**Investigation of mechanisms of supercritical CO₂ trapping in deep saline reservoirs using
surrogate fluids at ambient laboratory conditions**

Luca Trevisan^{a,*}, Abdullah Cihan^b, Fritjof Fagerlund^c, Elif Agartan^a, Hiroko Mori^a, Jens T.
Birkholzer^b, Quanlin Zhou^b, and Tissa H. Illangasekare^a

^{a)} Center for Experimental Study of Subsurface Environmental Processes (CESEP), Colorado
School of Mines, Golden, CO 80401, USA

^{b)} Earth Sciences Division, Lawrence Berkeley National Laboratory, University of California,
Berkeley, CA 94720, USA

^{c)} Department of Earth Sciences, Uppsala University, Villavägen 16, 75236 Uppsala, Sweden

*Corresponding author:

1500 Illinois Street

Golden, CO 80401

T – +1-303-273-3483

luca.trevisan@gmail.com

Abstract

Geological storage of carbon dioxide relies on the effectiveness of immobilizing CO₂ in the pore space of deep geological formations through a number of trapping mechanisms that include capillary, dissolution, and mineral trapping. Improved fundamental understanding of these processes is expected to contribute towards better conceptual models, improved numerical models, more accurate assessment of storage capacities, and optimized placement strategies. However, studying these processes at a fundamental level is not feasible in field settings because fully characterizing the geologic variability at all relevant scales and making observations on the spatial and temporal distribution of the migration and trapping of supercritical CO₂ (scCO₂) is not practical. The specific goal of this study is to develop and implement an experimental method in intermediate scale test tanks under ambient laboratory conditions to make observations and generate data to improve the understanding of capillary trapping affected by fluid and formation properties. Since it is challenging to visualize multiphase flow processes occurring at high pressure conditions at the meter scale, a testing method was developed based on the use of surrogate test fluids to replace the scCO₂ and formation saline water. To set a foundation for extrapolating experimental results to the field, we chose a set of dimensionless groups that define the relative contributions of buoyancy, viscous, and capillary forces to the displacement behavior of immiscible fluids. The experiments were designed with the goal of understanding and accurately quantifying the immobilization of the scCO₂ analog in a homogeneous formation confined by a slightly dipping structural barrier. A set of three displacement experiments through unconsolidated sands with variable permeability was conducted in a quasi-two-dimensional flow cell to gain insight into the influence of buoyancy forces on the propagation of the displacing phase. This work takes advantage of laboratory

42 experiments at the intermediate scale to investigate gravitational and hysteresis effects on
43 entrapment of scCO₂ currents in brine-saturated reservoirs. Understanding these phenomena at a
44 fundamental level represents a critical step to improve injection strategies and to enhance
45 capillary trapping mechanisms.

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47 **Keywords:** Multidimensional experimental analysis; capillary trapping; dimensional analysis;
48 trapping models.

49

1. Introduction

Geological storage of carbon dioxide in deep, brine-bearing formations has been proposed as a strategy to stabilize, and eventually reduce, the net loading of CO₂ from large stationary sources to the atmosphere (IPCC, 2005; Orr, 2009; Pacala and Socolow, 2004). The preference in using deep saline formations for CO₂ storage is motivated by their extensive presence in the U.S. and around the world (U.S. Department of Energy, 2012). The assessment of the storage capacity and safety of a potential reservoir, as well as the delineation of a specific area of review, represent critical steps for site selection and risk analysis that must be conducted prior to the deployment of carbon capture and storage (CCS). In this context, modeling tools based on numerical methods or analytical solutions are generally used to predict shape and extent of the scCO₂ plume during and after the injection (Akervoll et al., 2009; Celia and Nordbotten, 2009; Doughty and Pruess, 2004; Kumar et al., 2005; Schnaar and Digiulio, 2009). However, the limited availability of field data generally hinders a direct evaluation of model predictions; this motivates the development of experimental analyses to gain insight into the migration and trapping mechanisms involved in such multiphase systems. Since vertical migration and horizontal spreading underneath a physical barrier are two important flow components during the injection and redistribution of the scCO₂ plume, one-dimensional core-flooding tests have some inherent limitations in representing both phenomena. For example, since the magnitude of capillary trapping is spatially varying even in macroscopically homogenous porous media (Cihan et al., 2014), there is a value in studying the effect of flow dimensionality on the distribution of residual saturation in controlled laboratory settings. A series of intermediate-scale laboratory experiments aimed at reproducing flow dynamics and displacement patterns comparable to actual carbon storage scenarios were conducted. We use the term “intermediate-scale” to

describe the scale of our experiments based on the definition of Lenhard et al. (1995), according to which: 1) “the experimental configuration has to allow small-scale processes to manifest themselves at a larger scale so that their relative contributions to flow and transport phenomena can be studied and quantified”, 2) “the size of the experiment has to be small enough for the environment to be controlled”, and 3) “the dimensions of the flow-cell have to be compatible with measurement techniques”. Following the work of Cinar et al. (2009), we first selected a combination of immiscible fluids with comparable density and viscosity ratios to the real reservoir fluids. Afterwards, in order to replicate the vertical cross-sectional displacement of brine by scCO₂ that can be expected from an array of wells (Szulczewski et al., 2012), we performed immiscible displacements with the goal of characterizing the residual saturation of the invading fluid once a secondary imbibition had occurred. The outcomes of our experiments provide insight into the expected behavior of a buoyancy-driven plume migrating under an inclined impermeable layer under capillary-dominated flow and the related capillary trapping efficiency.

