Surface Monitoring for CO2 Geological Storage with H2 Tracer

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論 文 名 : Surface Monitoring for CO<sub>2</sub> Geological Storage with H<sub>2</sub> Tracer
(CO<sub>2</sub>地中貯留に対する H<sub>2</sub>トレーサを用いた地表でのモニタリング)
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論文内容の要旨

The CO<sub>2</sub> capture and geological storage (CCS) would be an effective method for offsetting human-induced climate change provided that the captured CO<sub>2</sub> can be safely stored in underground formations for long-term, i.e. for hundreds or thousands of years. In order to make CCS widely accepted by policy makers and the general public, particularly the residents living in the vicinity of the storage sites, the storage will have to be safe and demonstrate no leakage of CO<sub>2</sub> gas from a deep reservoir. Those sites, therefore, need to be evaluated using appropriate CO<sub>2</sub> leakage monitoring procedure and a comprehensive monitoring system.

The greenhouse gas emission control will be more effective if it is supplemented by reducing the carbon-based energy consumption and using lower- or zero-carbon energy sources, such as hydrogen (H<sub>2</sub>). Currently, some industries are producing H<sub>2</sub> extensively, however H<sub>2</sub> production plants from hydro-carbon resources are also emitting CO<sub>2</sub> and CCS can be embedded into their system to reduce net carbon emissions. The CO<sub>2</sub> captured from H<sub>2</sub> plants contains slight quantities of H<sub>2</sub> that would be injected along with CO<sub>2</sub> into deep geological formations. In addition, natural H<sub>2</sub> may also be produced in the deep subsurface area around the geological storage. On the other hand many studies have considered a wide range of gases to be utilized as tracers for CCS monitoring, and H<sub>2</sub> if being injected into the reservoir can be a suitable gas for this purpose. In this study, a new monitoring approach has been proposed to be used for CO<sub>2</sub> leakage detection by utilizing H<sub>2</sub> gas as a tracer.

This dissertation consists of six chapters.

Chapter 1 describes the background, objectives and the importance of the research to provide the fundamental insights into the study.

Chapter 2 presents a literature review of some basic concepts related to CCS and  $CO_2$  leakage monitoring.

Chapter 3 describes a field characterization and experimental design. This study was carried out at Ito Natural Analog Site (INAS) field in Fukuoka, Japan. The test field is administratively belonged to Itoshima City, and has a humid subtropical or temperate climate. Two wellbores for the gas injection, 24 m apart from each other, were drilled in the test field with depths of 100.5 m and 113.0 m. The property of rock fractures and underground water at the test field were investigated. The geology of study site is composed of Itoshima granodiorite,

commonly classified in unfractured to moderate fractured, except in 62 m, 79 m, 81 to-89m, and 88 to 95 m depths where intensive discontinuity has occurred. Fracture dip orientation is mostly in SE direction. The hydrogeology of the area is very complex. Both of the boreholes have different groundwater level and groundwater flow only in 24 meters: 19 and 15 m depth of water level, NNE, and EES direction, and 0.67 cm/s and 1.06 cm/s, respectively. The total of 95 monitoring pipes (initially, 65 monitoring points) were set at intervals of 1.25 or 2.5 m, forming a rectangular grid, to provide an accurate spatial setting for monitoring and measuring the  $CO_2$  release above the test field. Also, another three boreholes approximately 19.5 m in depth were developed for carrying out the field tests for  $CO_2$  leakage monitoring.

Chapter 4 presents the description of the near-surface gas leakage monitoring after  $CO_2$  released at INAS site. The first injection experiment showed a significant increase of  $CO_2$  concentrations as an anomaly in 4 points, identified Y-E4, Y-F4, Y-F5 and Y-F6, in 64 points installed the monitoring pipes, i.e., 1.2, 3.1, 3.6 and 0.62 %, respectively. However, the same trend was not observed in the second and third experiment. In this study, the direct  $CO_2$  measurement was considerably influenced by seasonal and rainfall intensity and  $CO_2$  concentration tended to decrease after the rainfall. This study also demonstrated that  $CO_2$  concentration measured by pipes gas method occasionally gives low confident result due to the noise of external factor such as the natural  $CO_2$ . Considering that the quantity of natural  $CO_2$  flux produced from soil carbon was larger than that of  $CO_2$  flux produced from the storage site, the anomaly level of  $CO_2$  flux could have been hindered on direct surface monitoring. To increase the reliability of leakage detection, a new monitoring approach that utilizes  $H_2$  gas as a  $CO_2$  tracer has been proposed in this study.

Chapter 5 presents another field test for surface monitoring with  $H_2$  as a tracer to detect and predict potential CO<sub>2</sub> leakage from geological storage sites. The mixed gas (CO<sub>2</sub>:  $H_2 = 99:1$ ) was released through the injector well-b into 20 m depth of shallow aquifer. Gas pipe measurements showed that  $H_2$  was detected immediately in 15 ppm at the monitoring point just after mixed gas was released to the water saturated zone. Repeated measurements were then conducted at some monitoring points and recorded the elevation of  $H_2$  continuously between 15 – 65 ppm for two weeks. Meanwhile, pipe gas measurement only showed CO<sub>2</sub> elevation at one monitoring point a day after mixed gas was released. The field result was confirmed by laboratory experiments of mixed gas release which suggests that  $H_2$  was detected earlier than CO<sub>2</sub>.

Chapter 6 presents the conclusions of the study together with suggestions for future research. The elapsed time between H<sub>2</sub> and CO<sub>2</sub> surface detection after the release of mixed gas was observed in this study suggesting that H<sub>2</sub> has the potential to be an early signal precursor for CO<sub>2</sub> leakage. It would be very useful for early detection of a potential leakage of CO<sub>2</sub> in advance, so that mitigation actions can be implemented. However, it is necessary to investigate the applicability of H<sub>2</sub> as an effective monitoring methodology at CO<sub>2</sub> storage sites in various scales. More research is also required to not only test the sutability of H<sub>2</sub> for CO<sub>2</sub> leakage detection, but also using it for quantifying the potential leakages.