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Experimental investigation on the feasibility of industrial methane production in the subsurface environment via microbial activities in northern Hokkaido, Japan – A process involving subsurface cultivation and gasification

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ABSTRACT

Future world energy consumption is projected to increase along with the high economic growth of emerging countries. This will make it necessary to expand the development of unconventional resources such as shale oil and gas and coalbed methane. The production of secondary methane gas using microorganisms at oil fields and coal mines is another area of great interest in the development of unconventional resources. This paper describes a feasibility study involving a new method to convert unused organic matter contained in sedimentary rock in northern Hokkaido into biogenic methane. The new method, subsurface cultivation and gasification (SCG), is proposed for the production of biogenic methane gas in the subsurface environment. Our approach uses hydrogen peroxide (H_2O_2) to decompose organic matter rapidly into usable substrates for methanogens. Two wells were drilled through several lignite seams in the Soya coal-bearing formations in the Tempoku coalfield in northern Hokkaido, Japan, and lignite cores were recovered. Batch tests using a 1 wt% H_2O_2 solution were performed on the lignite to estimate the amount of low-molecular-weight organic acids as substrates for methanogens. Oxidative decomposition of the lignite produced a high yield of low-molecular-weight organic acids. Moreover, it was confirmed that methanogens converted the H_2O_2 reaction solution containing organic acids into methane. H_2O_2 would be useful for effective SCG at lignite seams, and conversion of organic matter from lignite into biogenic methane with the help of microorganisms is expected to be highly profitable. The results reveal that using SCG to produce biogenic methane in this coalfield shows great commercial promise.

1. Introduction

Current development demands on petroleum and natural gas are expanding for both conventional resources and unconventional resources. However, unconventional resources in particular can be difficult to develop as future energy demand–supply conditions can be inflexible and difficult to predict. Typical unconventional resource development focuses on e.g., oil sand and shale oil as petroleum sources and shale gas, coalbed methane (CBM), and underground coal gasification to produce natural gas.

Microbial methanogenesis occurs in diverse subsurface environments, the processes of which play an important role in the global-scale carbon cycle and could assist in effectively utilizing biogenic methane as an energy resource. Strąpoć et al. [1] estimated that methane recovered from coal beds contributed to approximately 10% of the total natural gas production volume in the United States, with about half of this production being of biogenic methane. Subsurface methane is derived from microbial products, thermal decomposition, or chemical reactions in inorganic matter. Biogenic methane is produced via the metabolic processes of methanogens. With the advancement of the CBM recovery technique, the production of secondary biogenic methane [2] from strata is of great interest as a clean energy source. Some substrates for methanogens are contained in source rock masses or in groundwater in which the methanogens are active. Low-molecular-weight organic components, especially acetic acid, formic acid, and methanol, can be used as dominant carbon resources for methanogens [3]. Source rock

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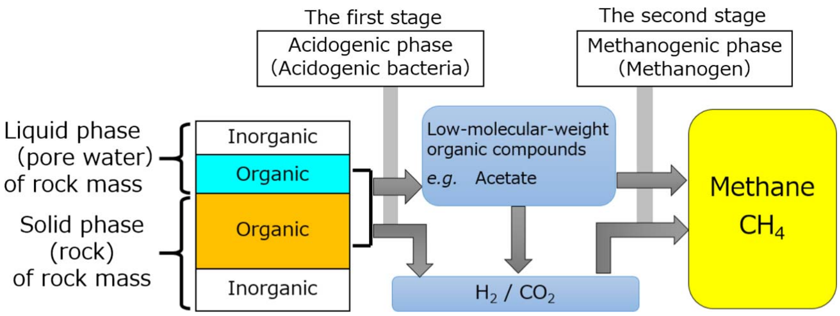

Fig. 1. Schematic of microbial methanogenesis in the subsurface environment.

masses such as coal beds comprise recalcitrant decomposable organic matter that can be converted into low-molecular-weight organic acids through chemical reactions (e.g., reactions between meteoric water and minerals in the rock [4,5]) or the metabolic activity of microorganisms [6] in the subsurface environment. Recent studies on the generation of biogenic methane in coal beds have focused on the bioavailability of coal carbon [7], the presence of microbial communities to convert coal carbon to methane [8], environments supporting microbial growth and methanogenesis [9], and process control for the generation of methane when a combination of the above factors is present [10,11].