Besides the structural containment provided by a confining impervious layer, three major trapping mechanisms may contribute to avoid long-term migration of a continuous phase scCO₂ plume. These mechanisms are referred to as residual, dissolution and mineral trapping. Each of these mechanisms is relevant on different temporal scales and their contribution to the total entrapped volume will depend on the interplay of forces acting on the system during the injection and post-injection stages (Kopp et al., 2009). In this investigation, we define this competition of forces through dimensionless groups, which are used to design controlled laboratory experiments aimed to reproduce scCO₂ injection and forced imbibition processes in a synthetic aquifer under ambient conditions. Since the mechanisms that are known to immobilize the moving scCO₂

plume occur on a wide range of time scales (from decades to millennia) without much temporal overlap, we base our experimental analysis on the assumption that they can be studied separately (Celia and Nordbotten, 2009). Moreover, given the immediacy of trapping due to favorable stratigraphy (Bradshaw et al., 2007) as well as the significant volumetric contribution of residual trapping (Kumar et al., 2005), for the purpose of this study we only take into account *structural* and *residual trapping* controlled by capillary forces. The first trapping mechanism results from a capillary barrier effect and is a function of caprock integrity and spatial continuity, being known to contrast the upward migration of scCO₂ and representing a requirement for candidate repositories. The latter is known to affect the “footprint” of the plume, where the pore space invaded by scCO₂ during the injection stage is subsequently imbibed by the resident brine, affecting mostly the trailing edge of the buoyant plume (Juanes et al., 2006).

1.1 Residual trapping mechanism

During the injection period of a typical storage scenario, the pressure of the CO₂ pumped into the reservoir is increased until the resident brine is displaced from the pore space, invading first the larger voids and gradually increasing the saturation of the plume. Once the injection ceases and the CO₂ pressure at the well suddenly drops, a redistribution of the fluids throughout the reservoir follows, producing a flow reversal in the saturation history and leading to an imbibition process. After the surrounding brine has displaced the mobile fraction of the plume from the pore space, isolated CO₂ droplets will be disconnected from the continuous phase, leading to its effective immobilization. The trapped fraction of CO₂ subsequent to the imbibition process is defined as residual saturation and is a function of the maximum CO₂ saturation, or initial saturation S_{nw}^i , reached at the time of flow reversal (Akbarabadi and Piri, 2013; Land, 1968; Pentland et al., 2010). This residual trapping mechanism, also referred to as capillary

trapping or relative permeability hysteresis trapping (Flett et al., 2004; Juanes et al., 2006; Suekane et al., 2008), is known to affect the flow of multiple fluid phases through porous media and it has been broadly studied in several areas of research, from soil physics, where the unsaturated zone above the water table represents a vital environment for plant roots, to the discipline of aquifer remediation and enhanced oil recovery (EOR), where capillary forces need to be overcome in order to clean up chemical spills or to increase production of hydrocarbons from reservoirs (Lake, 1989). In contrast, the ultimate goal of CCS is to maximize the storage capacity of deep geological formations by increasing CO₂ immobilization through several trapping mechanisms, including capillary trapping. Therefore, estimation of the residually trapped volume and saturation of CO₂ is fundamental for the assessment of trapping capacity of a potential reservoir (Bachu et al., 2007). Nevertheless, in situ measurements of this essential quantity are very challenging to obtain in the field, motivating this investigation in a dimensionally similar system under controlled laboratory settings.

1.2 *Dimensionless groups*

The application of dimensional analysis to characterize multiphase fluid flow is widely documented in the literature (Cinar et al., 2006; Gharbi et al., 1998; Kopp et al., 2009; Zhou et al., 1994). In the context of immiscible fluids affected by capillary, viscous, and buoyancy forces, it has been demonstrated experimentally and numerically that the volume of residually trapped non-wetting phase (CO₂ in the case of CCS, crude oil in the case of EOR) can be correlated to the magnitude of dimensionless groups such as capillary and Bond numbers (Chatzis and Morrow, 1984; Hilfer and Oren, 1996; Lake, 1989; Larson et al., 1981; Morrow et al., 1988). These parameters describe the relative influence of the governing forces on the flow regime and have two main advantages when applied to experimental studies: 1) they allow

comparing results obtained in a synthetic model with a geometrically similar prototype by maintaining the same dimensionless group's value (Bear, 1972) and 2) their use helps reducing the number of parameters that are needed to be considered in the problem statement, decreasing the experimental effort required (Shook et al., 1992). In addition to capillary and Bond numbers, expressed by the formulations adopted by Cinar et al. (2009) and Kuo and Benson (2013) among others, we consider the viscosity (μ_{nw}/μ_w) and density (ρ_{nw}/ρ_w) ratios between the non-wetting (subscript nw) and the wetting (subscript w) phases to compare the flow instability between scCO₂/brine and our fluid combination (see Table 1). The ratio between viscous and capillary forces is expressed by the capillary number (Ca), while the ratio between buoyancy and capillary forces is expressed by the Bond number (Bo).

$$Ca = \frac{U_i \mu_i}{\sigma} \quad (1)$$

$$Bo = \frac{\Delta \rho g k}{\sigma} \quad (2)$$

where U_i and μ_i are the Darcy velocity and viscosity of the injected fluid, respectively, σ and $\Delta \rho$ are the interfacial tension and density difference between the injected and displaced fluids, g is the gravitational constant, and k is the permeability of the porous medium.

