

# Subsurface CO<sub>2</sub> leakage : Lab-scale study of salient characteristics and assessment of borehole-based detection using resistivity tomography

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**ABSTRACT:** The geological sequestration of carbon dioxide (CO<sub>2</sub>) to reduce greenhouse gas emissions into the atmosphere faces difficulties related to non-homogeneous underground conditions, poorly characterized interconnected geo-systems, and complex hydro-chemo-mechanical effects that involve the reservoir rock and cap-rock mineralogy, the saturating fluid, and the injected fluid. Given these uncertainties, extensive monitoring of CO<sub>2</sub> injection projects is required. We developed a unique laboratory facility for the observation of subsurface CO<sub>2</sub> leakage evolution. A thin transparent tank is filled with different sizes of glass-beads to form controlled layered stratigraphies; then the medium is saturated with water mixed with a universal pH indicator. The flow-controlled injection of CO<sub>2</sub> is carefully controlled using pressure transducers with precise needle valve and, time-lapse photography permits capturing the evolution of gas invasion and diffusion. Results show the nature of CO<sub>2</sub> gas migration in the near surface, the effect of fine-grained layers such as the cap-rock, water acidification near conduits and subsequent diffusion, the convection of carbonated water. In addition to this trial to understand salient characteristics on subsurface CO<sub>2</sub> leakage, applicability of borehole based resistivity tomography is assessed. The measurement system for resistivity tomography is attached to the CO<sub>2</sub> gas migration monitoring system. An adequate inversion scheme is proposed based on 3-D forward modeling. The array types are dipole-dipole and modified pole-dipole, which were specially designed for this lab-scale test. A mixture of in-line and cross surveys is employed. Produced resistivity images are compared with time-lapse digital images taken during CO<sub>2</sub> gas leakage simulation. The visible feasibility check for the borehole resistivity tomography in detecting subsurface CO<sub>2</sub> leakage is expected.

## 1 INSTRUCTIONS

Global warming and extreme weather events are a hot issue in the world today. Carbon dioxide (CO<sub>2</sub>), one of the greenhouse gases, has been nominated as the main culprit of this international concern (United States Environmental Protection Agency, 1997). The carbon capture and storage (CCS) technique is grabbing attention because the current energy paradigm that focuses on fossil fuel cannot be changed abruptly. The CCS technique can be a stepping stone to a move to a new energy paradigm that focuses on new renewable energy (Intergovernmental Panel on Climate Change, 2005). However, CO<sub>2</sub> geological sequestration faces difficulties related to non-homogeneous underground conditions, poorly characterized interconnected geo-systems, and complex hydro-chemo-mechanical effects that involve reservoir rock and cap-rock mineralogy, saturating fluid, and injected fluid. Thus, it is considered as risky and uncertain, and there is the possibility of CO<sub>2</sub> leakage.

When injected CO<sub>2</sub> leaks, the CO<sub>2</sub> geological sequestration will be in vain at first. Moreover, we have already experienced disasters due to CO<sub>2</sub> leakage. In 1986, a massive CO<sub>2</sub> discharge from Lake Nyos in Cameroon suffocated many people and livestock (Smolowe, 1986). The trees at Mammoth Mountain in California are being killed by high concentrations of CO<sub>2</sub> gas in the soil (McGee et al., 1998). CO<sub>2</sub> vents lower the pH of the water column and it affects benthic ecosystems at shallow coastal sites (Orr et al., 2005). These examples are related to volcanic activity, but underground CO<sub>2</sub> storage can be another source for this kind of disaster. Owing to transportation cost, it is desirable that storage be located near power plants or human habitation. Failing to understand the phenomena related CO<sub>2</sub> geological sequestration will create another big problem to human life.

