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Multi-scale experimental study of carbonated water injection: An effective process for mobilization and recovery of trapped oil

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highlights

- Gas exsolution during carbonated water injection (CWI) is studied.

- Experiments are performed at macro and micro scales using X-ray CT imaging.

- Oil recovery factor increases significantly due to gas exsolution during CWI.

- CWI has applications in both environmental engineering and petroleum engineering.

 \bullet CWI offers a possibility for CO₂ sequestration in petroleum oil reservoirs.

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ABSTRACT

Steady flow of a disconnected gas phase (bubbles) is realized in porous media during carbonated water injection (CWI) under conditions that promote continuous exsolution of the dissolved gas. Using microfluidic pore networks etched on glass as well as a miniature core-flooding setup integrated with micro computed tomography (CT) imaging apparatus, we demonstrate capillary interactions of the flowing gas bubbles with a previously trapped oil phase (three-phase ganglion dynamics), which lead to mobilization of oil ganglia and remarkably high oil recovery. When three-phase ganglion dynamics are induced by carbonated water injection in low-permeability Berea sandstone core samples containing waterflood residual oil, more than 34% and 40% of the original oil in place additional recoveries are achieved in macro- and micro-scale flow tests, respectively, while a significant amount of $CO₂$ is permanently sequestered in the pore space as capillary-trapped and dissolved gas. It is observed that when oil globules come into contact with $CO₂$, they form thick spreading layers between brine and gas and are carried by moving gas clusters. The oil layers stay stable until the gas clusters leave the medium. Individual oil and gas blobs captured during micro-CT imaging are statistically analyzed to further examine underlying pore-level displacement physics of the process.

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1. Introduction

Sustained production of oil from a subsurface reservoir, initially driven by natural formation pressure and expansion of oil and reservoir rock, requires injection of water or gas in order to maintain reservoir pressure and displace oil toward production wells [\[1\]](#page--1-0). In the majority of cases, this so-called secondary recovery is achieved by waterflooding, a technique first considered in 1880 [\[2\]](#page--1-0). Waterflooding is a process of immiscible displacement leaving behind a significant amount of the original oil in place (OOIP), typically 30–50% in water-wet systems, in the form of disconnected oil

⇑ Corresponding author. E-mail address: mpiri@uwyo.edu (M. Piri). ganglia strongly held within rock pores by capillary forces [\[3\]](#page--1-0). Of various enhanced oil recovery techniques targeting this trapped oil, injection of $CO₂$ [\[4,5\]](#page--1-0) has recently received renewed attention. $CO₂$ is also a greenhouse gas that may be captured and sequestered $[6]$. Recent estimates indicate significant potential for $CO₂$ -based EOR for recovery of large volumes of incremental oil from watered-out reservoirs alongside storage of equally large volumes of $CO₂$ [\[7\]](#page--1-0).

The current paradigm of large-scale $CO₂$ sequestration is injection of $CO₂$ as a separate, usually supercritical, phase. The same is true of $CO₂$ injection for enhanced oil recovery. Recently, however, attention has been drawn to the alternative of dissolving the $CO₂$ in brine at the surface and injecting carbonated brine instead of $CO₂$ $[8,9]$ – an approach that would mitigate the risks associated with

buoyant migration of a free $CO₂$ phase since $CO₂$ is dissolved in brine and carbonated brine is denser than the native brine. Carbonated water injection (CWI) for oil recovery at the secondary and tertiary (EOR) stages was proposed more than fifty years ago [\[10\]](#page--1-0). Interest in this process has been recently renewed [\[11\]](#page--1-0) given the potential for coupling oil recovery with $CO₂$ sequestration. Previous pore-scale visualization studies in glass micromodels [\[12\]](#page--1-0) have shown that transfer of dissolved $CO₂$ from carbonated brine to oil causes swelling and coalescence of isolated oil ganglia, and that oil redistribution leads to local flow diversion. In combination with a reduction in the oil viscosity, these processes instigate displacement of additional oil. Sohrabi et al. [\[11\]](#page--1-0) reported 10% additional recovery of the original oil in place by CWI in a previously waterflooded high-permeability core sample, while nearly half of the injected $CO₂$ was permanently stored. This was achieved by direct transfer of $CO₂$ from brine to oil without appearance of a free $CO₂$ phase. In other words, the impact of $CO₂$ exsolution during CWI and the effects of free gas on the mobilization of oil ganglia were overlooked. It was concluded that the success of CWI as an enhanced oil recovery method depends on deliverability of the dissolved $CO₂$ to the trapped oil [\[12,11\].](#page--1-0) Alizadeh et al. [\[13\]](#page--1-0) presented the results of an experimental study where carbonated water injection and subsequent degassing and in situ development of a gas phase, as a result of pressure depletion, were used to mobilize and recover residual oil in Berea sandstone. It was reported that the gradual increase in the pressure drop led to liberation of gas from the aqueous phase, internal gas drive, mobilization of oil ganglia, and reduction of residual oil saturation. Parallel pore-scale visualization studies using transparent glass micromodels indicated that the effectiveness of the approach was linked to the interaction between a flowing, disconnected gas phase and oil ganglia (three-phase ganglion dynamics). In addition to oil recovery, the process offered the possibility for simultaneous $CO₂$ sequestration in the pore space as capillary-trapped and dissolved gas. The experiments were conducted at ambient temperature and relatively low pressure. Zuo and co-workers $[14-16]$, in a series of experimental studies, also investigated gas exsolution from carbonated water in micromodels and sandstone rocks but at elevated pressure and temperature conditions. It was observed that gas exsolution in water-filled pores resulted in water flow blockage, local water flow diversion into oil-filled elements, and mobilization of trapped oil. Additional oil recovery of 10% was obtained when this process was applied to a Berea core sample. The researchers mention that, under reservoir conditions, the exsolved gas phase has little or no mobility and its relative permeability remains very low, in the order of 10^{–5} to 10^{–3}, even at moderate gas saturations. Therefore, they conclude that, rather than flowing, the gas phase acts as a flow barrier (similar to a mobility control agent) and diverts water flow to oil-filled pores, thereby displacing oil. Expansion of gas bubbles was also considered to improve oil recovery by displacing trapped oil globules.