1.3 Flow behavior of the injected plume

The driving forces that control the migration of the scCO₂ plume while injection is occurring are gravity, capillarity, and the imposed pressure differential, while the spreading of the plume during post-injection involves the static balance between capillary and buoyancy forces. In the presence of an inclined sealing formation, the dipping angle of the stratigraphic limit will play a key role (Gasda et al., 2006), as well as the direction and magnitude of the regional groundwater flow (Larkin, 2010). An optimal injection scheme for an efficient storage

scenario is the “inject low and let rise” method (Bryant et al., 2008). This method is intended to maximize the sweeping area of the plume and **minimize** the accumulation of **mobile CO₂ phase** under the caprock by releasing the supercritical fluid as deep as possible in the reservoir. Another injection strategy known from EOR operations is the “chase brine” scheme (Ide et al., 2007; Qi et al., 2009), which has the goal of maximizing the contact area between the injected CO₂ and the resident brine, and eventually promoting dissolution. As sketched in Fig. 1, once the injection of scCO₂ has ceased and the continuous phase plume (in dark gray) has reached the top of the storage formation, a fluid redistribution occurs, leaving behind a portion of the reservoir (in light gray) characterized by immobile scCO₂ at residual saturation S_{nw}^r . This volume is known as the plume footprint and covers a variety of residual saturations, ranging from very small fractions to maximum residual saturation $S_{nw}^{r,max}$. In particular, higher residual saturations are expected to be found close the injection well, whereas the front is going to be characterized by lower amounts of trapped CO₂. Furthermore, when a wetting phase flow comparable to a water drive in EOR operations is established upstream, any mobile fraction of the plume pooling underneath stratigraphic barriers will be eventually displaced updip. This process will affect some areas of the reservoir with a secondary drainage cycle, which will eventually lead to larger amounts of residually trapped CO₂.

[Figure 1 here]

With the assumption of dimensional similarity between the laboratory model and the field prototype, we aim to conduct immiscible displacement experiments to replicate injection and migration of scCO₂ in deep saline aquifers. The residual saturation of non-wetting phase (NWP)

resulting from different Bond number scenarios will be analyzed in detail and correlated with the maximum saturation achieved at flow reversal to evaluate existing trapping models.

2. Experimental approach and materials

Several investigators have conducted laboratory experiments to simulate the dynamics of scCO₂ migration and trapping in deep geological formations. The methods employed to achieve this can be grouped under: 1) use of surrogate fluids in two- and three-dimensional flow cells at ambient conditions (Cinar et al., 2009; Neufeld et al., 2010; Wang et al., 2010), and 2) replication of actual reservoir conditions by injecting scCO₂ in 1-D core flooding experiments (Al-Abri and Amin, 2010; Krevor et al., 2012; Uemura et al., 2011). The experiments presented in this paper belong to the first category, in realization of the difficulty of test design and plume visualization in actual reservoir conditions. We used a pair of immiscible fluids and laboratory test sands to recreate the interplay of capillary, viscous, and buoyancy forces that are expected to govern the migration and entrapment of scCO₂ in deep, saline aquifers. In order to investigate the influence of variable buoyancy over capillary forces, defined by the Bond number, on the plume migration and trapping, we carried out three geometrically similar experiments with laboratory sands of increasing permeability. For each Bond number scenario, an injection stage of NWP was performed with equivalent flow rates during 5.5 hours. Afterwards, the plume was left to redistribute for several hundred hours until a steady state of NWP saturation was observed via x-ray attenuation. Finally, a forced imbibition event was carried out by injecting the denser, more viscous wetting phase from the same well, displacing any mobile fraction of the non-wetting plume that had accumulated underneath the confining top.

2.1 Surrogate test fluids

The application of analog fluids at ambient conditions to mimic the behavior of scCO₂-brine system has been widely documented in the literature for the simulation of miscible displacement (DiCarlo et al., 2007), dissolution enhanced by convective mixing (Backhaus et al., 2011; Neufeld et al., 2010), and immiscible displacement (Cinar et al., 2009; Pentland et al., 2010). To our knowledge, experimental studies of displacement and trapping of scCO₂ under deep reservoir conditions ($T > 31.1^{\circ}\text{C}$, $P > 8\text{MPa}$) have not yet been reported for two-dimensional test systems. This study analyzes the usefulness of a set of immiscible surrogate fluids at ambient conditions ($T = 20^{\circ}\text{C}$, $P = 0.1\text{MPa}$) to determine the flow regime and trapping capacity associated with interactions of capillary, viscous, and buoyancy forces in a synthetic aquifer. For the experiments that are presented below, we selected a set of surrogate fluids that represent a pseudo-CO₂ phase displacing a pseudo-brine phase within a synthetic aquifer with gently dipping caprock. For this purpose, an aqueous solution of glycerol (80% w/w) was chosen as analog for brine. Its viscosity was measured at 20°C with an AR-G2 rheometer (TA instruments) and was found to be in good agreement with the empirical parameterization presented by Cheng (2008). The pseudo-CO₂ phase consisted of Soltrol 220 (Phillips 66), a low-toxicity, isoparaffinic solvent, widely used for experimental purposes (Fagerlund et al., 2007; Schroth et al., 1998). In order to improve visual observation of the injected NWP and to increase the x-ray attenuation contrast with the wetting phase, Soltrol 220 was dyed with Sudan IV (Fisher Scientific) and doped with 10% w/w Iodoheptane (Alfa Aesar). The interfacial tension (IFT) between the two phases was measured with a CAM200 optical tensiometer (KSV instruments) at 20°C . The mass proportion of the glycerol-water mixture was 80:20 in order to obtain density and viscosity ratios between the two fluids that are in agreement with the parameter range observed in field settings (Table 1). For the case presented here, the density and viscosity ratios of the laboratory fluids can

be compared to a storage scenario in a ‘cold’ basin (geothermal gradient equal to 25°C/km and average surface temperature around 10°C) and injection into a rather ‘shallow’ reservoir (Nordbotten et al., 2005). At the Sleipner field in the North Sea, where scCO₂ is injected at 1012 meters below m.s.l., temperature and pressure conditions are estimated to be approximately 41°C and between 8 and 11 MPa, respectively (Arts et al., 2008). Under these conditions, viscosity and density ratios between injected and resident phases are 0.075 and 0.745 (using values from Singh et al. (2010)), respectively.

