International Journal of Greenhouse Gas Control 46 (2016) 175–186

Contents lists available at ScienceDirect

International Journal of Greenhouse Gas Control

journal homepage: www.elsevier.com/locate/ijggc

Impact of wettability alteration on 3D nonwetting phase trapping and transport

Anna L. Herring[a,b,*], Adrian Sheppard[b], Linnéa Andersson[a], Dorthe Wildenschild[a]

[a] School of Chemical, Biological and Environmental Engineering, Oregon State University, Corvallis, OR 97331, USA
[b] Department of Applied Mathematics, Research School of Physics and Engineering, Australian National University, Canberra, ACT 2601, Australia

ARTICLE INFO

Article history:
Received 15 April 2015
Received in revised form 29 October 2015
Accepted 15 December 2015
Available online 19 January 2016

Keywords:
Wettability
Drainage flow pattern
Capillary trapping
Residual trapping
Pore scale
Geologic CO_2 sequestration

ABSTRACT

We investigate capillary trapping and fluid migration via x-ray computed microtomography (x-ray CMT) of nonwetting phase (air) and wetting phase (brine) in Bentheimer sandstone cores which have been treated to exhibit different degrees of uniform wettability. x-Ray CMT scans were acquired at multiple steps during drainage and imbibition processes, as well as at the endpoints; allowing for assessment of the impact of wettability on nonwetting phase saturation and cluster size distribution, connectivity, topology and efficiency of trapping. Compared with untreated (water-wet) Bentheimer sandstone, cores treated with tetramethoxylsilane (TMS) were rendered weakly water-wet, and cores treated with octadecyltrichlorosilane (OTS) demonstrate intermediate-wet characteristics. As apparent contact angle increases, drainage flow patterns deviate from those derived for water-wet systems, total residual trapping and trapping efficiency decrease, and buoyancy plays a larger role during nonwetting phase mobilization; this has significant implications for CO_2 migration and trapping during CO_2 sequestration operations.

1. Introduction

Multiphase flow in porous media has long been a subject of intensive research, traditionally with application to oil extraction. More recently, geologic CO_2 sequestration has been suggested as a potential climate change mitigation strategy, which has initiated renewed research efforts into multiphase flow processes. While some basic principles of multiphase flow can be immediately transferred between the two applications, the fluid properties of CO_2 (particularly density and viscosity) are dramatically different to those of oil, necessitating additional investigation of CO_2 flow processes. This study uses ambient condition air as a proxy for CO_2 to investigate how the wettability state of the porous medium impacts fluid flow behavior, and highlights consequences of potential wettability alteration during CO_2 sequestration operations.

Geologic sequestration of CO_2 in subsurface reservoirs is a climate change mitigation strategy which prevents carbon emissions to the atmosphere. Sequestration involves capture of CO_2 from the effluent of a large power production point source, transport to the storage reservoir in a dense liquid state, and then injection into a subsurface storage reservoir. The CO_2 will warm to the temperature of the geologic formation, resulting in a phase change to a supercritical state (we refer to supercritical CO_2 as $scCO_2$). During injection, the CO_2 phase displaces the resident brine of the geologic formation, i.e. a brine drainage process. Subsequently, upward travel of the buoyant CO_2 plume allows for brine imbibition as the brine reenters the pore space. Accurate prediction of transport and trapping of subsurface CO_2 is necessary to ensure the safety, security, and effectiveness of a CO_2 sequestration operation (IPCC, 2005).

Multiple trapping mechanisms have been identified which prevent CO_2 migration and release at the surface (IPCC, 2005); one such mechanism is capillary (or residual) trapping, wherein CO_2 is held by capillary forces within the pore structure of the geologic matrix. Capillary trapping decreases the risk of inadvertent leakage of CO_2 due to cracks and faults (or other preferential pathways) of the geologic matrix; this mechanism also breaks up the injected CO_2 plume into small bubbles, increasing the surface area to volume ratio of the CO_2 and enhancing subsequent dissolution and precipitation reactions. Accordingly, understanding capillary trapping is critical for engineered geologic sequestration. The wettability state of a porous medium is a main factor influencing the strength of capillary forces (and thus, capillary trapping) during multiphase flow (Anderson, 1987b; Morrow, 1990; Iglauer et al., 2012; Wan et al., 2014).