Given these concerns, extensive monitoring of CO<sub>2</sub> injection projects is necessary. For the first step of this huge project, we developed a unique laboratory facility for observing the evolution of subsurface

CO<sub>2</sub> leakage where we observe the phenomena related to CO<sub>2</sub> injection at an imaginary vertical section of underground sample. Photos were taken at regular intervals and we attempted to capture the evolution of gas invasion and diffusion. Although our study was with normal pressure and temperature conditions and small scale physical modeling, coupled hydro-chemo-mechanical (HCM) processes, which are difficult to simulate through numerical modeling, could be observed. This study provides a foundation for understanding the real phenomena related to CO<sub>2</sub> geological sequestration.

For monitoring CO<sub>2</sub> storage sites, use of geophysical tomography methods has been studied (Arts et al., 2000). Use of the geophysical methods performed on the ground surface is limited because the effective storage depth is about 1000 m. When CO<sub>2</sub> is dissolved in the water, the concentration of carbonated water is increased. This lowers the resistivity of the medium. On the other hand, CO<sub>2</sub> gas migration displaces water in pores and this increases the resistivity of the medium. CO<sub>2</sub> gas migration due to storage or leakage changes the resistivity of the medium, and it can be monitored through resistivity tomography (Nakatsuka et al., 2010). If a resistivity survey can catch the complex phenomena caused by subsurface CO<sub>2</sub> leakage, it should be considered as a concrete option for monitoring CO<sub>2</sub> storage sites. We attached a resistivity measurement system based on borehole resistivity tomography to the CO<sub>2</sub> gas migration monitoring system. We attempted to get resistivity images when the phenomena related to CO<sub>2</sub> leakage were taking place in the tank. These images were compared with the photos. We analyze and discuss the applicability of the resistivity survey.

## 2 EXPERIMENTS

### 2.1 CO<sub>2</sub> gas migration monitoring

The schematic diagram of the testing systems in this study is shown in Fig. 1. A very thin transparent tank (W × H × D=300 mm × 600 mm × 2 mm) was used for specimen preparation. We wanted to see the phenomena at a certain imaginary vertical section of an underground sample. Thickness was only 2 mm and we assumed it was a two dimensional model. The tank was filled with different sizes of glass-beads to form controlled layered stratigraphies; then the medium was saturated with water mixed with a universal pH indicator. A universal indicator is a pH indicator composed of a blend of several compounds that exhibits several smooth color changes over a pH value range from 1-14 to indicate the acidity or basicity of the solutions (from Wikipedia). It is green when the pH of the solution is 7. The color changes to the yellow and red color families with decreasing pH values. The flow-controlled injection of CO<sub>2</sub> was

carefully controlled using pressure transducers and a precise needle valve. To capture the evolution of gas invasion and diffusion, photos were taken at regular intervals. Flow paths, displacements, invading volume, pH and density contours of carbonated water were extracted by subsequent image analyses. Bubble pressure was also monitored using a pressure sensor attached to the bottom of the injection needle.