[Table 1 here]

2.2 *Experimental setup*

The laboratory experiments were carried out in a rectangular quasi-two-dimensional flow cell with internal dimensions (LxWxH) of 91.4 x 5.6 x 61 cm³. Although the flow cell has three dimensions, due to the reduced width of the tank compared to its length, the flow component along the second horizontal direction can be discounted and the flow can be considered two-dimensional. A schematic drawing of the experimental setup is given in Fig. 2. The acrylic walls of the flow cell allowed for visual inspection of the migrating plume, so that photographic images and saturation measurements via x-ray attenuation could be directly compared. The injection well was placed down dip on the left boundary of the aquifer and consisted of a vertical PVC pipe (internal diameter, I.D.=2.3 cm) screened along a 11 cm length. The screened area consisted of 30 holes with I.D.=0.4 cm for a total area of 3.8 cm², and was wrapped in a very fine stainless steel wire mesh (#250) to prevent clogging by the surrounding sand. During injection, Soltrol 220 was constantly supplied to the well with a fixed pressure by a Mariotte bottle placed

on an electronic scale positioned on a motorized elevator to control head. This allowed the head difference (ΔH) between the Mariotte bottle and the constant head reservoir mounted on the right hand boundary to be controlled. Packed with coarse sand #8 (Unimin Corp.), this constant head boundary was designed to simulate a hydrostatic pressure condition at the right limit of the aquifer. The synthetic aquifer consisted of a homogeneous medium with an inclination of 2° underlying a practically impervious layer of very fine sand consisting of a 2:1 mixture of sieve sizes #110 and #250 (U.S. Silica Company) that acted as caprock. For each experiment, the height difference between the two reservoirs was adjusted according to the permeability of the sand to produce an injection flow rate of 0.7 ml/min. During injection, this flow rate was maintained constant throughout the three experiments corresponding to a capillary number of 1×10^{-5} . The magnitude of the capillary number for our experiments is in agreement with values reported by Bandara et al. (2011) for a high permeability reservoir. The darcy velocity used for calculating the capillary number was estimated by dividing the volumetric flow rate of the injected fluid by the total area of the screen. After the injection stage, the plume was monitored through photography and x-ray attenuation measurements until no variation in Soltrol 220 saturation was detected, indicating a fully immobilized plume. Afterwards, in order to establish a wetting phase flow corresponding to a chase brine event, the Mariotte bottle previously filled with Soltrol 220 was replaced with a reservoir filled with glycerol-water. In order to prevent the wetting phase from by-passing the NWP plume accumulating below the fine sand layer, the flow rate of this wetting phase flow event was maintained at 2 ml/min until all the mobile fraction of NWP had left the reservoir.

[Figure 2 here]

A set of unconsolidated porous media with variable permeability was used to study the multiphase flow behavior through a range of Bond numbers, expecting higher permeability to cause a faster vertical migration of the injected NWP due to a stronger influence of gravity over capillary forces. The corresponding Bond numbers changed from 2.61×10^{-5} (for experiment #1) to 3.3×10^{-6} (for experiment #3). The laboratory sands used for packing the synthetic aquifer are identified by the effective sieve numbers and consisted of hydrophilic silica sands commercialized under the brand Accusand (Unimin Corp.) with well-characterized physical properties (see Table 2). Although the viscosity and density contrasts of the fluids used in the experiments are representative for scCO_2 /brine at reservoir conditions, the permeability expected at field sites such as Sleipner will be 1-2 orders of magnitude lower than the laboratory sands.

[Table 2 here]

2.3 X-ray attenuation method

Central to the experimental approach is the use of X-ray attenuation to nondestructively determine the spatial and temporal distribution of the phase saturations in the test tanks. In fact, the attenuation of an X-ray beam traversing a porous medium is influenced by the saturation of fluid phases present in the pore space and the intensity of the beam (Oak and Ehrlich, 1988). The X-ray attenuation method is based on Beer-Lambert's law, which describes the change in energy intensity for a beam of mono-energetic photons traveling through a material with linear attenuation coefficient α and thickness x :

$$I/I_0 = e^{-\alpha x} \quad (3)$$

where I is the intensity of photons (photons/time) transmitted across the material and I_0 is the photon flux in the absence of a material or incident beam intensity. To measure the void fraction of the porous medium (i.e., porosity) as well as the amount of fluid phases through a non-invasive manner, we employed a custom-designed x-ray attenuation device. By moving the x-ray source (an AEG/Thomson x-ray tube with Tungsten filament) and photon detector (a PGT/HPGe connected to a multichannel analyzer) simultaneously along a predefined scanning grid, we were able to measure volumetric fractions of any of the phases inside the tank (glycerol-water, Soltrol 220, sand) and interpolate them in two dimensions to obtain contours of equal saturations. These measurements represent a depth-average along the second horizontal axis (y) and an areal average over a sampling volume determined by the radius of the collimated x-ray beam (1 mm). The x-ray generator was operated with tube settings of 70 kV and 10 mA, while 2 heavy rare earth element filters (Erbium carbonate and Samarium carbonate) were used as absorption filters to condition the spectra. A 10-s detector live time was used for photon counting at each point, resulting in an average total measurement time of 28 s, including the positioning routine. The procedure used to extrapolate sand porosity and NWP saturation from material path lengths follows that of Fagerlund et al. (2007) and is outlined in Fig. 3, where each step describes the acquisition process of a phase path length. Glycerol-water (x_w), sand (x_s), and Soltrol 220 (x_n) path lengths are measured to estimate flume thickness, porosity, and NWP saturation, respectively. Prior to any path length estimation, linear attenuation coefficients for each phase had to be characterized.

