Greenhouse Gas Sequestration by a Novel Microbubble Injection Technology in Saline Aquifers

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ABSTRACT

Natural gas has been defined as a clean energy source, which may contribute to increasing fuel efficiency and fuel switching in the short and medium term (until 2020). But in the mid and long term (2020 to 2050), the unfavorable possibility exists that natural gas will be included in the category of the fossil fuels. The consumers will preferentially select alternative clean energy source if the gas industry does nothing. It is essential for city gas suppliers to take measures to reduce CO_2 emissions not only for the short term but also for the mid and long term. Carbon dioxide capture and sequestration (CCS) is one of the key technologies for controlling CO_2 emission caused by natural gas consumption in the mid and long term.

In Japan, sites with a CO_2 storage capacity of over 1×10^6 t-CO2/yr are limited owing to the fact that the geological structures in Japan are generally complicated, with alternate layers of sand and clay, which limit the expansion of saline aquifers. A method that is more efficient and safer than the conventional technology used for injecting CO_2 into the saline aquifers in Japan would be required for medium- or small-scale CO_2 storage sites.

CO_2 microbubble injection was carried out in laboratory experiments as an alternative to the conventional CCS technology. Using a newly developed filter, we successfully produced CO_2 microbubbles under simulated conditions similar to those in a deep saline aquifer. This microbubble injection method can serve as a CCS technology for realising efficient and safe CO_2 injection in saline aquifers; it will be especially effective for medium- or small-scale CO_2 storage sites, which have complicated geological structures. This microbubble production method may be applicable not only to saline aquifers but also to oil reservoirs for enhanced oil recovery (EOR).

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1. Introduction

  Global warming has been attributed to the increasing atmospheric concentrations of the greenhouse gases emitted during heavy consumption of fossil fuels such as coal and petroleum oil. The total amount of the greenhouse gases in CO_2 equivalent emitted in Japan was $1,209 \times 10^6$ tons in 2009. This amount was reduced by 4.1% relative to the base year of the Kyoto Protocol. However, it is unclear whether the treaty can be accomplished, in which Japan has pledged to reduce its 1990 emissions by 6% by 2012, unless the efforts required to mitigate the greenhouse gas emissions are sustained.

  CO_2 accounts for 95% of the greenhouse gas emissions in Japan. The major emission sources of CO_2 include coal- and oil-fire power plants, steel plants, chemical complexes etc. Carbon dioxide capture and sequestration (CCS) technology should play an important role in reducing the CO_2 emissions from these major sources. According to the IEA Energy Technology Perspective 2010, CO_2 emissions worldwide will be reduced by 50% in 2050 from the level in 2005. Ensuring high fuel efficiency, fuel switching, using nuclear power and renewable energy sources, and CCS are the major countermeasures for emission reduction. CCS, in particular, is expected to be responsible for about 20% of the total emission reduction in 2050,[1] as shown in Figure 1. This technology has already been adopted at several demonstration and commercial sites, e.g., Sleipner,[2] In Salah,[3] and Nagaoka.[4]
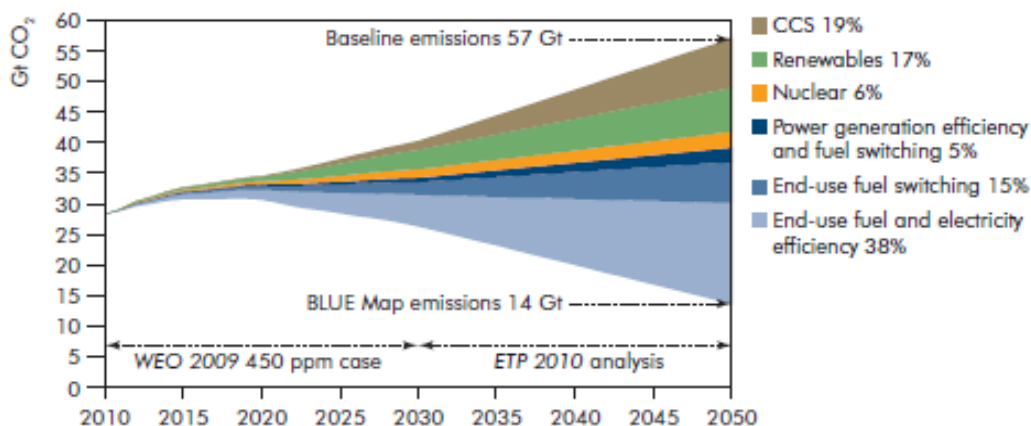

