Greenhouse Gas Control Technologies for Distributed Energy Systems in Near-to-mid and Long-term Perspectives

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Introduction

Global warming is caused mainly by increasing atmospheric concentrations of greenhouse gases emitted from the heavy consumption of fossil fuels such as coal, petroleum oil, and natural gas. The total amount of greenhouse gases in CO_2 equivalent emitted in Japan was $1{,}256 \times 10^6$ tons in 2010 [1]. It is unclear whether the goals of the Kyoto Protocol, in which Japan agreed to a 6% reduction from its 1990 greenhouse gas emission levels by 2012, can be met unless the efforts required to mitigate 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-, oil-, and gas-fired power plants, steel plants, chemical complexes, etc. Carbon dioxide capture and storage (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 2005 levels. Increased fuel efficiency, fuel switching, using nuclear power and renewable energy sources, and CCS are the major countermeasures for CO_2 emission reduction. CCS, in particular, is expected to be responsible for approximately 20% of the total CO_2 emission reduction by 2050 [2], as shown in Figure 1. This technology has already been adopted at several demonstration and commercial sites, e.g. Sleipner, In Salah [3], and Nagaoka [4].

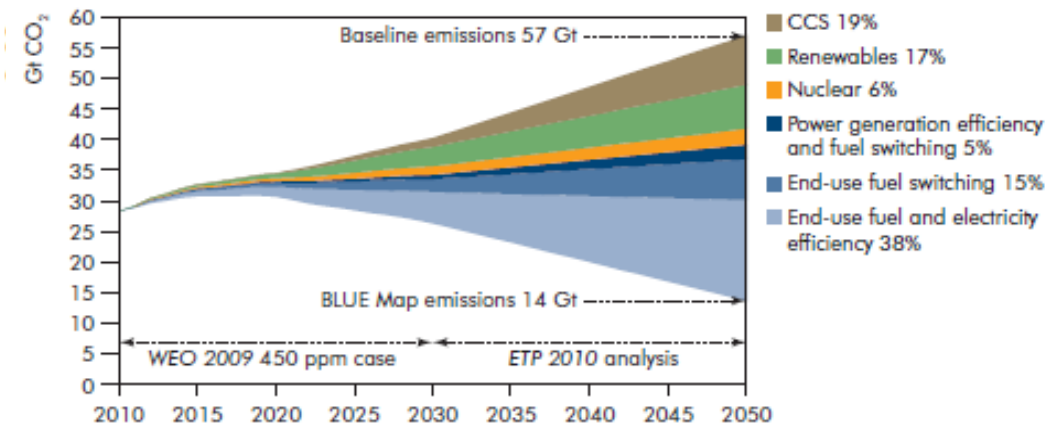

Figure 1. Key technologies for reducing CO_2 emissions (IEA 2010).

In the case of municipal natural gas supply businesses, the CO_2 emissions accompanying 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 emission generated by the regasification and supply processes of Tokyo Gas was approximately 264,000 tons in 2010, and the total amount of the city gas supply was 11.25×10^9 m^3. However, the CO_2 emission generated by gas consumption has been estimated to be approximately 2.2 kg/m^3. The consumer emission from Tokyo Gas in 2010 was estimated at 24.86×10^6 tons CO_2 based on the total natural gas consumption [5].

In the short- and mid-term (until 2020), natural gas has been defined as a clean energy source that contributes to increasing fuel efficiency and fuel switching. A tentative goal for the incorporation of CCS in distributed energy systems is to attain 'CCS-ready' status, wherein sufficiently improved technology for capturing and transporting CO_2 are available. CO_2 can be recovered from distributed energy systems at the appropriate scale, cost, and level of additional energy required for capture. By promoting commercial utilization of captured CO_2, CCS-ready status will soon be achieved.

Considering mid- and long-term periods (after 2020), CCS may be essential for natural gas utilization to reduce CO_2 emission levels much less than CO2 emission reductions achieved by improving the efficiency and fuel switching, because natural gas is a fossil fuel. The CO_2 reduction target set for 2050 cannot be achieved only by improvement of efficiency and fuel switching. CCS is considered to be a CO_2 reduction method for large-scale CO_2 emission sites such as coal power plants, but it can also be used for distributed CO_2 emission sites such as distributed natural gas power generation sites. For sustainable growth of the gas industry, an appropriate amount of CO_2 from distributed energy systems should be captured and stored.