[Figure 3 here]

The accuracy of the x-ray attenuation device was tested prior to each experiment by measuring material path lengths across known thicknesses. The method consisted of placing two 1.0-cm thick disposable spectrophotometry cuvettes within the path line of the x-ray beam. First the two cuvettes were filled with the wetting phase (i.e. glycerol-water mixture) for the baseline measurement “ I_0 ”, then one cuvette was replaced with another containing the NWP, for the “I” measurement. The application of the Beer-Lambert law allowed for the estimation of the path length of the NWP, which was within 98% agreement (0.02 cm underestimated path length of 1 cm) of the exact width of the cuvette.

2.4 Flume thickness

In previous laboratory experiments conducted in the same flow cell by Fagerlund et al. (2007), the vertical Plexiglas™ walls displayed a noticeable bulging across the y-axis, which could affect the estimation of sand porosity and NWP saturation. In order to normalize the sand and NWP path lengths at each scanning point by the variable thickness of the flume, we followed the first step in Fig. 3 to determine the width variation for the whole flow domain. The contour map resulting from interpolation of x-ray attenuation data in Fig. 4 revealed an increasing thickness (in cm) approaching the center of the flume. In order to reduce the noise from acquired data, before proceeding to the porosity estimation, a smooth surface was fitted to the interpolated contours and these estimates were used to represent the two-dimensional variation of flume thickness (Fig. 4).

[Figure 4 here]

2.5 Porosity

After wet-packing with a known mass of sand and prior to the injection of the NWP into the synthetic reservoir, a complete x-ray scan of the flow domain was performed to characterize the spatial distribution of sand porosity. A total of 611 measuring (or scanning) points covered the flow domain with a measurement spacing of $1.5 \times 1.5 \text{ cm}^2$. Fig. 5 shows the resulting contour maps of the porosities for the three packing configurations, indicating that a certain degree of heterogeneity exists even though all attempts were made to pack the tank homogeneously. However, it was observed during each displacement experiment that there was no evidence of preferential flow pathways due to porosity changes in the sand pack. Without accounting for the small fraction of sand volume around the well, a total pore volume of 2270, 2379, and 2462 cm^3 was measured for experiments #1, #2, and #3, respectively. These values agreed with the porosity estimated after the total mass used to pack the tank. Fig. 6 shows porosity histograms from x-ray data analysis for the three packing configurations, where mean values are in good agreement with measurements from Zhang et al. (2008) (Table 2).

[Figure 5 here]

[Figure 6 here]

3. Results

3.1 *NWP saturation*

We adopted two approaches for the visualization of the evolution of NWP saturation during the plume migration: 1) a qualitative comparison between digital photography and linear interpolation of NWP saturation measurements via x-ray attenuation, and 2) a temporal series of

saturation observed at selected locations. For the latter approach, we omitted the plots corresponding to those locations that did not show saturation changes above the detection limit. Well refined x-ray measurements provided saturation data for quantitative analysis and allowed for spatial moment analysis in order to estimate the total NWP volume present in the flow domain. For this purpose the zeroth order spatial moment as reported by Freyberg (1986) and Fagerlund et al. (2007) was used to match the injected volume of Soltrol 220 tracked via electronic scale. In Fig. 7 we show a comparison of cumulative NWP volume measured by electronic scale versus volume estimated from zeroth moment calculation. Fig. 7 shows a 30 ml-mismatch (12.7% of total volume injected) in volume estimation between zeroth moment and NWP weight measurements acquired from the electronic scale. This is attributed to the unavailability of x-ray data at the well location, causing the NWP volume around the well to remain out of reach for spatial moment calculations.

$$V_{NWP} = M_{00} = \int \int \varphi(x, z) S_{NWP}(x, z) dx dz \quad (4)$$

[Figure 7 here]

Fig. 8 illustrates the distribution of 25 observation points that were chosen to compare temporal plots of saturation measured using x-ray attenuation for different Bond number scenarios throughout all three experiments. The vertical spacing between each point is 1.5 cm. The variable horizontal spacing between the vertical arrays was intended to better capture the evolution of plume saturations in the proximity of the injection location.

[Figure 8 here]

The highest Bond number scenario corresponds to experiment #1, where the synthetic reservoir is packed with #30/40 Accusand (see Figure 9). By interpolating the saturation data acquired via x-ray attenuation throughout the whole flow domain, contour maps of the NWP saturation were generated, allowing identification of regions with mobile and disconnected phases. The larger influence of buoyancy versus capillary forces produced by the higher permeability of this sand pack led to strong gravity segregation between the two phases, as well as NWP plume propagation on the very top of the flow domain, along the caprock interface. As a result, poor volumetric sweep efficiency and rapid NWP build-up near the injection well were observed (Fig. 9a). This behavior has been shown in previous studies presented in the literature (e.g. Ide et al., 2007; Tchelepi and Orr, 1994), however this experiment represents an attempt to demonstrate the validity of some of the predictions made using numerical simulations, in the laboratory. A total volume of 231 ml of Soltrol 220, corresponding to approximately 10% of the reservoir pore volume, was injected during 5.5 hours, which resulted in NWP saturations greater than 0.9 near the injection well at the end of injection (Fig. 9a). Once the plume had completely redistributed as driven by buoyancy after about 590 hours (Fig. 9b), a forced imbibition event was created with the intent of displacing any mobile fraction of the plume away from the well and maximizing the contact area between the plume and the reservoir, which is known to affect the final sweep efficiency (Fig. 9c). Even though there is a clear boundary effect on the right limit of the reservoir, we focus our attention on the near-well area, where NWP phase saturations measured at the onset of forced imbibition still had saturations values higher than the expected residual value associated with this porous material, which is 0.26 for sand #30/40.

