Microbubble carbon dioxide injection for enhanced dissolution in geological sequestration and improved oil recovery

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Abstract

Microbubble CO2 can be generated by injecting CO2 through special porous filters attached to borehole casing or gas tubing. When injecting microbubble CO2 into saline aquifers, dissolution of injected CO2 into formation water can be accelerated up more than 20%, compared to conventional CO2 injection. As a result, microbubble CO2 injection will minimize the free CO2 fraction in the subsurface and consequently contribute to the long term safety of large-scale CO2 storage. Microbubble CO2 injection will also lead to effective use of pore space within the reservoirs. P-wave velocity and resistivity changes obtained when injecting CO2 in microbubble and normal bubble sizes into artificial brine-saturated porous sandstones indicate more pore water displaced by the injected microbubble CO2 in terms of sweep efficiency. Combined effects of enhanced dissolution and sweep efficiency in microbubble CO2 injection can reduce the potential risk of CO2 leakage from the subsurface and enable us to store more CO2 in same reservoirs compared to normal CO2 injection as well as economic benefit of achieving higher oil recovery.

Keywords: CO2 microbubble; X ray CT; enhanced dissolution; sweep efficiency; mobility; oil recovery

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1. Introduction

Microbubble technology has a long history in water and wastewater treatments, to remove particles from water in the air flotation process [1]. Microbubbles with diameters on the order of 10 μm have many different characteristics resulted from their small size [2]: (1) high specific interfacial area, for a given gas holdup the decrease of bubble size obviously results in the increase of specific interfacial area; (2) slow rise velocity, after formation a bubble rapidly accelerates to its terminal velocity which is proportional to the square of bubble diameter; (3) high inner pressure, gas pressure inside a bubble is greater than that of outside due to the surface tension. With the recent discovery of its bioactivity in fish culture and water purification, the microbubble is attracting more attentions due to the unique characteristics, such as slow rise velocity and negative zeta-potential [3, 4].

Microbubbles are often generated by pressurization, cavitation and rotating-flow, to overcome the difficulty in preventing bubble coalescence when generating microbubbles using porous media, constant flow nozzles, membrane and gas spargers with mixers [3]. However, applications of such methods are restricted to low pressure (<1 MPa) and ambient temperature conditions. Koide and Xue [5] proposed a novel concept to inject microbubble CO₂ from the small- to middle-scale emission sources (< 100 kiloton/year) into widely existing monoclinic reservoirs in Japan. Injection CO₂ in microbubble can be achieved by installing special porous filters on borehole casing or gas tubing [6].

Compared to the conventional microbubble generation methods listed in Table 1, this new method also using porous media but easy to maintenance and less operation expense. The most important points are: it can be easily deployed to deep formations (high pressure and temperature) and has advantages in preventing corrosion in saline water. Xue et al. [7] successfully generated microbubble CO₂ in gaseous, liquid and supercritical phases by using the special porous filters. They also showed the enhanced effect for dissolution of CO₂ microbubble. From the quantitative analysis with images recorded by a high-speed digital video camera system, they concluded that the enhanced effect of microbubble was higher than 20% (Figure 1).

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<th>Microbubble generation methods</th>
<th>Enhanced processes will take place in the same manner as we inject CO₂ microbubble into saline aquifer and oil reservoirs. To improve understanding of enhanced processes in deep reservoirs, we also compared the difference of CO₂ saturation estimated from CT values obtained when injecting normal bubble (NB) and microbubble CO₂ (MB) into the artificial brine-saturated Berea sandstone (Fig.2).</th>
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Fig. 1. Comparison of dissolution rate between normal and microbubbles. Upper: Shrinking of a swarm of microbubbles with diameter ranged 200 - 50 μm at a time step of 0.34 sec.; Lower: Ratio of decrease of bubble volumes against observation time.