In this paper, an available option for reducing CO_2 emission levels in distributed energy systems in both near-to-mid-term and mid-to-long-term periods is suggested. Fuel cells and hydrogen stations are considered main targets for capturing CO_2 because CO_2 emissions from these systems are highly concentrated. These distributed energy systems have already been launched at the demonstration stage. The CO_2 captured will be utilized as an industrial gas in factories and dry ice manufacturing plants. It can be also used as a raw material for chemical production. Such developments in the capture and utilization of CO_2 in these commercial systems will lead to the realization of CCS-ready status in the short- and mid-term periods. After 2020, captured CO_2 will be permanently sequestered in saline aquifers. The geological structures in Japan are generally complicated, with alternating layers of sand and clay that limit the expansion of saline aquifers. Medium- or small-scale CO_2 storage sites would be considered as an option for CCS in Japan. To implement CCS in distributed energy sources, a more efficient and safer method for injecting CO_2 into these saline aquifers in Japan compared to conventional technology would be required for small- or medium-scale CO_2 storage sites. A novel microbubble injection technology [6] will meet this requirement.

Capture and utilization of CO_2 for realization of CCS-ready status

CO_2 capture from phosphoric acid fuel cells

Fuel cells are presently considered to be one of the appropriate emission sources among distributed energy systems for CO_2 capture. Figure 2 indicates the difference in mechanisms between a fuel cell and an internal combustion engine such as a turbine or piston engine. The CO_2 density of exhaust gases from the reformer in a phosphoric acid fuel cell (PAFC) is four times higher than that from a gas engine or turbine. In a PAFC, fuel first reacts with steam and produces hydrogen in the fuel electrode. The hydrogen moves to the air electrode, which is separated by electrolytes from the fuel electrode, and produces electricity by reacting with oxygen in the air. The off-gas from the fuel electrode mixes with air and burns to produce CO_2. In an internal combustion engine, fuel mixes with air before power generation to combust and produce CO_2. The amount of air that is in contact with fuel in a gas engine or turbine is four times more than that in contact with fuel in a PAFC. Because of this difference, the amount of energy required to separate CO_2 from the exhaust gas is less for a fuel cell than for an internal combustion engine.

CCS technology needs a large amount of energy to separate CO_2 gas from exhaust gas. A higher concentration of CO_2 in the exhaust gas means less energy is needed for separation. Therefore, the required energy of CO2 separation for fuel cells is considered to be lower than that for engines or turbines..

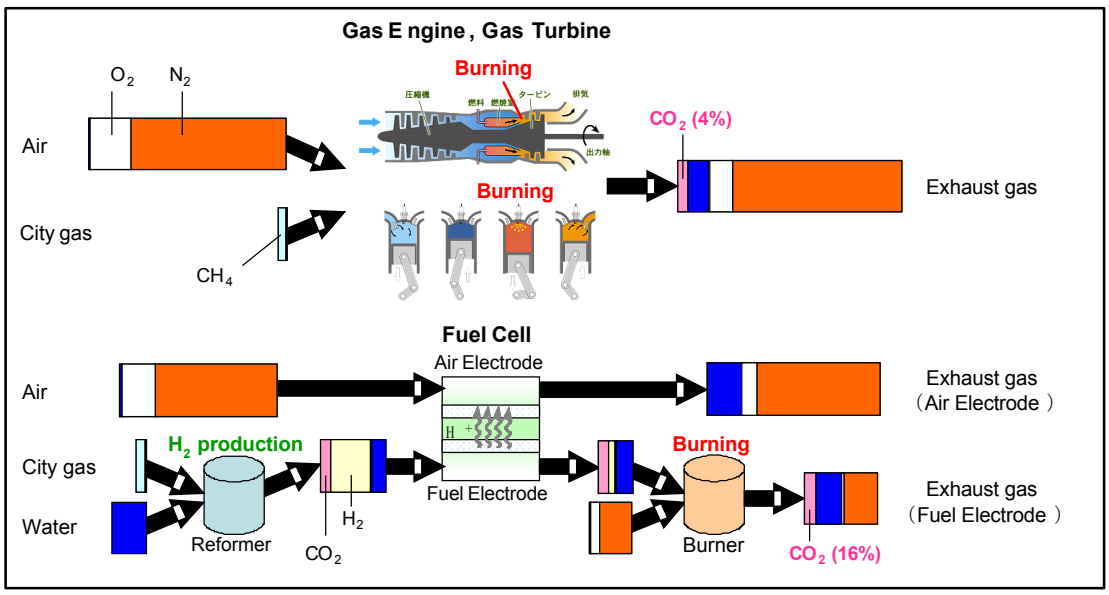

Figure 2. Difference in mechanisms for a fuel cell and internal combustor.