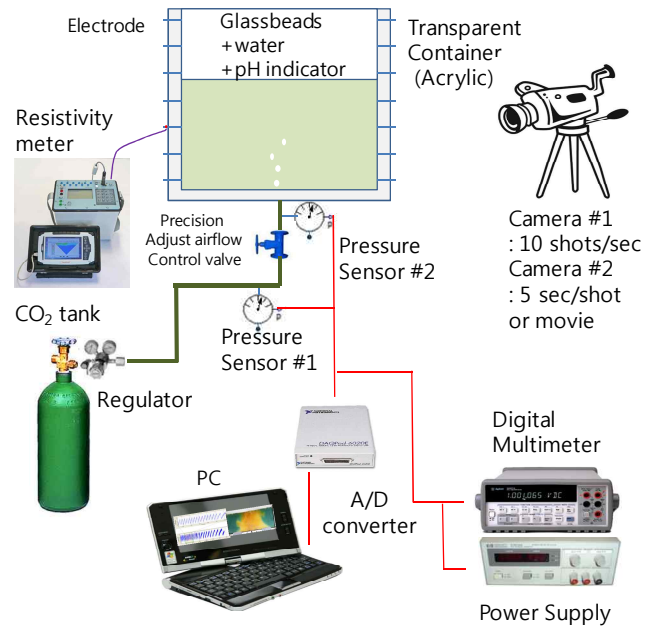


Figure 1. Schematic diagram of the testing systems for monitoring CO<sub>2</sub> gas migration. Depressurized CO<sub>2</sub> gas was injected into a transparent thin gap container through a needle. Bubble pressure was monitored with a pressure sensor attached near the injection point. Pictures were taken for recording and image processing.

To observe various phenomena, several models were used: a pH 7 water model, a one layer homogeneous soil-water mixture model, a sea bottom mountain model, a three-layered model containing a fine middle layer and so on.

### 2.2 Resistivity measurement and inversion

The measurement system for resistivity tomography was attached to the CO<sub>2</sub> gas migration monitoring system. Eleven electrodes were installed at the left and right boundaries of the tank. Two or more electrodes were installed at the top plate. The testing equipment for the resistivity survey was Syscal Pro from IRIS instruments. The survey method was a mixture of in-line and cross surveys. The connected CO<sub>2</sub> gas bubbles traversing the medium of the tank can be a barrier to measure reasonable potential between the left and right parts of the electrodes. The in-line survey will help this kind of problem. The array types were modified pole-dipole and dipole-dipole, which were specially designed for this lab test.

Various inversion schemes for resistivity tomography have been developed. A 3D inversion algorithm considering topography and location of elec-

trodes has already been in general (Yi et al., 2001). Now a 4D inversion algorithm for the dynamic earth has been developed (Kim et al., 2009). Conventional inversion algorithms are not applicable for this study because the tank is perfectly isolated. A modified inversion scheme based on general 3D forward modeling was developed. It considered the shape of the container and the bottom line was put to earth to find solutions in forward modeling. Active constraint balancing was used to enhance the resolving power of least squares inversion (Yi et al., 2003).

The main soil model for our resistivity survey monitoring is anticline structure, which is a typical type of structural/stratigraphical trapping for CO<sub>2</sub> geological sequestration (Fig. 2a). It is a three-layered model. Coarse glass-beads were used for the reservoir and upper layers. Fine glass-beads were used for the cap-rock. Red circles indicate trapped CO<sub>2</sub> gas.

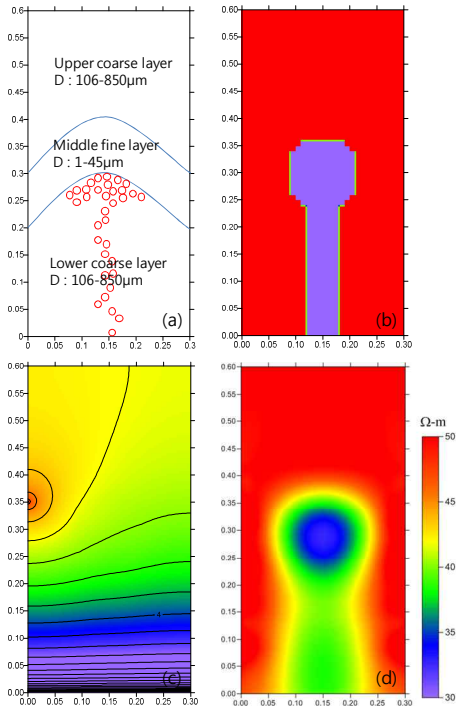


Figure 2. Verification of the inversion procedure for resistivity tomography through numerical modeling: (a) soil model for the feasibility test of resistivity tomography and expected form of trapped CO<sub>2</sub> gas bubble inside the medium, (b) simple model for numerical verification, (c) a typical potential contour during mono-pole current, (d) inversion result using numerical data.