Several sedimentary rocks of Paleogene and Neogene age form unconformable strata on a Cretaceous age basement rock in northern Hokkaido, Japan. Tamamura et al. [12] analyzed dissolved gas in groundwater sampled from several strata of less than 1000 m depth in the area, with the presence of bubbles in the sampled groundwater confirmed at the sampling. Methane concentration in a headspace of vials containing sampled groundwater was measured to be over 50% in most samples, revealing that the groundwater in the area was supersaturated with methane. In addition, the dissolved methane in the groundwater was determined to be of microbial origin from the relationship between the methane/(ethane + propane) ratio and the carbon isotope ratio of methane ($\delta^{13}C$ (CH_4)) [13]. Sedimentary rocks of Neogene age are included in several coal seams comprising the Soya coal-bearing formation in the Tempoku coalfield in northern Hokkaido. Methane in these coal seams was also confirmed to be of microbial origin [14], with the presence of methanogens including some new strains of a methanogenic archaeal community confirmed in the coal seam and diatomaceous shale formations of northern Hokkaido [15,16]. However, the amount of methane in the subsurface environment of northern Hokkaido that is available as a commercial energy resource has not been confirmed.

Microbially Enhanced Coalbed Methane, an enhanced recovery method utilizing microorganisms, is currently under development [10]. From the perspective of ensuring stable production, recovery, and supply of biogenic methane as an energy resource, there are concerns that the production rate of suitable substrates for methanogens from persistent geomacromolecules might be a bottleneck during methanogenesis in subsurface environments. As one method of solving this bottleneck [17], our approach uses hydrogen peroxide (H_2O_2) in the chemical decomposition of organic matter in sedimentary rocks into usable substrates for methanogens [18,19]. Because H_2O_2 is a representative regent in synthetic organic chemistry, many studies have been conducted on the oxidation reactions of organic matter with H_2O_2 [20]. Miura et al. [21] showed that oxidation of lignite using a 30 wt% H_2O_2 solution at low temperature under ambient pressure was a promising method for producing small-molecule fatty acids such as acetic acid and formic acid in high yields and with high selectivity. Using batch experiments, Tamamura et al. [19] examined the potential of dilute H_2O_2 (0.3 wt%) to solubilize lignite at common subsurface temperatures (10–50 °C). Dilute H_2O_2 was also found to be useful for producing organic acids as a substrate for methanogen. They further conjectured that the aromatic-C content in lignite markedly decreases during reaction with H_2O_2, implying high reactivity of the catechol structures of lignite with $H_2O_2 \cdot H_2O_2$ solubilizes lignite with the production of methanogen substrates to produce organic acids, resulting in significant enhancement of the bioavailability of lignite.

Our laboratory has studied biogenic methane production in the subsurface environment from the viewpoints of microbiology, geochemistry, and geomechanics. The ultimate goal of these efforts is to develop a new biogenic methane production method in the subsurface environment through an organic linkage of these viewpoints. This paper presents a feasibility study of a new method to convert unused organic matter contained in the coal seams of northern Hokkaido into biogenic methane. The feasibility study comprises two major tasks: (a) a coal chemical study involving geotechnical engineering and (b) a microbial engineering study. The new biogenic methane production methodology, which is called “subsurface cultivation and gasification” (SCG), is first proposed in the following section. Subsequently, the two major tasks above are examined experimentally. The feasibility of SCG is then discussed based on the results of each task.

2. Methodology of SCG and approach

2.1. Subsurface cultivation and gasification

Fig. 1 shows schematically the process of microbial methanogenesis under an anaerobic condition in the subsurface environment [22]. To provide a commercial energy resource, a sufficient amount of methane must be stored in the subsurface environment. Typically, this takes geological time in processes such as the formation of natural gas fields.

The stable production and recovery of biogenic methane in the subsurface environment, requires the development and application of a technical production process. To extract the methane reserves suggested by the preceding analysis of the Tempoku coalfield, we propose a new technical method for the production of biogenic methane in the subsurface environment.

Fig. 2 shows an image of the proposed method for the production of biogenic methane in the subsurface environment, which is called SCG. The SCG process is roughly classified into two stages: “reclamation” and “cultivation”. The reclamation stage involves developing a gas deposit in the subsurface environment for microbial methanogenesis. First, a gas well is drilled through a target stratum comprising recalcitrant decomposable organic matter (Fig. 2(a)) and formation of methanogenic substrates is then assisted through a geological improvement method applied to rapidly produce low-molecular-weight organic components as substrates of methanogens from the source rock (Fig. 2(b)). The geological improvement method might involve injection of useful microorganisms [6] or a reagent into the stratum to accelerate the decomposition of organic matter in the source rock. This is the same approach as the in situ contaminated soil remediation method to decompose harmful organic substances in soil/groundwater. In the cultivation stage, biogenic methane is generated from various substrates (e.g., acetic acid, formic acid, hydrogen, carbon dioxide) via methanogen metabolism. Methanogens can utilize substrates produced technically in a previous stage. To increase the production of biogenic methane following geological improvement, a solution containing