Fig. 2. Comparison of CO₂ saturation when injecting normal and microbubble CO₂ in Berea sandstone.
2. Experimental Procedure and Visualization with X-ray CT Scanner

2.1. Sample preparation

In our previous study on effects of P-wave velocity and resistivity changes during MB-CO₂ and NB-CO₂ injection, we already confirmed the advantages of MB-CO₂ injection in use of pore space in Berea sandstones. To assist interpretation of experimental results, we used X-ray CT to visualize CO₂ migration in pore spaces by injecting CO₂ into the same sandstone sample through the special porous filter (MB-CO₂) and the grooved disc with small holes (NB-CO₂). In this paper we present the advantages of CO₂ microbubble in both saline aquifer storage (two-phase) and enhanced oil recovery (three-phase) from low-production and depleted oil reservoirs. Cylindrical Berea sandstone samples (35 mm in diameter and 70 mm in length, 18% porosity) were used in this study.

The whole sample assembly was covered by silicon sealant to prevent the leakage of CO₂ from the sample and invasion of hydrostatic pressure medium (oil) into the sample. After setting the sample into the pressure vessel, it was pressurized up to 13MPa under hydrostatic pressure conditions. Then the sample was saturated with artificial formation water (0.01Mole, KCL solution, 1ohm-m) in 10MPa. Supercritical CO₂ was injected into the water-saturated sandstone at 10.5MPa, 40 °C and 0.05ml/min. The hydrostatic pressure and pore pressure were kept constant throughout the CO₂ injection test.

In this study we used the Toshiba Aquilion One X-CT Scanner with a resolution of 0.35mm x 0.35mm, to monitor and evaluate changes in the fluid saturations in Berea sandstone. Figure 3 shows the sample prepared for the X-CT visualization test. The grooved disc was set to left end and the special filter was set to right end of the sample. The assembled sample was set into a special pressure vessel designed for X-CT scanner. CO₂ was injected first through the grooved disc from the left end of the sample and then the sample was flushed with artificial formation water to remove CO₂ in pore spaces until the CT value recovered to the initial saturated condition. Then CO₂ was injected through the special filter from right end of the sample. X-CT images were recorded during the two injection tests, to investigate advantages of CO₂ microbubble injection.

3. Results and Discussion

3.1. Results of the two-phase (brine and CO₂) experiments

P-wave velocity will decrease and resistivity will increase as a result of CO₂ and water displacement in pore spaces in water-saturated sandstones. Usually changes in P-wave velocity and resistivity depend on CO₂ flooding...
Higher sweep efficiency causes large changes in P-wave velocity and resistivity [7]. From the two-phase experiments we confirmed that: (1) Breakthrough time of MB-CO$_2$ injection is almost double compared to that in NB-CO$_2$ injection. CO$_2$ injected through the grooved disc migrated at a steady pace in NB-CO$_2$ injection and this result suggests once a flow path formed, the path will dominate the CO$_2$ flow in this sample (Figure 4). Because the CO$_2$ does not penetrate into other pore spaces, the sweep efficiency would be low and ultimately cause an early breakthrough of injected CO$_2$. (2) Increase in resistivity during MB-CO$_2$ injection is greater than that in NB-CO$_2$ injection. These results suggest that CO$_2$ injection through the special filter leads to better sweep efficiency in MB-CO$_2$ injection.

Fig 4. Changes of mean CO$_2$ saturation (left: NB-CO$_2$ injection; right: MB-CO$_2$ injection).

Fig 5. CO$_2$ saturation difference within low permeable zones in Berea sandstone during NB- and MB-CO$_2$ injections.
3.2. Results of the three-phase (brine, oil and CO₂) experiments

The two-phase experiments strongly indicate the higher sweep efficiency during MB-CO₂ injection. If the same mechanism works well in three-phase experiments, there is a big potential to recovery oil from the reservoirs. Xue et al. [8] used the experimental system to investigate effects of NB- and MB-CO₂ into brine and oil saturated Berea sandstones. The samples were firstly saturated with artificial brine with the similar process in two-phase experiments and then flooded with oil (decane) until no displaced brine recovered from the samples (irreducible water saturation). Figure 6 shows migrations and distributions of injected NB- and MB-CO₂ in the Berea sandstone samples.