The proposed inversion scheme was verified using numerical data. Fig. 2(b) is a simplified resistivity model for the main soil model shown in Fig 2(a). The resistivity of the anomaly caused by CO<sub>2</sub> dissolved water was 30 Ohm-m while the resistivity of the background medium was 50 Ohm-m. The size of model was 300 mm x 600 mm x 2 mm and the interval of the imaginary electrode was 50 mm. This is same dimensions as the experimental setup. The size of the elements was 1 mm x 1 mm x 0.5 mm and the number of nodes were 904,505 (301x601x5). Fig. 2(c) shows an example of the potential contour when a current was injected through an imaginary mono-

pole electrode at 0.35 m of the left boundary. The bottom line was put to earth while the other boundaries were considered as having the Neumann condition. Thus, the potential was zero at the bottom line. Using all twenty-two electrodes, in-line, cross, reverse-cross surveys were performed through dipole-dipole electrode arrays. Inversion was done using a total of 216 measurement data items. The constructed image of resistivity is shown in Fig. 2(d). The block size for the inversion was 5 mm x 5 mm and total number of blocks was 450. Due to the smoothness constraint, it appears that the boundary of the anomaly has expanded, but it shows well the low resistivity anomaly at the center of the model. Furthermore, the resistivity value is similar to the original value of the model. Thus, it appears that the proposed inversion scheme for resistivity survey works well for this kind of lab-scale study.

### 3 RESULTS AND DISCUSSION

#### 3.1 Gas migration and water acidification near the conduit

Fig. 3 shows the representative time-lapse images taken by a digital camera for the simulation of a sea bottom mountain model. The path of CO<sub>2</sub> gas migration is easily observed. The color of the pore water near the conduit is changed due to acidification because CO<sub>2</sub> is dissolved and makes carbonated water. Pore-water is pH sensitive because the universal pH indicator is mixed. It changes color with different pH values.

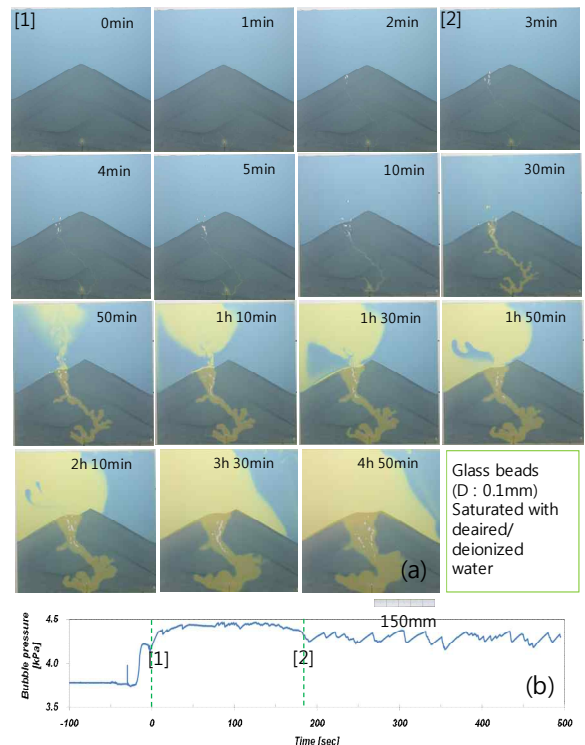


Figure 3. The results of CO<sub>2</sub> gas injection into a sea bottom mountain model. (a) time-lapse digital images, (b) time history of gas bubble pressure.

In this way, the path of the CO<sub>2</sub> gas is clearly detected. The direction of gas migration is upward due to buoyancy. When medium is not homogenous, injected CO<sub>2</sub> tries to find an easier way to migrate, so the direction of gas migration can be horizontal and sometimes downward. This model was non-homogeneous due to segregation of the glass-beads when the specimen was prepared. Therefore, the path of the CO<sub>2</sub> gas was not simple and it changed with time. Fig. 3(b) shows the time history of the bubble pressure inside. When the gas migrated, the pressure of the gas bubbles changed. There is a drop point [2] in the graph which means the gas bubble broke through the medium of glass-beads. After that, there were repeated fluctuations of pressure due to continuous gas migration.