---

[*] Corresponding author at: Department of Applied Mathematics, Research School of Physics & Engineering, The Australian National University, Garran Road, Building 58, Canberra, ACT 2601, Australia.
E-mail address: anna.herring@anu.edu.au (A.L. Herring).

http://dx.doi.org/10.1016/j.ijggc.2015.12.026
1750-5836/

Wettability is defined as the affinity of a solid surface for one fluid in the presence of other immiscible fluids. From a pore-scale perspective, wettability can be measured by the contact angle (θ) created by the intersection of the fluid–fluid and fluid–solid interfaces. As measured through the aqueous phase, a contact angle between 0° and 75° is considered water-wet, 75–105° is intermediate-wet, and measurements of 105–180° are oil/$scCO_2$-wet (Treiber and Owens, 1972; Anderson, 1987a). The wettability of a porous medium affects the distribution of wetting and nonwetting fluids within the pore structure and can have a dramatic effect on permeability and connectivity of fluids. Depending on the saturation state of the medium, nonwetting phase fluid clusters tend to occupy the larger pores, while wetting fluid will exist in thin layers throughout the smallest throats and within crevices and roughness on the surface of the medium (Blunt and Scher, 1995; Kumar et al., 2009). As fluids redistribute in a water-wet medium due to changes in saturation or capillary pressure, the nonwetting phase can become isolated and trapped via "snap-off" (Roof, 1970); this occurs during imbibition, when encroaching wetting fluid films surround a nonwetting fluid element in a pore body and the wetting fluid menisci touch as the pore throat constricts. This mechanism will be very different, and potentially completely absent if the system wettability is altered to be less strongly water-wet. Thus, wettability is a dominant factor controlling the occurrence of snap-off (as wetting films are only present in strongly-wetting systems), as well as the morphology and topology of fluid phases within the porous medium (Blunt, 1998); it is therefore crucial to understand the degree to which wettability will affect residual trapping of the nonwetting phase for application in CO_2 sequestration scenarios.

The wettability state of a CO_2–brine–rock system is dependent on complex geochemical, ionic, and physical chemistry interactions, and there is currently no real consensus regarding how interaction with $scCO_2$ will impact rock surfaces in geologic systems. Transition to intermediate wettability after exposure to $scCO_2$ and brine has been reported for shale minerals (Chiquet et al., 2005), silica micromodels (Kim et al., 2012), sand columns (Plug and Bruining, 2007), and sandstones (Iglauer et al., 2011; Akbarabadi and Piri, 2013; Berg et al., 2013). However, other studies investigating $scCO_2$ in Berea sandstone have reported relative permeability and capillary pressure-saturation curves similar to gas-phase fluids, indicative of a water-wet system (Pini and Benson, 2013), and significant residual trapping due to snap-off of $scCO_2$ during drainage-imbibition studies, also implying water-wet conditions (Pentland et al., 2011; Krevor et al., 2012; Pini and Benson, 2013). Recently, Kaveh et al. (2014) showed that, in contrast to pure quartz, transition of CO_2 from subcritical to supercritical has no effect on the wettability of the Bentheimer sandstone/water system (despite the 90–96% quartz composition of Bentheimer sandstone). In carbonate rocks, Andrew et al. (2014) measured a $scCO_2$–brine in situ average contact angle of approximately 45°. El-Maghraby and Blunt (2013) report that experiments with Indiana Limestone show increased levels of residual trapping of CO_2 under supercritical conditions as compared to experiments conducted at subcritical pressure; they deduce that geochemical reactions force the system to a more strongly water-wet state than the same system at lower pressure. Wan et al. (2014) provide an overview of studies demonstrating the wide variation of contact angles reported for $scCO_2$–mineral systems and conclude that several factors contribute to the variation in reported contact angles: (1) nominally similar substrates can have different surface properties (i.e. reactivity or roughness) either intrinsically or due to cleaning methods; (2) contaminants may adsorb onto substrate surfaces and are difficult to avoid; (3) dissolution of CO_2 droplets due to non-equilibrium conditions affect contact angle measurements; and (4) the CO_2-substrate contact time, flow process order (i.e. which fluid first contacts the substrate), and contact repetition may affect water film/CO_2 stability. Given the range of observed wetting behavior, it seems prudent to further explore how wettability alteration affects CO_2 migration and capillary trapping.