Fig 6. Schematic view of the sample assembly for X-CT visualization tests.
Figure 6 shows the differences of CO\textsubscript{2} distribution (warm color area) along the sample length between NB- and MB-CO\textsubscript{2} into the sample. During the NB-CO\textsubscript{2} injection the CO\textsubscript{2} mostly flooded the upper part of the sample, while the injected CO\textsubscript{2} flooded both upper and lower parts during the MB-CO\textsubscript{2} injection. It is worth to noting that such differences appeared at the same pressure and temperature condition, and the same CO\textsubscript{2} injection rate. In addition, there is less difference in porosity and permeability between the two Berea sandstone samples. They were drilled from the same block and a few thin layers with higher permeability are aligned longitudinally of the drilled samples. The NB- and MB-CO\textsubscript{2} were injected perpendicular to such high permeable layers.

As mentioned above that NB-CO\textsubscript{2} has greater buoyancy than MB-CO\textsubscript{2} due to the difference of bubble size. The higher upward buoyant force works well in the porous sandstone sample and this is the main reason why we observed the early breakthrough when injecting NB-CO\textsubscript{2}. Once the CO\textsubscript{2} breakthrough occurs at the opposite end of the sample, the path will dominate the flow of injected CO\textsubscript{2} and consequently leads low sweep efficiency. This explanation is strongly supported by the experimental result in Figure 4. When injecting CO\textsubscript{2} through the special filter the MB-CO\textsubscript{2} flooded not only the high permeable upper part but also the bottom and slightly low permeable middle parts. Lower buoyant force and better ability to penetrate small or narrow pores of MB-CO\textsubscript{2} lead the late CO\textsubscript{2} breakthrough and higher sweep efficiency (Figure 7).

The late breakthrough also depends on CO\textsubscript{2} dissolution. Microbubble CO\textsubscript{2} dissolution can be clearly confirmed in case of 0.045 PV in Figure 7. It is interesting that both NB- and MB-CO\textsubscript{2} flows appeared in the high permeable layers but MB-CO\textsubscript{2} can migrate into the vicinity of the high permeable layers. These results suggest the usefulness of CO\textsubscript{2} microbubble injection in enhanced oil recovery.

![Fig 7. CO\textsubscript{2} distribution in two-phase experiments during NB- (upper) and MB-CO\textsubscript{2} (lower) injections.](image)

4. Conclusions

We examined the microbubble CO\textsubscript{2} injection into saline aquifers can accelerate CO\textsubscript{2} dissolution and minimize the free CO\textsubscript{2} in the subsurface and contribute to the long-term safety of large-scale geological storage. The enhanced dissolution of the microbubble CO\textsubscript{2} is higher than 20%, based on the quantitative analysis with images recorded by a
high-speed digital video camera system. Mito and Xue [9] reported the field survey results on CO\textsubscript{2} dissolution in formation water from the first Japanese pilot CO\textsubscript{2} injection site (Nagaoka) and they also pointed out the potential that CO\textsubscript{2} will be converted into stable carbonate minerals as a result of geochemical reactions. The practical use of the microbubble technology is to install the special filter directly to the gas tubing or steel casing in injection wells. In the near future CO\textsubscript{2} microbubble sequestration would be a novel technology to store CO\textsubscript{2} from the small- to middle-scale emission sources such as oil refinery and hydrogen filling station.

Results from three-phase experiments suggest that normal bubble CO\textsubscript{2} has greater buoyancy than microbubble CO\textsubscript{2} due to the small size. The grate upward buoyant force resulted in an early breakthrough as the NB-CO\textsubscript{2} only flooded the high permeable part of the sample. The MB-CO\textsubscript{2} can flood almost whole sample and advantages in the effective use of pore space indicate the higher sweep efficiency of microbubble CO\textsubscript{2} injection and this technology will contribute to the enhanced oil recovery from the low permeable and depleted reservoirs.

Acknowledgements

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References

[8] Xue Z., Akai T., Kiyama T. and Nishizawa O., Experimental study on microbubble CO\textsubscript{2} in enhanced oil recovery, the fall meeting of the Mining and Materials Processing Institute of Japan (in Japanese), 2014, Kumamoto Japan.