PAFC systems have been commercialized and put into practical use by Fuji Electric Co., Ltd. Tokyo Gas and Fuji Electric developed a PAFC system with CO_2 separation equipment. The

developed PAFC system is considered to be commercial-ready and is expected to contribute to drastic CO_2 emission reduction.

CO_2 is recovered from the exhaust gas of the reformer in the PAFC, where CO_2 comprises approximately 25% of the gas. Figure 3 is the system flowchart of the CO_2 recovery equipment. Using this system, approximately 70% of CO_2 emissions from the PAFC can be recovered and easily transported as liquefied CO_2 in bottle containers.

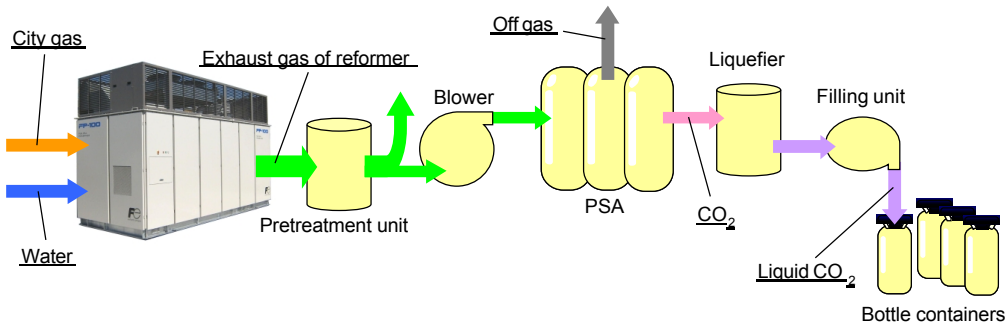

Figure 3. System flowchart of the CO_2 recovery equipment in the PAFC system.

Table 1 shows the specifications of the newly developed PAFC system with the CO_2 separation unit. Although output and power efficiency are reduced in the developed system, the emitted CO_2 is successfully recovered. By using combined heat and power (CHP), the newly developed PAFC system can be a substitute for boilers. The newly developed PAFC system is being regarded as virtually CO_2-free on the assumption that not only the recovered CO_2 but also originally emitted CO_2 from the boilers are included in the amount of the total CO_2 reduction.

Table 1. Specifications of the conventional and developed PAFC systems.

	Conventional PAFC system	Developed PAFC system
Power transmission end output [kW]	100	72
Power generation efficiency [%] (transmission end)	40	29
Heat recovery efficiency [%]	49	48
Total energy efficiency [%]	89	77
Amount of CO_2 emitted [kg-CO_2/h]	51.4	15.4
CO_2 emission intensity [kg-CO_2/kWh]	0.51	0.21
CO_2 emission intensity by using CHP [kg-CO_2/kWh]	0.34	−0.01

Using this system, approximately 70% of CO_2 emissions from PAFCs can be recovered and easily transported from the site as liquefied CO_2 in bottle containers. Although total energy efficiency is reduced by around 13% in the developed system, around 70% of the emitted CO_2 is successfully recovered.

Utilization of CO_2 captured from a hydrogen station

Tokyo Gas built a hydrogen station in December 2010[7] and has been operating it under contract with Hydrogen Supply/Utilization Technology (HySUT) within the context of the Demonstration Program for Establishing a Hydrogen-based Social System, sponsored by the Ministry of Economy, Trade and Industry of Japan[8]. Figures 4 and 5 show an image and the system flowchart, respectively, of the new hydrogen station. Natural gas is reformed to hydrogen at the station, and the CO_2 generated during this process is captured. For the first time in history, CO_2 has been captured at a hydrogen station during hydrogen production. The station has commenced the refuelling of fuel-cell vehicles and airport limousines. Approximately 80 kg/day of liquefied CO_2 is captured during the demonstrational operation of the station.

Figure 4. Image of the newly built hydrogen station with CO_2 capture equipment.

