraints and mechanism – A mini-review

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ABSTRACT

Microbial enhancing coal-bed methane generation (MECMG) is a feasible and environment-friendly approach for improving coal-bed methane production. In order to understand the potential of biogenic methane generation and factors controlling the process, the biogenic methane yield, coal intrinsic characteristics and incubation conditions are assembled from the published data. Results show that sub-bituminous and high volatile C-B bituminous coal have higher biogenic methane generation yield than that in peat and anthracite. Contents of H, N, and volatile matter in the coal contribute positively to the biogenic methane generation yield. The optimized conditions for *ex-situ* biogenic methane generation are headspace gas type, culture temperature, particle size, solid-liquid ratio and pH of N_2:CO_2 (4:1 v/v), 28–30 °C, 60 μm, 1:10, and 7.0–7.5, respectively. The organic composition and molecular structure collected from coal-bed produced water, culture solution, and biodegraded residuals would provide effective evidences for revealing biogenic methane generation mechanisms.

1. Introduction

Biogenic gas is a renewable and environment-friendly energy resource, accounting for more than 20% of the total methane reserves on the earth (Rice and Claypool, 1981; Rice, 1992). The generation process is still active and on-going in some sedimentary basins, such as San Juan Basin (Scott et al., 1994), Powder River Basin (Jin et al., 2007), southern Sydney Basin (Faiz et al., 2007), southeastern Illinois Basin (Strąpoć et al., 2007), Surat Basin (Li et al., 2008), and Huainan-Huaibei coal fields (Tao et al., 2007; Bao et al., 2014). The fact that coal can be metabolized by microbes has been realized since the early 20th century (Potter, 1908), but the microbes endemic to coal were reported several decades later (Rogoff, 1962). Coal biodegradation by aerobic bacteria and fungi in the laboratory (Cohen and Gabriele, 1982) and coal-bed methane generation by anaerobic microbial process have not been recognized until 1980s (Rightmire, 1984).

Recently, coal biodegradation by microbes to form methane became a hot topic because the process not only improves the yield of coal-bed methane but also reduces the environmental hazard of coal mining. Numerous results have been published; however, the relationship between coal rank and biogenic methane generation potential is still a controversial issue. For example, Rice and Claypool (1981) and Orem and Finkelman (2003) reported that there was more biogenic methane generated from low-rank coal than the high-rank one. However, Fallgren et al. (2013) insisted that bituminous coals could generate more biogenic methane than lignite and sub-bituminous coals. In addition to coal rank, other factors controlling the potential of secondary biogenic methane generation are largely unknown.

In this work, both internal factors such as coal rank, element composition, proximate composition, and petrographic composition and external factors such as temperature, particle size, solid-liquid ratio, pH, and salinity are investigated to understand the controls on the potential of biogenic methane generation. Large data sets including coal rank, particle size, solid to liquid ratio,

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cultural temperature, incubation time, pH, pressure and headspace gas types (83 data, 26 references) and element content, proximate composition, and petrographic composition of coal samples (52 data, 11 references) are gathered and analyzed. The constraints and optimized conditions for *ex-situ* methane generation are inferred. Meanwhile, the experimental procedures and analytical approaches are summarized as well. The problems encountered in the current research and future challenges are subsequently discussed.

2. Materials and methods

Experimental methods, operational processes and other important information related to *ex-situ* biogenic methane generation simulation are collected from available literature and summarized briefly here.

2.1. Sample preparation

Most coal samples were collected from the underground and preserved in tin-foil under low temperature in the laboratory for long-time storage (Papendick et al., 2011; Strąpoć et al., 2011; Wawrik et al., 2012). The storage and preparation places should be purged in case of volatile substances are present (Robbins et al., 2016). Samples were generally grinded or lumped (rarely used in laboratory) for the utilization in subsequent inoculation experiments (Green et al., 2008; Jones et al., 2010; Orem et al., 2010; Papendick et al., 2011). Because coal samples were crushed and exposed a period of time before inoculation, no previous gas remnants exist in coal matrix and observed methane yield represents biogenic methane generation potential from coal.

In order to better understand the basic coal properties and compare the factors influencing the hydrocarbon generation, coal rank (vitrinite reflectance, R_o), elemental composition (C, H, O, N and S contents), proximate composition (moisture, ash and volatile matter contents) and petrographic composition (vitrinite/huminite, inertinite and liptinite contents) of coal samples should be measured synchronously.