Figure 1. Key technologies for reducing CO_2 emissions (IEA 2010).

  In the case of the city gas supply business, the CO_2 emissions that accompany the regasification and supply of gas from liquefied natural gas (LNG) in LNG receiving terminals and pipelines are very low. The amount of CO_2 emissions generated by the regasification and supply processes of Tokyo Gas was approximately 250 thousand tons in 2009, and the total amount of city gas supply was 11×10^9 m^3. However, the emission generated by gas consumption has been estimated to be approximately 2 kg of CO_2 for every 1-m^3 consumption of city gas on a per consumer basis. The consumer emission from Tokyo Gas in 2009 was estimated at 24.5×10^6 tons CO_2 on the basis of the total city gas consumption. Natural gas has been defined as a clean energy that contributes to increasing the fuel efficiency and fuel switching in the short and mid term (until 2020), but in the mid and long term (2020 to 2050), an unfavorable possibility exists that natural gas will be listed as one of the fossil fuels and consumers will select alternative clean energy sources. It is imperative for city gas suppliers to take measures to reduce CO_2 emissions not only for the short term but also for the mid and long term. CCS is one of the key technologies for controlling CO_2 emission caused by natural gas consumption in the mid and long term.

In Japan, the CO_2 storage potential of saline aquifers covered by cap rocks forming a dome structure (category A) is estimated to be approximately 30×10^9 tons; the CO_2 storage potential of saline aquifers covered by cap rocks forming the dome or a horizontal structure (category A & B) is estimated to be approximately 146×10^9 tons.[5] However, sites with a CO_2 storage capacity of over 1×10^6 t-CO_2/yr are limited owing to the fact that the geological structures in Japan are generally complicated with alternate layers of sand and clay, which limits the expansion of saline aquifers.

A method that is more efficient and safer than the conventional technology for injecting CO_2 into these saline aquifers in Japan would be required for medium- or small-scale CO_2 storage sites (storage capacity of less than 1×10^6 t-CO_2/yr). For this purpose, we focused on using CO_2 microbubbles. A microbubble is generally defined as a bubble with a size of 1~100 μm. Microbubbles have several unique features. First, they move very slowly in water; second, their surface area of a swarm of microbubbles is greater than that of a single normal bubble of a comparable size. Figure 2 shows a conceptual diagram of microbubble behavior. By using an injection of technique for introduction of the CO_2 microbubbles, the areas of contact between the CO_2 microbubbles and saline water in the aquifer can be increased, and the time required for the microbubble to move from the injection points to the cap rocks can be prolonged. The injected CO_2 microbubbles are expected to immediately dissolve in saline water. Thereby mitigating any increase in pressure beneath the cap rocks. These unique features of microbubbles can be exploited in CCS for efficient and safe CO_2 injection in a saline aquifer.