[Figure 9 here]

The observation for an extended period of time of NWP saturations higher than residual near the injection well was confirmed by analyzing the time series shown in Fig. 10. Even though NWP saturation slowly decreased between 40 and 590 hours, all the observation points near the injection well (A, B, C, D, E) remained above residual saturation before the impact of forced imbibition. Faster vertical migration, together with a local smoothing of the caprock inclination near the well and a lower porosity at the middle top of the reservoir (see Fig. 5) due to a tighter packing of the sand may have limited further migration of the plume in the updip direction. Except for point A, which lays above the interface between the reservoir sand and the caprock sand, the other 24 points recorded increasing saturation values at higher vertical locations, as a proof of the presence of a saturation transition zone produced by capillary forces. Also, saturations higher than 0.3 were detected at the two uppermost points of the left half of the reservoir (F, G), suggesting the presence of continuous phase in variable amounts, possibly due to the lower porosity zone just mentioned. The persistence of high saturations at the mentioned locations was observed during the next two days after the injection ceased, hence the plume was left to redistribute for longer times (>3 weeks), before starting the forced imbibition event.

All these points, as well as those nearby the well (B, C, D, E), recorded saturations below 0.3 once the forced imbibition was completed, indicating that a residual saturation had been reached (see Table 2). Some locations, especially in the upper region of the aquifer, exhibited a rapid spike in NWP saturation after the onset of the glycerol-water injection (i.e., forced imbibition), due to the piston-like displacement of mobile NWP phase from left to right. The most important result that can be derived from the observation points in the right half of the

reservoir (P through Y) is the increase in NWP residual saturation achieved after the forced imbibition event. This behavior represents an effect of a secondary drainage/imbibition cycle and it can be translated to field-scale projects, where such an injection strategy can eventually lead to enhanced storage of CO₂ at locations far from the injection well.

[Figure 10 here]

Experiment #2 was conducted in less permeable sand to reduce the relative magnitude of buoyancy versus capillary forces and correspondingly decrease the value of the Bond number (1.47×10^{-5}). In this case, the sand pack consisted of #40/50 Accusand, a medium with permeability 40% lower than #30/40 sand. Examining the photo images and two-dimensional contour maps of measured NWP saturation shown in Fig. 11, it is noticeable that saturation of Soltrol 220 (surrogate scCO₂) near the injection well quickly rose to 0.8. Then, after the cease of the injection stage, the NWP saturation slowly decreased until it reached an immobile condition as a result of capillary trapping. Once the wetting cycle was completed, residual saturations achieved at these locations represented the endpoint $S_{nw}^{r,max}$, or the maximum historical NWP saturation, which represents a critical parameter in multiphase flow simulations that implement the Land's trapping model (Land, 1968).

[Figure 11 here]

During experiment #2 there was no evidence of the strong gravity segregation observed in the previous one. For this reason, the forced imbibition stage did not lead to the marked effect

on residual saturation that had arisen from the piston-like displacement of the mobile NWP accumulating on the left section of the reservoir during experiment #1. However, a feature that can be readily observed from the temporal series in Fig. 12 is the different depletion rates in NWP saturation at observation points at different vertical locations during the 240 hour natural imbibition or the recovery stage. Also, a delayed response in saturation depletion at downstream points represents an effect of the updip migration of mobile NWP from the well area. This behavior is expected to be more pronounced in higher permeability reservoirs where the effect of buoyancy forces is stronger. In field sequestration scenarios, these outcomes are relevant to the reliability of capillary trapping at deeper regions, where scCO_2 at the trailing edge of the plume may become immobile with a faster rate compared to regions underlying the caprock.

[Figure 12 here]

In order to provide an injection scenario characterized by a lower influence of buoyancy compared to capillary forces, we performed a third experiment in a geometrically similar setting with sand of even lower permeability. In this case, we wet-packed the reservoir with #50/70 Accusand, which has a permeability 47% lower than #40/50 Accusand used in experiment #2. Once the Soltrol injection was stopped, the plume configuration appeared more compact compared to experiments #1 and #2, as evidence of increased sweep efficiency (Fig. 13a). This process can eventually affect the storage capacity of a given reservoir (Cinar et al., 2009).

[Figure 13 here]

Similarly to what was observed in experiment #2, NWP saturations measured at higher locations were characterized by slower depletion rates. Particularly, some tendency to plume accumulation was observed at point A, although a residual saturation of 0.25 was achieved after leaving the fluids to redistribute spontaneously during approximately one week (Fig. 14).