The simultaneous flow of fluid phases in porous media is of great interest not only in petroleum reservoir engineering but also in many other areas of science and technology, such as environmental engineering. In the environmental context, multiphase flow occurs when petroleum products and volatile organic compounds, usually referred to as non-aqueous phase liquids (NAPLs), leak from storage tanks and migrate through the vadose zone toward the water table, acting as a source of groundwater contamination. Significant efforts have been devoted to developing clean-up methods capable of remediating contaminated zones [\[17,18\].](#page--1-0) Recently, carbonated water injection has also been employed to recover NAPL ganglia trapped below the water table. Nevertheless, the application is limited to only a few studies [\[19–21\]](#page--1-0), emphasizing the need for more detailed examination of the underlying physics.

Due to the applicability of CWI in environmental engineering and to understand the displacement physics under simplified conditions, we intend to investigate this process first at low pressure and temperature conditions prevailing at the groundwater table. However, we will address potential applications of the CWI process in petroleum oil reservoirs as well, even though the conditions relevant to petroleum engineering applications are the subject of our future studies. In this study, we focus on the impact of in situ degassing during carbonated water injection on oil recovery at conditions more relevant to the remediation of petroleum-contaminated zones. For this purpose, we deploy an array of experimental techniques to study this process and shed light on some of the subtle displacement physics. We use experiments in a two-dimensional micromodel as well as flow tests in naturally-occurring rocks with various sample sizes. For the latter, we utilize X-ray computed tomography (CT) techniques with significantly varying resolutions for in situ saturation measurements. We perform a set of proof-of-concept experiments on a large core sample (macro scale) to examine the effectiveness of CWI in recovery of trapped oil from a waterflooded core sample. We then conduct micro-scale flow tests (both in a small rock core sample and in a glass micromodel) to investigate the pore-level displacement physics responsible for the trends observed at the macro scale. We report pore-scale observations of three-phase flow involving two immiscible disconnected non-wetting phases (i.e., oil ganglia and gas bubbles) and one connected wetting phase (i.e., water). Steady flow of the discontinuous gas phase is established by carbonated water injection under conditions that promote in situ degassing [\[22,23\]](#page--1-0). This is a non-equilibrium process driven by supersaturation of the aqueous phase with $CO₂$, namely by the condition HC – $P_w > 0$, where C and P_w are the local $CO₂$ concentration and pressure of the aqueous phase, respectively, and H is Henry's constant. We demonstrate that oil ganglia are effectively and immiscibly mobilized by flowing gas bubbles. Additional recoveries of more than 34% and 40% of OOIP at the macro and micro scales, respectively, are achieved when this process is applied to low-permeability sandstone cores containing trapped oil due to a waterflood.

In the following sections, we first provide the specifics of the porous mediums and fluids used in the experiments at different scales. We then describe the details of experimental setups and procedures deployed to perform the flow tests. This is followed by the results of the experiments and discussion. We conclude the paper with a set of final remarks.

2. Flow experiments

Experiments presented in this paper were performed at two scales: macro (Category A) and micro (Category B). At the macro scale, we used a long consolidated naturally-occurring sandstone core sample to perform a set of core-flooding experiments in order to examine the effectiveness of the immiscible CWI (in situ degassing and consequent three-phase ganglion dynamics) in recovering trapped oil at two different carbonation pressures. We then used micro-scale studies to examine some of the complex pore-scale phenomena leading to the trends observed in the experiments under Category A. We performed miniature core-flooding experiments (Category B) in a similar, but smaller, core sample while using a micro-CT scanner to obtain high-resolution images of pore-scale fluid occupancies. We also investigated, still under Category B, more details of the pertinent displacement mechanisms using a two-dimensional glass micromodel.

Below, we present details of the porous mediums, fluids, and experimental conditions, setups, and procedures utilized in each of the above-mentioned categories of experiments.

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