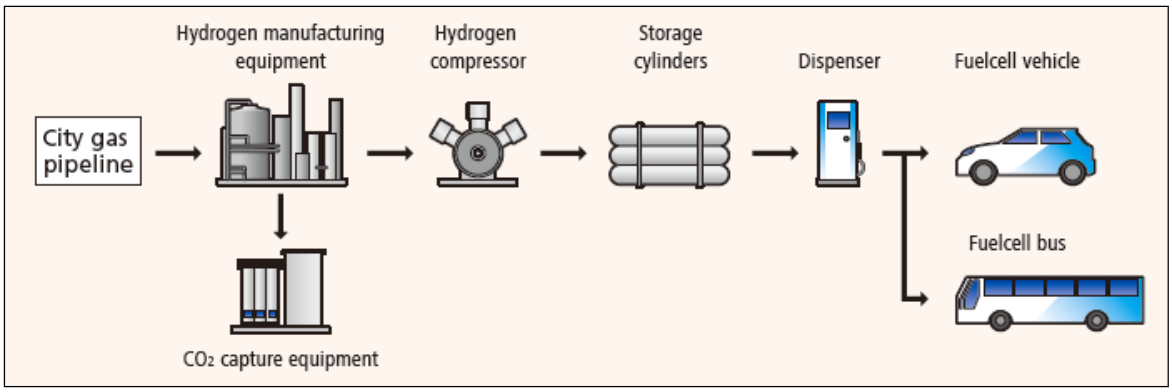

Figure 5. System flowchart of the hydrogen station with CO_2 capture equipment.

The liquefied CO_2 captured from the station is transported by truck and utilized for enhance the growth of tomatoes on in a plant factory the greenhouse at Chiba University. A schematic flow of CO_2 utilization on in the plant factory greenhouse is shown in Figure 6. The supplied amount of the CO_2 is approximately 320 kg/month. The site area of the greenhouse is approximately 1000 m^3. The CO_2 concentration inside the greenhouse is maintained around 1000 ppm as compared the density level of CO_2 in the atmosphere, which is approximately 400 ppm. Photosynthesis in the tomatoes is enhanced, and the yield and sugar content of the

tomatoes are expected to be improved by the increased CO_2 uptake.

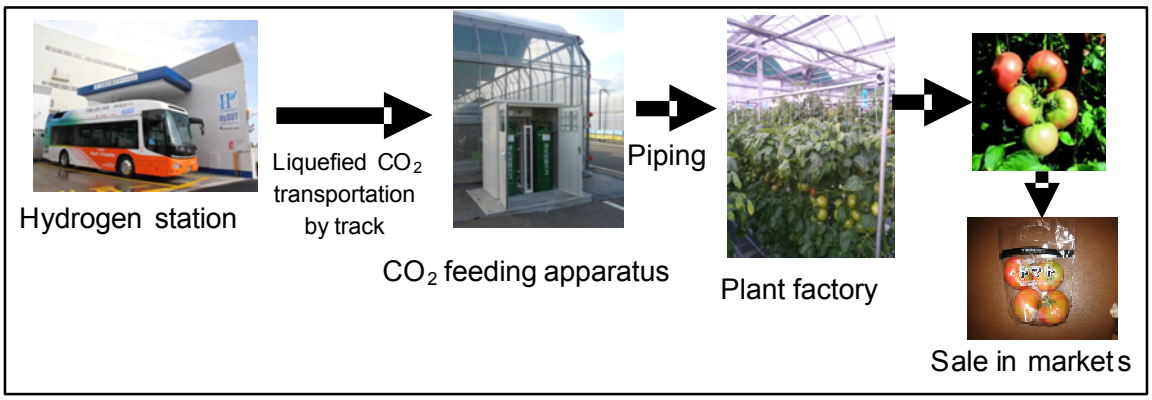

Figure 6. Schematic flow of CO_2 utilization for tomato plant factory.

Development of storage technology for CO_2 from distributed energy sources

In Japan, sites with a CO_2 sequestration capacity of more than 1×10^6 t/yr are limited because the geological structures are often complicated by alternating layers of sand and clay that limit the expansion of the saline aquifers. For CO_2 sequestration at medium- or small-scale sites, Tokyo Gas and RITE are jointly developing technology for microbubble injection into saline aquifers. A microbubble is generally defined as a bubble with a size of 1~100 μm. Microbubbles have several unique features. They move very slowly in water, and the surface area of a swarm of microbubbles is much greater than that of a single bubble containing the same volume. Consequently, CO_2 microbubbles have a much larger area of contact with the saline water in the aquifer, and the time required for a microbubble to move from the injection points to the cap rocks can be prolonged. The injected CO_2 microbubbles are expected to dissolve immediately 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 sequestration in a saline aquifer.