The microbubble technology can increase the feasibility of the case, in which CO_2 is captured from distributed energy sources and sequestrated into medium- or small-scale storage sites. Furthermore, it is also considered to be beneficial to CO_2 sequestration into large storage sites.
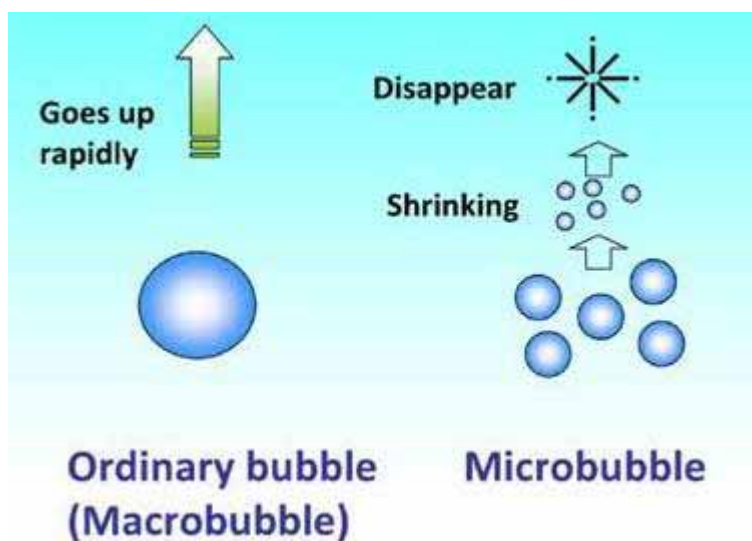

Figure 2. Conceptual diagram of microbubble behavior.

2. Fundamentals of technology

2.1 CO_2 trapping mechanisms

Several CO_2 trapping mechanisms have been evaluated and proposed. The dominant trapping mechanisms for CCS include physical trapping, dissolution, residual gas trapping and mineralization.[6] A general representation of the evolution of trapping mechanisms over time is shown in Figure 3. In the physical trapping mechanism, the injected CO_2 floats up into the underside of the cap rocks because of the buoyant force generated by the difference in the density of the CO_2 bubbles and the saline water. The CO_2 then remains trapped under the cap rocks for a

long time. Physical trapping is one of the most important mechanisms for CCS in saline aquifers. Injected CO_2 in saline aquifers is expected to dissolve in the saline water with time. This mechanism is defined as dissolution and is considered to have greater long term stability than physical trapping. The saline water in which CO_2 is dissolved acidulates, and the dissolved CO_2 gradually reacts with cations contained in the rocks of the underground layers. Finally, minerals are formed, which are the most stable substances in CO_2 trapping. The mineralized CO_2 is safely stored within the saline aquifers for distinctly long periods. However, mineralization proceeds over thousands or tens of thousands of years. Dissolution and mineralization can be facilitated using the CO_2 microbubble injection technique.

Incidentally residual gas trapping is one trapping mechanism that has attracted attention for academic and practical reasons. This mechanism is recognizes as the phenomenon, in which natural gas localizes in the void spaces of natural gas reservoirs by hydraulic injections. In CO_2 trapping during CCS, the injected CO_2 has also been confirmed to localize in the porous structure of the layers. The amount of CO_2 trapped by this mechanism is thought to account for 20% of the total trapped CO_2 in saline aquifers, as previously reported.[7]
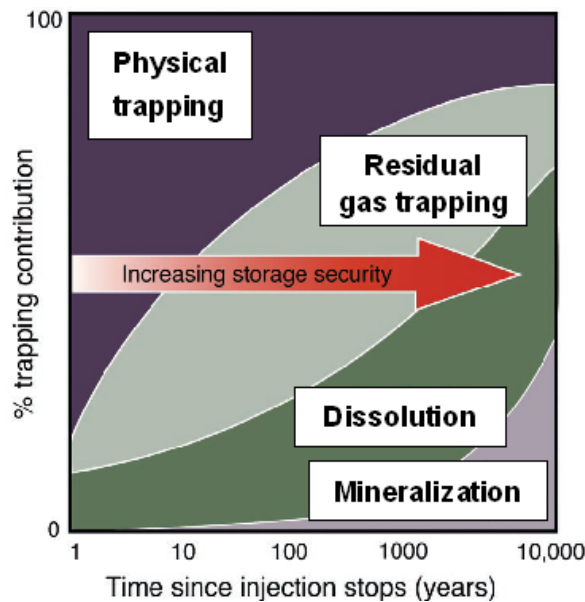

Figure 3. General representation of evolution of trapping mechanisms over time (IPCC).