In the three-layered model containing a fine layer, other interesting phenomena related to gas migration were observed as shown in Fig. 4. There were three stages. In the first stage, CO<sub>2</sub> gas migrated into interconnected pores displacing pore-water in the bottom layer. There was no big pressure build-up. There are just small pressure fluctuations during the gas migration. Otherwise, in the second stage, high pressurized gas made cracks in the fine middle layer to advance. The bubbles pressure increased continuously and dropped a little repeatedly when the gas advanced into the fine layer making cracks.

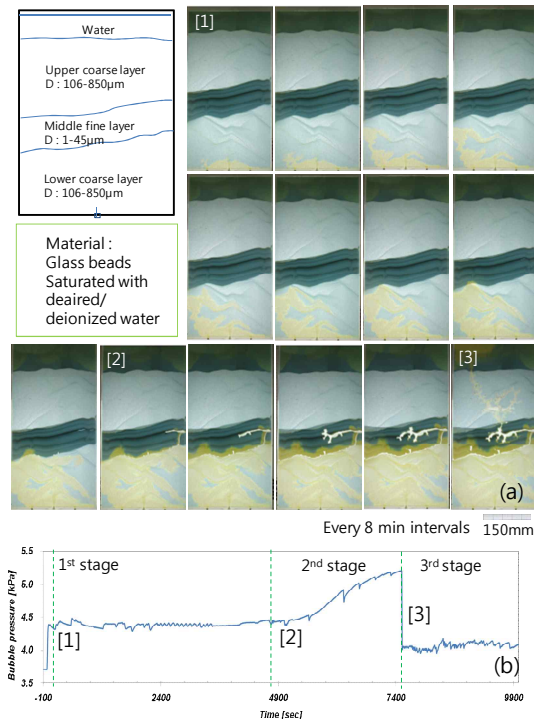


Figure 4. The results of CO<sub>2</sub> gas injection into a layered glass-bead-water mixture system having a fine middle layer: (a) time-lapse digital images, (b) time history of gas bubble pressure.

When the gas broke through into the second fine layer, the bubble pressure dropped abruptly at point [3] in the Fig. 4(b). After that, the CO<sub>2</sub> gas migrated freely through the percolated path that was the third

stage. When the bubble pressure is larger than the air entry value of the medium and smaller than confining pressure, it migrates into the interconnected pores. On other hand, when the bubble pressure is smaller than the air entry value of the medium and larger than the confining pressure, it migrated by open mode discontinuity.

### 3.2 Subsequent diffusion and convection of carbonated water

Dissolved CO<sub>2</sub> (carbonated water) moves in the medium in two major ways. One is diffusion and the other is convection. Due to a concentration difference, carbonated water near a conduit moves to the far area. In this case, there is no movement of the pore-water. Only carbonated ions move inside the pore-water. Fig 5(a) shows the time-lapse images for the diffusion test using a horizontal plate having a thin gap. When a small amount of CO<sub>2</sub> gas was injected at the center of a plate filled with water, it started to dissolve into the water. There was no convection due to gravity because the plate was horizontal. The carbonated water zone expanded only through diffusion.

Fig. 5(b) shows the diffusion within the fine glass-bead layer saturated with water in a thin vertical tank. These pictures are from the results of the three-layered model containing a fine layer in the previous section. It was impossible for the CO<sub>2</sub> gas to migrate into the interconnected pores in the fine layer because the air entry value was much larger than the CO<sub>2</sub> bubble pressure. The expanding of the carbonated zone was only from diffusion in this area. That means products of CO<sub>2</sub> gas can leak from storage or a reservoir even if there is no crack in the cap-rock.

