

A Processing Method to Increase Production of Biomethane from Lignite

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Abstract. This study evaluated the bioavailability of lignite from Shanxi Province (China) via using domestic anaerobic sludge as the source of exogenous flora. After pretreatment of lignite with hydrogen peroxide at different concentrations, the levels of soluble organic carbon and organic acid were increased significantly. hydrogen peroxide pretreatment can improve the solubility and bioavailability of lignite and influence methane production, making them viable processing options for increasing microbial production of coalbed methane.

Introduction

As a resource with abundant reserves, coal occupies more than 70% of global fossil fuels [1]. Currently, over 90% of lignite is used for conventional combustion and power generation among all of coals. However, the greenhouse gases and other harmful substances released during combustion process can cause serious environmental pollution [2]. In recent years, the process of lignite biogasification has been developed as the emerging and alternative cleaner energy source to produce methane [3]. Compared with traditional combustion methods, biogenic coalbed methane has many advantages, such as low energy consumption, moderate methane production conditions, high methane productivity efficiency, and simple equipment requirements etc. [4].

The heterogeneity and complexity of coal limit its direct use by microorganisms; therefore, without improvement of the physical, chemical, and biological pretreatment methods to enhance the solubility and bioavailability of coal, it will remain difficult to increase biomethane output in a coal seam [5]. There are two primary advantages in using hydrogen peroxide to pretreat lignite. First, the dissolution of lignite by hydrogen peroxide can produce organic acids (e.g., acetic acid) that are easily used by methanogens, which can promote the bioavailability of lignite [6]. Second, the decomposition of hydrogen peroxide produces water and oxygen without introducing harmful elements into the coal seam.

The objectives of this study were as follows: (1) to pretreat lignite with different concentrations of hydrogen peroxide; (2) to determine the total organic carbon and organic acid contents in the supernatant after reaction, and evaluate lignite solubility and methane production after the pretreatment.

Materials and methods

Sample. The coal samples were obtained from the Qiaojiawan Coal Mine.

Pretreatment of coal samples. At room temperature, 2 g lignite was added to 100 mL hydrogen peroxide solution at different concentrations (0.5, 1.5, and 2.5 M) with triplicates, and the

control experiment was conducted in a same mass of 2 g lignite added but with 100 mL water. After reaction for 10 days, the supernatant was filtered using a 0.45- μm organic system filter membrane prior to the chemical analyses.

Setup for experiments. According to the method of Huang et al. [7], a basic culture solution, a trace element culture solution, and a vitamin solution were configured. The pH of the medium was adjusted to 7.0 ± 0.2 using sodium hydroxide. Anaerobic sludge from domestic sewage treatment plants (sourced from Bengbu Jingsheng Environmental Protection Technology Co. Ltd., Anhui, China) was used as the source of exogenous flora. After domestication for several times, the supernatant was used as the inoculum. To investigate the effects of bioavailability and solubility on methane production, the following experiments were designed. In all assays, 5 mL of the inoculum was added and the samples were cultured in an anaerobic incubator at 35°C . This study referred to the method proposed by Huang et al. [8], i.e., 2 g lignite with different concentrations of hydrogen peroxide treated was added to 25 mL of methanogenic nutrient solution. The concentration of the methanogenic nutrient solution used in this part was twice of the original methanogenic nutrient solution in order to keep the concentration of methanogenic nutrient solution used in all experiments consistent. Seen Table 1 for other conditions.

Table 1. Description of processing methods and conditions

Processing methods	Description
Treatment 1	Add 25 mL of pretreated supernatant of coal and water mixture
Treatment 2	Add 25 mL of supernatant pretreated with 0.5 M hydrogen peroxide
Treatment 3	Add 25 mL of supernatant pretreated with 1.5 M hydrogen peroxide
Treatment 4	Add 25 mL of supernatant pretreated with 2.5 M hydrogen peroxide

Chemical analyses. A Dongxi 4100A gas chromatograph equipped with a thermal conductivity detector was used to measure the methane concentration in the bioassay. Nonpurgeable organic carbon concentration in the solution was measured with a the total organic carbon analyzer (Vario the total organic carbon, Elementar) as the dissolved organic carbon concentration. Monitoring of volatile fatty acids (formic and acetic acids) was conducted via the high-performance liquid chromatograph equipped with a C18 reversed-phase column (10 μm , 4.6 mm i.d. \times 250 mm). A phosphate buffer (10 mM) was used as a mobile phase with a flow rate of 1 mL min^{-1} .