2.2 Mechanisms for microbubble generation

Several methods have been proposed for the generation of microbubbles. Table 1 shows the conventional methods used for generating microbubbles. In many cases, the generated microbubbles are intended to be utilized under atmospheric pressure conditions such as in water treatments, cleaning and medical applications. However, for utilization in CCS, the pressure at the CO_2 injection points can be as high as 6~10 MPa. We selected a filtering method in which high-pressure CO_2 is filtered through a microporous structure because this method of microbubble generation is both simple and easily applicable under hyperbaric environments.

Table 1 Mechanisms of microbubble generation

Method	Mechanism	Diameter of bubbles
Cavitation	Boiled bubbles are generated under rapid pressure reduction by screw rotations.	< 10 μm
Crushing	Crushed bubbles are generated under rapid pressure fluctuation by microwaves.	< 10 μm
Pressurized dissolution	Dissolved bubbles are generated in static state after the solubility is increased under high pressure.	10~100 μm
Turbulent shear flow	Separated bubbles are generated by shear stress in turbulent flow of gas and water.	10~100 μm
Filtering	High-pressure bubbles are generated through microporous structures	1~100 μm

The filter used in this method must have very fine pores and the pore sizes must be fairly uniform for the effective generation of microbubbles. Therefore, further studies on the materials and the structures of the filters are necessary.

3. Experimental

3.1 Selection of filter for microbubble generation

Prior to performing laboratory experiments on CO_2 injection in a simulated saline aquifer, the materials and the structures of the filters were investigated for optimized CO_2 microbubble generation. A series of screening experience on a number of materials and structures revealed that filters made from grindstone had very fine pores with a fairly uniform size.

Figure 4 shows the filter made from grindstone. This filter has been traditionally used for wafer processing in the semiconductor industry. The filter has very fine pores (with sizes less than approximately 10 μm) and the pore size is fairly uniform as shown in a micrograph of the filter surface (Figure 5). The filter made from grindstone was selected for the effective generation of microbubbles in this study.

For reference, filters made from other materials were also evaluated and found to be less effective for the generation of microbubbles. Figure 6 shows a filter made from sintered stainless steel. This filter is typically used in various industries relevant to gas and water. However, the pores of the stainless steel filter are not as fine as those of the grindstone filter and the pore size is irregular as shown in a micrograph of the filter surface (Figure 7).
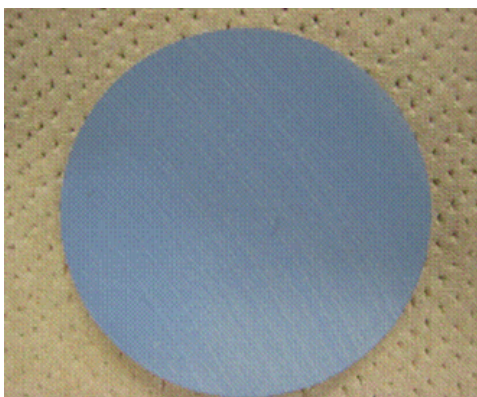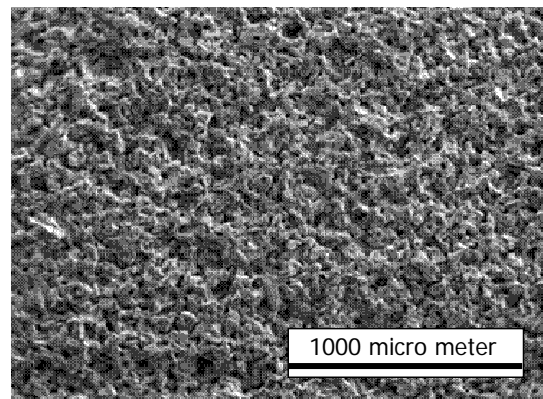

Figure 4. Grindstone filter.