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

Several methods have been proposed for the generation of microbubbles. In many cases, the generated microbubbles are intended to be utilized under atmospheric pressure conditions such as in water treatment, cleaning, and medical applications. However, for utilization in CCS, the pressure of the CO_2 at the 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 conditions.

Laboratory experiments were performed using an apparatus that simulates the pressure and temperature conditions in a deep aquifer. Figures 7 and 8 show a photograph and a

schematic, respectively, of the experimental apparatus. 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 using a syringe pumps; 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 was 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.

Figure 7. Photograph of experimental apparatus.

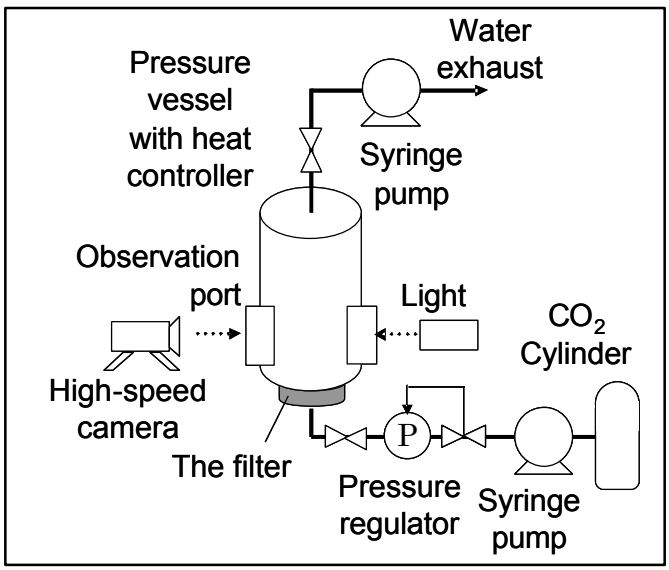

Figure 8. Schematic of experimental apparatus.

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

software.

CO_2 microbubble injection was carried out at 40 °C and 10 MPa, which simulates the conditions in a saline aquifer at a depth of 1000 m. Figure 9 shows the behaviour 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 upward 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 stored efficiently and safely by microbubble injection.

Figure 9. Behavior of injected CO_2 microbubbles (40 °C, 10 MPa).

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 10 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 compared to that of the large bubble. The ratio of the decreased volume to the original volume of the microbubble swarm (decreased volume of bubbles/original volume 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 demonstrates increased efficiency of CO_2 microbubble injection in saline aquifers.

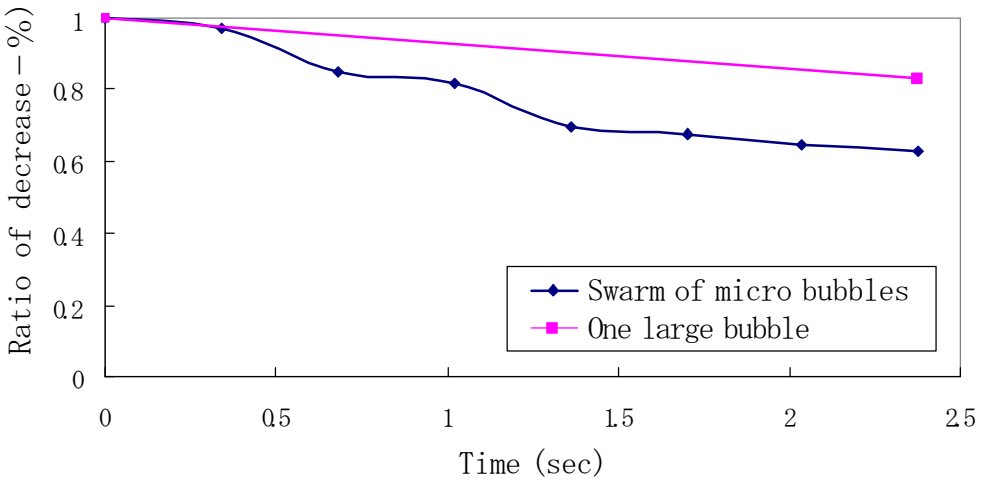

Figure 10. Comparison of decrease in volumes of microbubbles and large bubble over time.