Results and discussion

As an oxidant, hydrogen peroxide can cause pretreated lignite to produce more microorganisms to facilitate the use of hydrophilic functional groups (e.g., ketone, hydroxyl, and carboxyl) [9]. The concentrations of the dissolved organic carbon in lignite pretreated with different concentrations of hydrogen peroxide (Treatments 2-4) and Treatment 1 are shown in Fig. 1. With the increase of hydrogen peroxide concentration, the concentration of the total organic carbon increased continuously. The concentration of the dissolved organic carbon after pretreatment with 2.5 M hydrogen peroxide was $628.3 \text{ mg C L}^{-1}$, which was 99% and 39% higher than that of the dissolved organic carbon pretreated with 0.5 and 1.5 M hydrogen peroxide, respectively. Compared with Treatment 4 (1.6 mg C L^{-1}), the the dissolved organic carbon concentration in the supernatant after treatment with hydrogen peroxide was increased significantly. In addition, more organic acids were dissolved from the lignite pretreated with hydrogen peroxide, and the organic acid concentration increased with the increase of hydrogen peroxide concentration. The concentrations of formic and acetic acids after pretreatment with 2.5 M hydrogen peroxide were 138.2 and $101.6 \text{ mg C L}^{-1}$, respectively. No organic acid was detected in Treatment 1. This indicated that the dissolved organic carbon and organic acids cannot be extracted from lignite using water alone. The dissolved the dissolved organic carbon and organic acids were derived via the reaction of hydrogen peroxide and lignite, which revealed that the hydrogen peroxide pretreatment method can increase the solubility

of coal and produce substrates (formic and acetic acids) easily used by methanogens to increase the bioavailability, resulted in achieving the increase of final methane production ultimately.

Methane production continued to increase with increasing concentration of hydrogen peroxide (Fig. 1). The final yield of methane from lignite pretreated by 2.5 M hydrogen peroxide was $193.1 \mu\text{mol g}^{-1}$, which was 30% and 22% higher than that of lignite pretreated with 0.5 and 1.5 M hydrogen peroxide. Treatment 1 produced the lowest amount of methane ($126.5 \mu\text{mol g}^{-1}$), which might be attributable to the fact that less organic matter could be dissolved from the lignite. As shown in Fig. 1, the final methane yield was related to the concentration of organic matter dissolved from the aqueous solution. As the concentration of dissolved organic matter increased, the final methane yield also improved.

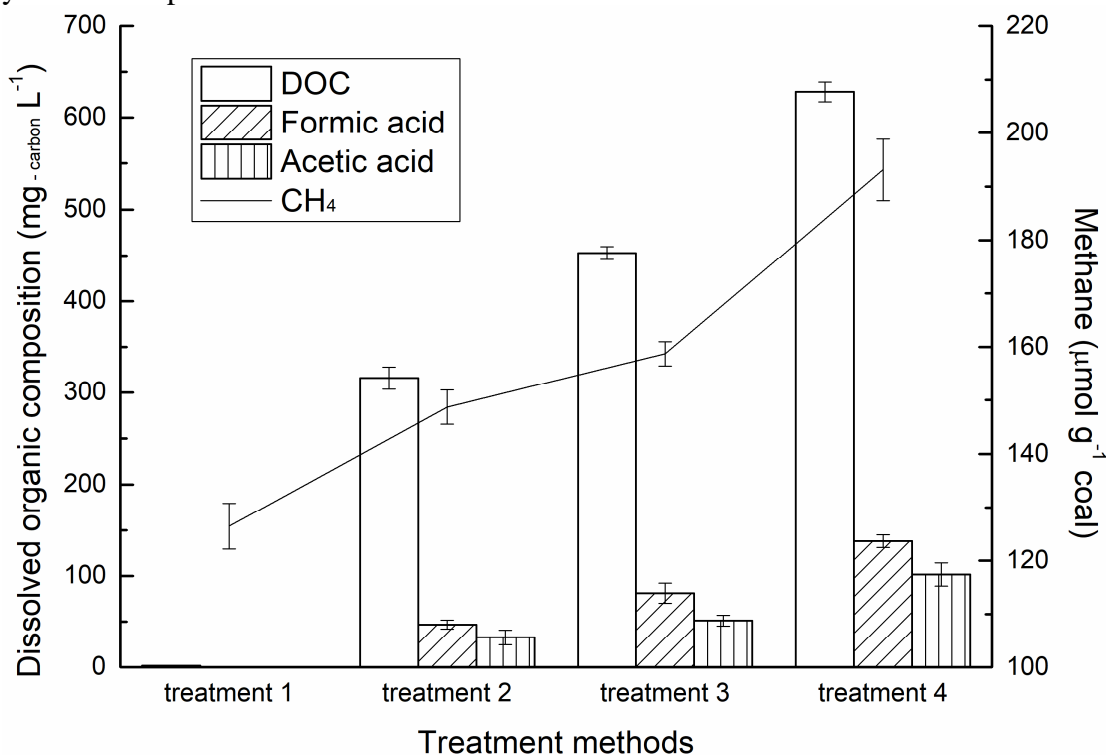


Fig. 1 The dissolved organic carbon, formic acid, and acetic acid concentrations in the supernatants with different pretreatments, and the final methane yield after 60 days of culture. Error bars represent the standard deviations of triplicate cultures.

Conclusions

This study demonstrated that utilizing hydrogen peroxide to pretreat lignite dramatically increased the concentration of the dissolved organic carbon, which increased the organic acids in the aqueous solution. Under the conditions of this experiment, when 2.5 M hydrogen peroxide was used as the pretreatment method, methane production increased by 53% compared with untreated lignite. These results illustrated the solubility of coal in water can greatly affect bioavailability.

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