1000 micro meter

Figure 5. Micrograph of surface of grindstone filter.

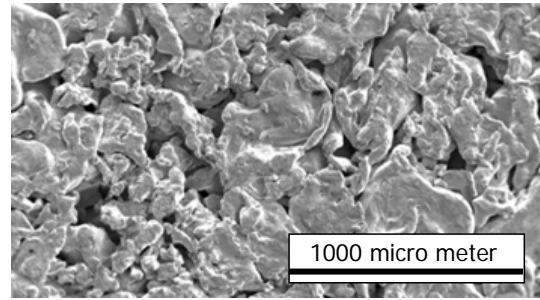Figure 6. Sintered stainless steel filter.

1000 micro meter

Figure 7. Micrograph of surface of sintered stainless steel filter.

3.2 Laboratory experiments for observation of microbubbles in simulated saline aquifer

Laboratory experiments were performed using an apparatus that simulates the pressure and temperature conditions in a deep aquifer. Figures 8 and 9 show a photograph and a schematic of the experimental apparatus, respectively. The pressure vessel in the apparatus was filled with saline water, and the conditions of a saline aquifer were simulated. CO_2 was injected into the pressure vessel at a specified injection rate by using a syringe pump, and the pressure in the vessel was maintained at a constant level by means of a regulator valve located upstream of the vessel and a syringe pump for water exhaust placed downstream. The temperature of the saline water was controlled using a ribbon heater covering the vessel. The filter made from grindstone was placed at the entrance of the vessel. The range of temperatures, pressures, and CO_2 injection speeds employed in the experiments were 20~40°C, 6~10 MPa, and 0.1~5.0 ml/min, respectively.

A high-speed camera was used to observe and record the behavior of the generated microbubbles. The behavior of the microbubbles was also analyzed using image analysis software.

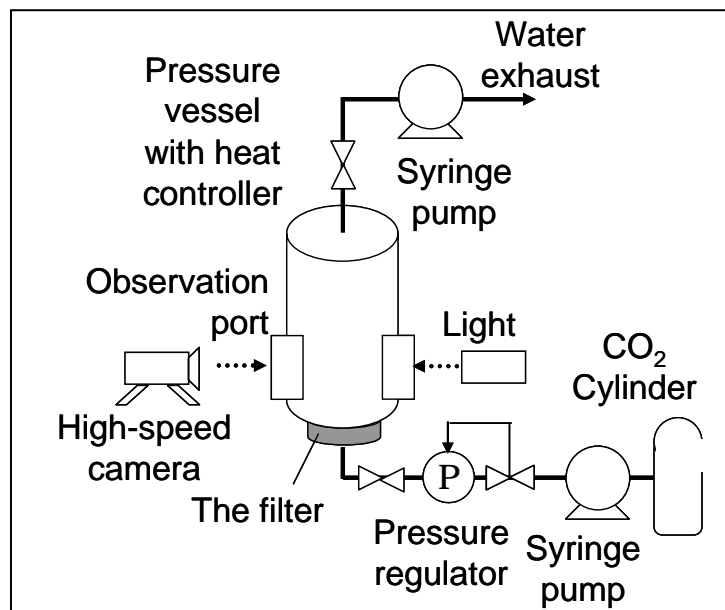Figure 8. Photograph of experimental apparatus.

Figure 9. Schematic of experimental apparatus.

4. Results and Discussion

4.1 Observation of injected CO_2 microbubbles

CO_2 microbubble injection was carried out at 40 ^{o}C and 10 MPa, which simulates the conditions in a saline aquifer at a depth of 1000 m. Figure 10 shows the behavior of the CO_2 microbubbles; the injection flow is upward from the bottom of the figure. As shown in the area encircled by the red ellipsis, the injected CO_2 produced a large number of microbubbles. It was observed that the CO_2 microbubbles were concentrated in the region close to the filter surface because the floating speed of the microbubbles was very slow. In addition, as compared to normal bubbles, these bubbles dissolved immediately in the water in the pressure vessel. It was experimentally confirmed that CO_2 dissolution in the water was enhanced, and that CO_2 was sequestered efficiently and safely by microbubble injection.