2.2. Culture medium selection

The formula of methanogens culture medium has been widely reported in the literature (Balch and Wolfe, 1976; Zehnder and Wuhermann, 1977; McInerney et al., 1979; Fedorak and Hrudey, 1984; Müller et al., 1986; Qian and Min, 1986; Tanner, 2002; Bonin and Boone, 2006; Jones et al., 2006, 2008, 2010; Green et al., 2008; McIntosh et al., 2008; Strąpoć et al., 2008; Orem et al., 2010; Opara et al., 2012; Khelaifia et al., 2013; Lavania et al., 2014; Zhang et al., 2016). Even though there are some differences in chemical compositions, the common components in medium contain the following six ingredients: major minerals (K, Na, Ca, Mg, N, P, and Cl), organic nitrogen source (yeast extract, peptone, or tryptone), vitamin solution (biotin, calcium pantothenate, folic acid, mercaptoethanesulfonic acid (coenzyme M), nicotinic acid, *p*-aminobenzoic acid, pyridoxine HCl, riboflavin, thiamine HCl VB_1, thioctic acid, and vitamin B_{12}), trace mineral solution (Mn, Co, Fe, Zn, W, Cu, Se, Mo, and B), reducing agent (Na_2S, cysteine-HCl, or mercaptoethanesulfonic acid), and redox indicator (resazurin) (Park and Liang, 2016). The maximum methane yield was obtained by Tanner (2002) and Green et al. (2008) in a standard medium (Table 1).

In order to keep medium free from contamination, the basal medium (Tanner, 2002) should be prepared at the beginning of experiment, while the trace mineral solution and vitamin solution (Green et al., 2008) are supposed to be prepared once for multiple usages. The mineral and vitamin solutions must be sealed and preserved under low temperature.

2.3. Methanogens enrichment and incubation

After medium preparation, methanogen enrichment should be done next. Methanogens can be introduced from the coal-bed-associated water in the investigated CBM fields or from previous enrichment culture solution. The former is indigenous microbial consortium; while the latter can be exogenous consortium. To obtain enriched methanogens, low molecular weight carbon substrates, such as formate, acetate, propionate, bicarbonate, 1-propanol, 1-butanol, or ethanol, could be added into medium separately to enhance methanogen metabolism and rapid growth until the carbon substrate has been exhausted (Widdel, 1986; Wawrik et al., 2012; about 30 days or several months), and then transferred into several small serum bottles for incubation (with coal added). At last, the coal samples and enriched methanogens solution were added into the anaerobic bottles which were sealed with butyl rubber stoppers and aluminum crimps. All the apparatus and amendments should be sterilized prior to use.

The mediums are autoclaved and flushed with nitrogen 3 times to remove oxygen. The ratios of coal (unit g), medium (ml) and enrichment solution (ml) are generally 1:10:1. The headspace remained ~1/2 vol, and is flushed with nitrogen for 20 min, in order to remove oxygen from the bottles, at 1.01×10^5 Pa (1 atm) pressure. pH value of medium is set at 6.5–7.5 (Green et al., 2008; Gupta and Gupta, 2014). The temperature of anaerobic box is 22–37 °C (Opara et al., 2012; Wei et al., 2014; Susilawati et al., 2015). The coal-contained bioreactor treatments are incubated without shaking in the dark for 20–35 days (Opara et al., 2012; Haider et al., 2013; Susilawati et al., 2015), 50–90 days (Harris et al., 2008; Fallgren et al., 2013; Robbins et al., 2016), 105 days (Orem et al., 2010), 120–145 days (Jones et al., 2008), or 1 year (Harris et al., 2008) for biogenic methane generation. Meanwhile, each incubation treatment should be prepared in triplicates with one blank (without coal inoculums).

2.4. Gas analysis

Headspace gas composition for each microcosm is determined at regular intervals (every 3–5 days, Orem et al., 2010; Wei et al., 2014) using gas chromatography on 0.3 ml of bioreactor headspace that was sampled with a gas-tight syringe (Jones et al., 2008). The septum of each microcosm is sealed with silicone immediately after sampling at the location of the needle puncture to prevent gas leakage (Fallgren et al., 2013).

3. Biogenic methane generation potential and influential factors

3.1. Coal rank and biogenic methane generation potential

The correlation between coal rank and biogenic methane generation potential has been discussed in several studies. The exploration and development practice in CBM fields have revealed that biogenic CBM can be generated from various coals with different coal ranks (Faiz and Hendry, 2006). However, the correlation between coal rank and biogenic methane yield is still ambiguous. Most studies suggested that low-rank coals were able to form more microbial gas than high-rank counterparts (Rice and Claypool, 1981; Orem and Finkelman, 2003; Opara et al., 2012). There is a negative correlation between coal rank and methane yield in 13 (Robbins et al., 2016) or more samples when vitrinite reflectance is less than 1.0% (Strąpoć et al., 2011). However, other studies noted that bituminous coals can generate more methane than lignite and sub-bituminous coals in 4 samples (Fallgren et al., 2013).

In this paper, 83 datasets regarding methane generation