

## Bioelectrochemical Conversion of geologically sequestered Carbon Dioxide into Methane by using Indigenous Microorganisms

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### [Introduction]

We are proposing a sustainable carbon cycle system which gives a solution not only to mitigate global warming but also to supply a carbon-neutral energy resource. Carbon dioxide Capture and Storage (CCS) technology, which is currently being developed around the world, could become a practical countermeasure to reduce emission of the greenhouse gas. As potential CO<sub>2</sub> geological storage site in CCS, utilization of depleted oil/gas reservoirs and aquifer has been proposed. The long-term aim of this research is to establish a biotechnological system to microbiologically convert geologically stored CO<sub>2</sub> into methane.

Our recent study revealed that methanogen (methane producing archaea) and exoelectrogen (electron emission bacteria) inhabiting subsurface reservoir are involved in the recently discovered bio-electrochemical reaction called electromethanogenesis ( $\text{CO}_2 + 8 \text{H}^+ + 8\text{e}^- \rightarrow \text{CH}_4 + 2\text{H}_2\text{O}$ ). In this reaction, methanogen receives a proton ( $\text{H}^+$ ) from reservoir brine and electron from a solid electrode, as a result, reduces CO<sub>2</sub> into methane. Required electricity for the methane conversion can be obtained from renewable energy sources such as wind or photovoltaic power generations.

In this report, we introduce the latest result based on the experiment that used the microbes collected from actual oil field about the possibility of utilization of electrochemical hydrogen reduction power as a method to supply hydrogen to methanogen.

### [Experiments]

Single-chambered electromethanogenic reactors were used for an evaluation.

The anode and cathode were made of a plain carbon cross of 4cm x10cm.

The reactors were inoculated with formation water anaerobically collected from Yabase oil field, located in Akita, Japan (oil formation depth: 1,293m - 1,436m, oil formation temperature: 40-82 degrees Celsius). Each reactor headspace was filled with mixed gases of N<sub>2</sub>/CO<sub>2</sub> (80/20). The reactors were incubated at 55 °C with an applied voltage of 0.75 V.

In addition, methane production rate and conversion efficiency of electricity-to methane were observed changing the applied voltage level with 0.4, 0.5, 0.6, 0.7, 0.8 V.

### [Results]

With an applied voltage of 0.75 V, the reactors produced methane at a rate of 386mmol/day m<sup>2</sup>. The current-methane conversion efficiency was almost 100%. On the other hand, no significant methane production was detected in the reactors without applied voltage. Furthermore, the methane production rate increased from 84 mmol/day m<sup>2</sup> to 1,103 mmol/day m<sup>2</sup>, with increasing applied voltage from 0.4 V to 0.8 V. The current-methane conversion efficiency surpasses 90% in all reactors with applied voltages.

To investigate the mechanism of electromethanogenic reaction, the phylogenetic diversity of the microbes on the cathode was analyzed by constructing 16S rRNA gene clone library. As for archae, the result shows methanogene closely related to Methanothermobacter thermoautotrophicus dominated the consortium. On the other hand, as for bacteria, Therminecola ferriacetica, one of the exoelectrogen, was the dominant species. Our research demonstrated for the first time that the possibility of bioelectrochemical methane conversion of carbon dioxide by utilizing microbes indigenous to oil field.

Keywords: Carbon dioxide Capture and Storage, electromethanogenesis, methanogen, exoelectrogen, carbon cycle