Figure 10. Behavior of injected CO_2 microbubbles (40°C, 10 MPa)

4.2 Dissolution rate of microbubbles

Quantitative evaluation was carried out to compare the dissolution speed of injected CO_2 microbubbles with that of a large bubble; the evaluation was conducted using a high-speed camera and image analysis software. Figure 11 shows that the dissolution speed of the swarm of microbubbles was faster than that of the large bubble. At the time of injection, the overall volume of the microbubble swarm was equal to that of the large bubble. The figure shows that, with time, the volume of the swarm of microbubbles decreased rapidly as compared to that of the large bubble. The ratio of the decreased volume to the original volume of the microbubble swarm (decreased size of bubbles/original size of the bubbles) was estimated to be 0.7 within 2 s of starting the injection. In contrast, this ratio was estimated to be 0.85 for the large bubble, which indicates that the microbubbles dissolved at least 20% faster than the large bubble. This result is considered to contribute to increased efficiency of the CO_2 injection in saline aquifers.
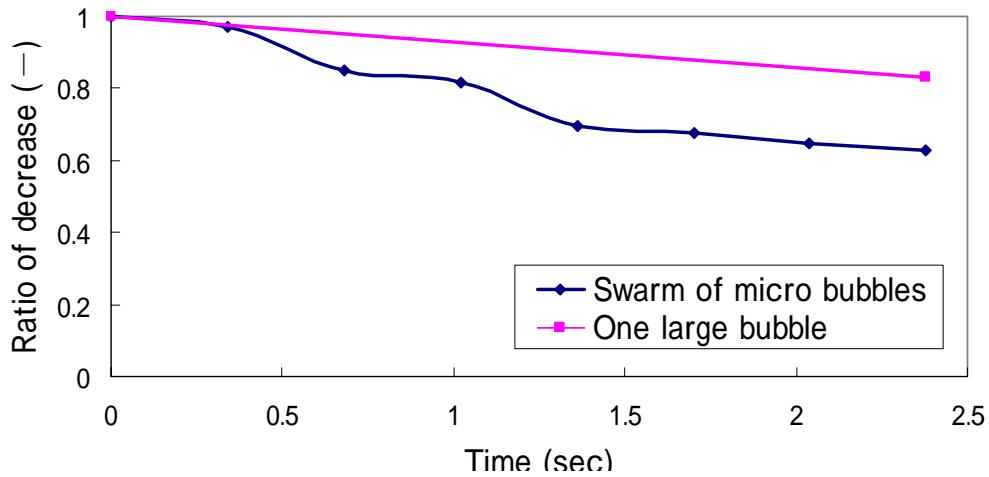
Figure 11. Comparison of decrease in volumes of microbubbles and large bubble over time.