[Figure 14 here]

To evaluate the ability of a number of trapping models to fit our experimental data, we followed the analysis by Pentland et al. (2010) and compared four trapping models from the literature (Jerauld, 1997; Land, 1968; Ma and Youngren, 1994; Spiteri et al., 2005). A comparison between the mentioned trapping models and the correlation for the maximum NWP saturation reached at flow reversal (S_{nw}^i) and the residual NWP saturation (S_{nw}^r) for the three experiments is shown in Fig. 15. A common feature among these plots is the higher occurrence of measurements at initial saturations that are lower than 0.2, suggesting that most of the NWP is residually trapped at low saturations. Also, as previously observed in Krevor et al. (2012) from core-flood experiments with scCO₂ in several sandstones, the Land's fitting coefficient C increases (reducing the capillary trapping) as the permeability of the porous medium decreases. Our displacement experiments conducted in unconsolidated sands indicate values of C varying from 3 (for #30/40 sand) to 5.4 (for #40/50) to 8.6 (for #50/70). However, the fitting of Land's model to the experimental data of experiment #3 was significantly biased by the larger use of measurements at small initial saturations. The red dots in the initial-residual saturation plots denote NWP saturations detected prior to the onset of glycerol-water injection, while the black dots indicate NWP saturations achieved once the forced imbibition stage had been completed. In

the case of experiment #1, where an accumulation of Soltrol 220 was observed underneath the confining layer, the red dots follow a linear trend that is typical for 100% trapping (Stegemeier, 1977) or pooling of NWP due to capillary barrier effect (Fagerlund et al., 2007; Krevor et al., 2011).

[Figure 15 here]

4. Discussion and Conclusions

A set of laboratory experiments was conducted in a quasi-two-dimensional flow cell with surrogate fluids at ambient conditions. A fluid combination was selected to reproduce density and viscosity contrasts between scCO₂ and brine at deep reservoir conditions. This approach avoided the complexity of dealing with high-pressure cells and allowed for the observation of multidimensional immiscible displacements. We believe that this is the first attempt to conduct displacement experiments at the meter scale to obtain a fundamental understanding of the trapping of scCO₂ using surrogate fluids. The capillary number and the Bond number were selected to describe the relative influence of viscous, capillary, and buoyancy forces on the propagation of the injected fluid phase. The magnitude of these dimensionless groups was maintained within a range of expected values for actual deep injection scenarios of scCO₂ in deep saline formations. As expected by theory, less pronounced gravity segregation due to lower Bond number led to a better vertical sweep efficiency, which is defined by Lake (1989) as the ratio between the cross-sectional area of reservoir contacted by the plume and the total cross-sectional area. A simple photographic analysis allowed for the evaluation of the vertical sweep efficiency as well as the migration distance of the plume, while the amount of NWP that left the

flume before and after the forced imbibition stage could be quantified after phase separation of the effluent. Table 3 summarizes the results of these evaluations for all the experiments, giving an overall outline of the displacement efficiency for reservoirs with different permeability. Except for experiment #1, where a considerable fraction of the injected fluid (76%) remained in the reservoir before the onset of forced imbibition, the rest of the experiments exhibited a depletion of the NWP plume during the fluid redistribution stage. Consequently, the forced imbibition stage had a minor effect on the entrapped plume for experiments #2 and #3, displacing only 2% of the NWP remaining in the reservoir.

As a further comparison of displacement efficiency between the three experiments, we calculated the ratios between the tip position of the plume at the end of the injection stage (x_{tip}) and the total length of the reservoir. While for the largest permeability scenario (experiment #1) the plume spanned the whole length of the reservoir, ‘leaking’ faster through the right boundary, the distance traveled by the plume during experiments #2 and #3 was 16% shorter.

[Table 3 here]

We investigated the correlation between initial and residual NWP saturation for three experiments conducted with variable sand permeability and we were able to show the effect of structural versus residual trapping based on the inclination of the confining sand. We showed that single two-dimensional displacement experiments allow for the observation and measurement of a range of initial NWP saturations that would be otherwise acquired through a series of one-dimensional core flooding experiments at different injection flow rates, as reported by Pentland et al. (2010) or Akbarabadi and Piri (2013) among others. Four trapping models

from the literature were able to reproduce reasonably well ($R^2 > 0.6$) the initial-residual NWP saturation correlation for the experiments.

After evaluating the results achieved from the experiments conducted in this study, we draw the following conclusions:

1. Intermediate-scale experiments conducted with surrogate fluids under highly controlled laboratory conditions facilitate the study of fundamental processes that occur under equivalent interplay of capillary, viscous, and buoyancy forces, that is not feasible in the field;
2. Our experiments provide data sets to test the applicability of numerical multiphase flow models to simulate saturation hysteresis and capillary trapping of wetting and non-wetting fluids;
3. Single immiscible displacement experiments conducted in a quasi-two-dimensional flow cell allow for the measurement of a wide range of initial NWP saturations. This makes comparison of theoretical trapping models with actual trends observed in experimental data possible;
4. For displacements carried out at an equal capillary number $Ca \sim O(10^{-5})$, gravity segregation is predominant when the flow is characterized by higher Bond numbers, which for equal density difference are highly dependent on sand permeability;
5. A comparison of the rates at which the saturations reach final residual values at different depths below the caprock shows that the saturations at deeper location reaches the

residual faster. This observation supports the convenience of “inject low and let rise” injection schemes.

6. To minimize the occurrence of mobile NWP in the reservoir, a forced imbibition event such as a chase brine injection can eventually initiate a secondary drainage/imbibition cycle, leading to an enhanced NWP residual saturation at locations far-off from the injection well.

Since natural heterogeneities are an inherent feature of geologic reservoirs, further experimental study of migration and trapping of CO₂ in non-homogeneous aquifers is needed. These heterogeneous experiments could help clarify the importance of constitutive models and upscaling methods commonly used for multiphase flow through heterogeneous media. In addition, a modeling study of these experimental systems could verify the applicability of continuum multiphase flow models to the prediction of residual trapping of scCO₂ in deep saline aquifers.

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797 vertical sweeping efficiency, and plume propagation.