A number of previous studies have investigated the impact of wettability on nonwetting phase trapping from the perspective of oil recovery. A pattern of minimum residual oil saturation at intermediate rock wettability and increased residual oil saturation at strongly water-wet and strongly oil-wet conditions is established in a review by Anderson (1987) (which presents the experimental results of Lorenz et al. (1974), Kennedy et al. (1955), and Amott (1959)); as well as in more recent work by Morrow (1990). This pattern is due to a combination of several factors. Snap-off dominates in water-wet conditions, leading to high residual oil saturations. Under intermediate-wet conditions, both fluids can easily flow through small constrictions; snap-off is suppressed (Mahmud and Nguyen, 2006), and oil can remain connected via smaller pore throats and drain to low residual saturation. Under oil-wet conditions, snap-off with respect to the oil-phase does not occur, but oil resides in the small pore spaces and throats of the media, again resulting in relatively high residual oil saturations. Blunt (1998) performed numerical simulations which demonstrate a similar pattern to the experimental results described above, but also indicate reduced residual saturations in strongly oil-wet ($\theta > 135°$) media due to the formation of oil films which allow oil to drain to very low saturations during water flooding.

However, results derived from oil–brine systems are not directly applicable to $scCO_2$–brine systems due to differences in fluid properties between oil and $scCO_2$. The viscosity of $scCO_2$ is lower than that of brine, resulting in an inverted viscosity ratio as compared to oil–brine systems. Such systems for which the invading phase is less viscous than the defending phase (e.g. CO_2, during a CO_2 injection) are said to have an "unstable" viscosity ratio, as the invading phase penetrates the medium via long fingers; this is in comparison to a "stable" displacement, where the invading phase moves through the medium as a solid front (Lenormand et al., 1988). In addition, gravity-buoyancy forces are generally stronger for $scCO_2$–brine systems compared to oil–brine systems due to the relatively large density difference between $scCO_2$ and brine; these differences in fluid properties between scCO2–brine and oil–brine systems limit the applicability of oil recovery literature to $scCO_2$–brine systems. Drainage flow patterns (nonwetting fluid invading wetting fluid) have been studied in water-wet two dimensional (2D) systems for a wide range of viscosity ratios and capillary numbers (Lenormand et al., 1988; Zhang et al., 2011), as well as a function of gravitational force (Méheust et al., 2002; Chau and Or, 2006). However, similar flow regimes are not well-defined for systems of intermediate wettability, or for three dimensional (3D) systems. In one of only a few supercritical condition CO_2–brine studies, Chaudhary et al. (2013) measured residual saturation of $scCO_2$ in glass (water-wet) and teflon ($scCO_2$-wet) bead cores, and found that residual trapping was greatly reduced in the $scCO_2$-wet system; however, the bead systems used in that study had large pore spaces, the size of which impacts the relative importance of snap-off and fluid-solid adhesive forces, and thus may not be directly applicable to natural geologic systems.

The impact of wettability alteration on capillary trapping of $scCO_2$ during geologic sequestration operations is difficult to investigate due to the inherent difficulty involved in conducting and quantifying wettability during supercritical-condition experiments. In this work we use an ambient-condition system of air, brine, and Bentheimer sandstone. While absolute values of fluid properties (e.g. density, viscosity and interfacial tension with brine) differ widely between air and $scCO_2$, the ambient condition experimental system used in this study does approximate the capillary, gravitational, and viscous forces present in a $scCO_2$–brine–sandstone system (Herring et al., 2015). Further,