4.3 Sedimentation phenomenon of microbubbles

With respect to the microbubbles near the filter surface, a sedimentation phenomenon was observed when CO_2 was injected upward from the bottom of the pressure vessel. The detailed behavior of sinking microbubbles is shown in Figure 12. Given that the density of the injected CO_2 is never greater than the density of water under the conditions of the pressure vessel, it is very interesting that the counter-intuitive sedimentation phenomenon was observed. The CO_2 microbubbles dissolve rapidly in the saline water as soon as the CO_2 is injected. The partial density of water increases rapidly upon CO_2 dissolution. A number of microbubbles are sunken by the downflow of the 'heavyish' water. This sedimentation phenomenon is considered to contribute to increased safety of the CO_2 injection in saline aquifers because the sinking CO_2 bubbles stay near the injection point.
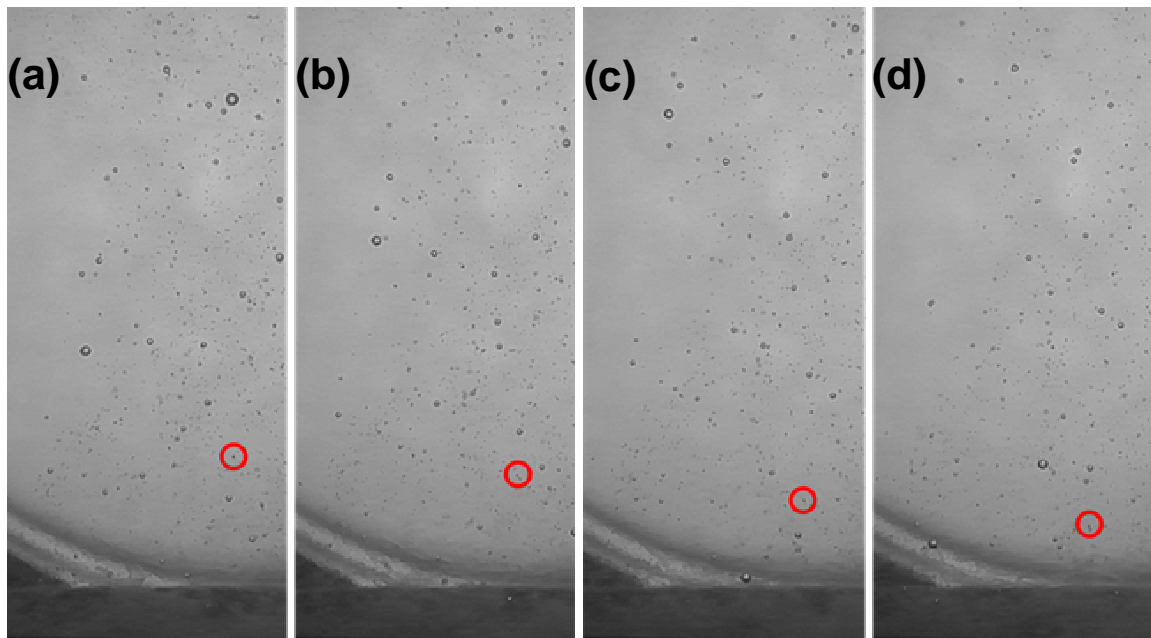

Figure 12. Sedimentation phenomenon of injected microbubbles.
(a) Initial state of phenomenon, (b) after 0.1 s (c) after 0.2 s and (d) after 0.3 s.
The red circles indicate the motion position of a microbubble.

5. Conclusion

The technology for CO_2 microbubble injection in CCS was improved by using a newly developed filter made from grindstone. Owing to the fine pores and the uniform pore size of the developed filter, CO_2 microbubbles were successfully generated in laboratory experiments under simulated conditions similar to those in a deep saline aquifer. Image analysis of the injected CO_2 microbubbles indicated faster dissolution of the injected CO_2 microbubbles as compared to a large bubble under the simulated aquifer conditions. Further, a sedimentation phenomenon near the injection points was confirmed to contribute to the safety of the CO_2 injection technology for saline aquifers.

This microbubble injection method can be employed as a CCS technology for realizing efficient and safe CO_2 injection in saline aquifers; it should be especially effective in the case, in which CO_2 is captured from distributed energy sources and sequestrated into medium- or small-scale storage site, which have complicated geological structures. For the practical implementation of this technology, the behavior of the injected CO_2 microbubbles should be investigated using a typical monitoring methodology, i.e., by performing resistivity and elastic wave measurements and direct observation using a high-speed camera. These measurements will be conducted in our future experiments.

This microbubble generation method may be applicable not only to saline aquifers but also to oil reservoirs for enhanced oil recovery (EOR). Further research on microbubble behavior in oils is required.

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