Table 1

phase	ρ (kg m ⁻³)	μ (mPa· s)	μ_{nw}/μ_w	ρ_{nw}/ρ_w	IFT (mN m ⁻¹)
Soltrol 220	860	4.9	0.072	0.71	15
Glycerol-water	1210	68			
scCO ₂	266-733 ^a	0.023-0.0611 ^a	0.026-0.20 ^a	0.22-0.75 ^a	19.8 ^b
	(760 ^c)	(0.06 ^c)			
Brine	945-1230 ^a	0.195-1.58 ^a	(0.075 ^c)	(0.745 ^c)	
	(1020 ^c)	(0.8 ^c)			

^a estimates from Nordbotten et al. (2005), T = 35-155°C, P = 10.5-31.5 MPa

^b measurement from Bennion and Bachu (2006), T = 43°C, P = 20 MPa, brine salinity = 2.7% wt.

^c estimates from Singh et al. (2010) for Sleipner field

Table 1. Summary of density and viscosity for surrogates at ambient conditions and for actual fluids at reservoir conditions.

Table 2

Experiment	Sand	$k \text{ (m}^2\text{)}$	$\Phi_{\text{avg}} \text{ (-)}$	$d_{50} \text{ (mm)}$	$d_{60}/d_{10} \text{ (-)}$	$S_{\text{nw}}^{r,\text{max}}$	$\text{Bo (x10}^{-5}\text{)}$
1	#30/40	$1.14 \times 10^{-10} \text{ }^a$	0.354 (0.355 ^a)	0.45 ^b	1.22 ^b	0.26	2.61
2	#40/50	$6.42 \times 10^{-11} \text{ }^a$	0.39 (0.355 ^a)	0.28 ^b	1.21 ^b	0.22	1.47
3	#50/70	$3.46 \times 10^{-11} \text{ }^a$	0.379 (0.372 ^a)	0.23 ^b	1.28 ^b	0.25	0.33
1-3	#8	$1.55 \times 10^{-9} \text{ }^c$	0.396 ^c	1.4 ^c	1.6 ^c	N/A	N/A
1-3	#110	$5.21 \times 10^{-12} \text{ }^c$	0.34 ^c	0.12 ^c	1.6 ^c	N/A	N/A

N/A: not available.

^a from Zhang et al. (2008).

^b estimated from sieve data provided by the manufacturer.

^c from Sakaki and Illangasekare (2007).

Table 2. Physical properties for the silica sand grades used in the displacement experiments with corresponding Bond numbers.

Table 3

	Experiment #1	Experiment #2	Experiment #3
NWP left before forced imb./Total NWP injected	76 %	37 %	30 %
NWP left after forced imb./Total NWP injected	40 %	35 %	28 %
vertical sweep efficiency	25 %	25 %	35 %
$\frac{\max x_{tip}}{x_{reservoir}}$	100 %	84 %	84 %

Table 3. Summary of the displacement experiments with assessments of NWP mass balance, vertical sweeping efficiency, and plume propagation.

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Figure 14 B&W

Figure 14. Experiment #3: temporal series of NWP saturation at the observation points shown in Fig. 8 throughout the injection and recovery stages (left column) and forced imbibition stage (right column). The solid and dotted vertical lines highlight the end of the injection stage and the beginning of the forced imbibition event, respectively.

Figure 15 (B&W)

Figure 15. Initial-Residual saturation data observed before and after forced imbibition compared with four different trapping models. Regression analysis of Land’s trapping coefficient performed on data observed after forced imbibition.

Figure 2 color

Figure 2. Sketch with dimensions of the synthetic aquifer and experimental setup.

Figure 3 color

Figure 4 color

Figure 4. Empty flume with overlapped interpolated contours of: 1) flume thickness (in grey shades) and 2) smooth surface (red lines).

Figure 5 color

Figure 5. Contour maps of sand porosity for the three packing configurations.

Figure 7 color

Figure 7. Cumulative NWP volume measured via electronic scale (red line) and derived from zeroth moment calculation (black circles).

Figure 8 color

Figure 8. Experimental flume after wet-packing with location of the 25 observation points; broken lines delineate flow domain, injection well, coarse sand boundary.

Figure 9 (color)

Figure 9. Image and saturation contours of NWP plume at different times during experiment #1 (#30/40 Accusand): at the end of the injection stage (a), and before and after the forced imbibition stage respectively b, c).

Figure 10 color

Figure 10. Experiment #1: temporal series of NWP saturation at the observation points shown in Fig. 8 throughout the injection and recovery stages (left column) and forced imbibition stage (right column). The solid and dotted vertical lines highlight the end of the injection stage and the beginning of the forced imbibition event, respectively.

Figure 11 (color)

Figure 11. Image and saturation contours of the NWP plume at different times during experiment #2 (#40/50 Accusand): at the end of the injection stage (a), and before and after the forced imbibition stage respectively (b, c).

Figure 12 color

Figure 12. Experiment #2: temporal series of NWP saturation at the observation points shown in Fig. 8 throughout the injection and recovery stages (left column) and forced imbibition stage (right column). The solid and dotted vertical lines highlight the end of the injection stage and the beginning of the forced imbibition event, respectively.

Figure 13 (color)

Figure 13. Image and saturation contours of NWP plume at different times during experiment #3 (#50/70 Accusand): at the end of the injection stage (a), and before and after the forced imbibition stage respectively (b, c).

Figure 14 color

Figure 14. Experiment #3: temporal series of NWP saturation at the observation points shown in Fig. 8 throughout the injection and recovery stages (left column) and forced imbibition stage (right column). The solid and dotted vertical lines highlight the end of the injection stage and the beginning of the forced imbibition event, respectively.

Figure 15 (color)

Figure 15. Initial-Residual saturation data observed before and after forced imbibition compared with four different trapping models. Regression analysis of Land's trapping coefficient performed on data observed after forced imbibition.