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. A detailed image of the sinking behaviour of microbubbles is shown in Figure 11. 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 this counter-intuitive sedimentation phenomenon was observed. The CO_2 microbubbles dissolved rapidly in the saline water as soon as the CO_2 was injected. The partial density of water increases rapidly upon CO_2 dissolution. A number of microbubbles are sunk by the downward flow of the 'heavyish' water. This sedimentation phenomenon is considered to contribute to increased safety of the CO_2 injection into saline aquifers because the sinking CO_2 bubbles stay near the injection point.

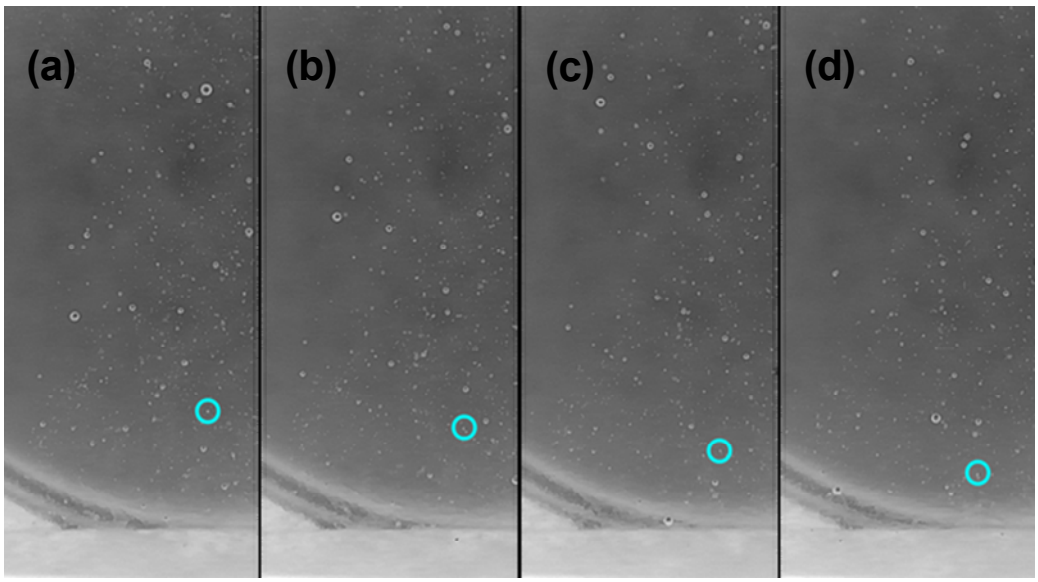

Figure 11. Sedimentation phenomenon of injected microbubbles.
(a) Initial state of phenomenon, (b) 0.1 s, (c) 0.2 s, and (d) 0.3 s.
The red circles indicate movement of a microbubble.

CO_2 microbubbles were successfully generated in laboratory experiments under simulated conditions similar to those in a deep saline aquifer, owing to the fine pores and the uniform pore size of the developed filter. 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 which should contribute to the safety of this CO_2 injection technology for saline aquifers.

Conclusion and future issues

Both near-to-mid-term and mid-to-long-term countermeasures for reducing CO_2 emission levels in distributed energy systems have been suggested. As a near-to-mid-term solution, CO_2 capture and utilization technologies in distributed energy systems have been developed to reach CCS-ready status. CO_2 recovery equipment was developed for actual distributed energy systems such as a 100-kW-class PAFC and a hydrogen station. Significant value is considered to be added to CCS by distribution of captured CO_2 as a valuable substance to a CO_2 utilization facility such as greenhouse. To increase the usage of CO_2 from CCS beyond greenhouses, new methods of CO_2 utilization, such as for dry ice and as a raw material for chemical production, should be developed.

With a long-term view beyond 2020, development was initiated for storage of captured CO_2 from distributed energy sources. The microbubble injection method can be employed as a CCS technology for realizing efficient and safe CO_2 sequestration in saline aquifers. This should be especially effective in the case of CO_2 captured from distributed energy sources and sequestered in small- or medium-scale storage sites, which have complicated geological structures. The behaviour of the injected CO_2 microbubbles should be investigated using typical monitoring methodology, for example, by performing resistivity and elastic wave measurements. Research and development of these monitoring technologies are required. For the feasibility of CCS technology, it is important for the gas industry to start developments and demonstrations at the present.

CO_2 transportation technology is also important. After 2020, the technological perspective of CO_2 pipelines will be critical for the realization of CCS. Further research on CO_2 pipelines, including issues like durability and safety, is required.

Acknowledgments

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