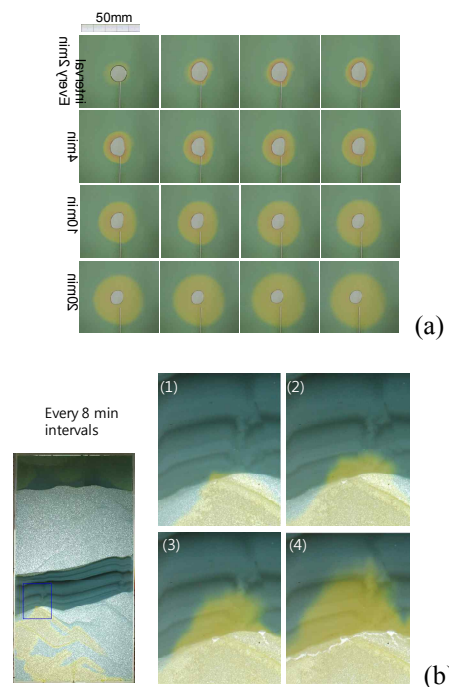


Figure 5. Diffusion of carbonated water: (a) in a horizontal plate having a thin gap. (b) within a fine glass-bead layer saturated with water in a thin vertical tank.



Fig. 6(a) shows the convection of carbonated water caused by gravity. When some CO<sub>2</sub> gas was injected at the bottom of the plate just containing pH 7-water, it was partially dissolved into the water before going out to the air. A carbonated water zone with a half circle shape was created at the top of the water. The density of carbonated water is denser than the surrounding water (Garcia, 2001), so carbonated water tends to move downward. Carbonated water with a lower pH has a relatively larger density. The downward movement of water with a lower pH is relatively faster.

When CO<sub>2</sub> gas was injected into the glass-bead medium saturated with water in a thin vertical tank, the shape of the carbonated water zone was a reverse triangle in the beginning period of CO<sub>2</sub> injection. Later, the shape changed to a normal triangle because of the current of carbonated water caused by convection (Fig. 6(b)). The size of glass-beads used for the model in Fig. 6(b) was 0.5 mm, which is larger than that in Fig. 3(a). If the size is coarser, the convection is superior. If the size is finer, the diffusion is superior when the carbonated water moves inside a saturated medium.

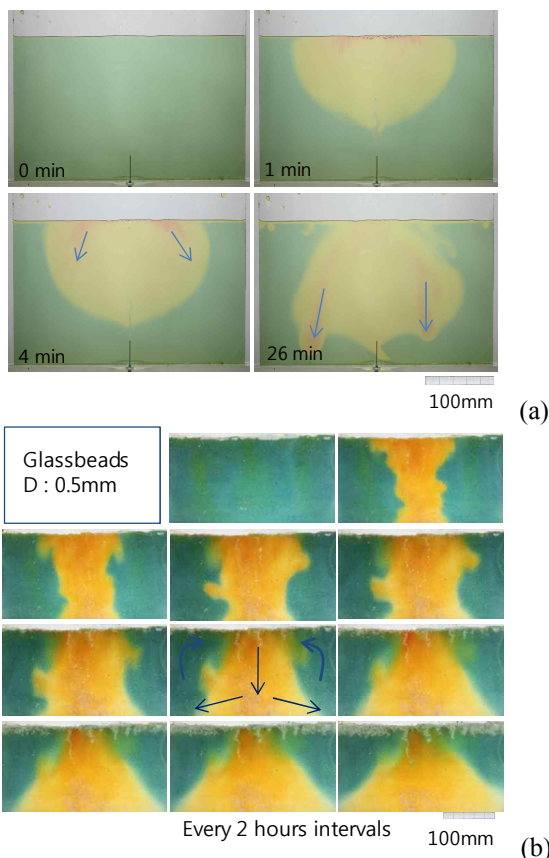


Figure 6. Convection due to density difference of carbonated water: (a) within the water in a vertical plate having a thin gap, relatively faster, (b) within glass-bead medium saturated with water in a thin vertical tank.

### 3.3 Time-lapse images of resistivity survey

Fig. 7 shows representative photos of the test in which CO<sub>2</sub> gas was injected into the tank containing

only water. The initial color was in the blue family. The color changed into the yellow and red color families around the path of the bubble rising. Furthermore, the area of carbonated water expanded or enlarged from diffusion and convection. During the CO<sub>2</sub> gas injection, a resistivity tomography survey was performed periodically. However, those resistivity images didn't match well with the photos. It may be that there are artifacts due to erroneous measurement procedure and inversion scheme. An additional experiment is being done to modify the results of the resistivity survey. Another application test using the main soil model of Fig. 2(a) is also being done. The feasibility of the resistivity tomography survey for complex phenomena related to CO<sub>2</sub> leakage will be discussed later.

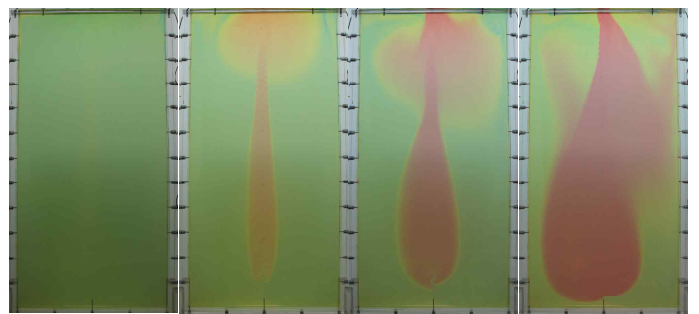


Figure 7. Representative photos for CO<sub>2</sub> gas injection and application of resistivity tomography survey.

## 4 CONCLUSION

A unique laboratory facility for observing the evolution of subsurface CO<sub>2</sub> leakage was developed. A resistivity measurement system based on borehole resistivity tomography was included. A universal pH indicator especially helped the movement of carbonated water to be monitored effectively. The nature of CO<sub>2</sub> gas migration, the effect of fine-grained layers such as the cap-rock, water acidification near conduits and subsequent diffusion, the convection of carbonated water were observed. This study was a first step to understanding the salient characteristics of subsurface CO<sub>2</sub> leakage and an assessment of the applicability of borehole-based resistivity tomography. Some of significant observations and discussions from this study are as follows:

The path of CO<sub>2</sub> gas migration in the medium of a tank is easily observed. The color of water near the path of the CO<sub>2</sub> changes from acidification because CO<sub>2</sub> is dissolved and makes carbonated water. When the gas migrates the pressure of gas bubbles changes. When the bubble pressure is larger than the air entry value of the medium and smaller than the confining pressure, it migrates through the interconnected pores. On other hand, when the bubble pressure is smaller than the air entry value of the medium and larger than the confining pressure, it migrates through open mode discontinuity.

Carbonated water tends to move downward because carbonated water is denser than the surrounding water. Carbonated water with a lower pH has a relatively larger density. The downward movement of water with a lower pH is relatively faster. If the grain size of the medium is coarser, the convection is superior and if the grain size is finer, the diffusion of the carbonated water is superior. Products of CO<sub>2</sub> gas can leak through diffusion from storage or a reservoir even if there is no crack in the cap-rock.

An inversion scheme was developed based on 3-D forward modeling that considers the shape of the container. Through numerical modeling, the proposed inversion scheme was verified. Nevertheless, to use the scheme with real experimental data, a refined testing procedure and modification of the proposed inversion scheme are necessary and will be pursued.

## 5 ACKNOWLEDGEMENT

Support for this research was provided by the USA Department of Energy, the Goizueta Foundation and the basic research project of KIGAM.

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