

## Displacement and mass transfer between saturated and unsaturated CO<sub>2</sub>–brine systems in sandstone

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### ABSTRACT

The process of displacement and mass transfer between CO<sub>2</sub> and brine, which are relevant for the prediction of plume migration and pore-space utilization during CO<sub>2</sub> injection in saline aquifers, were studied by conducting unsteady-state core flood experiments in nearly homogeneous Berea sandstone rock. Mutually saturated and unsaturated CO<sub>2</sub> and brine phases were injected in the rock under realistic sequestration conditions.

Relative permeability and capillary pressure curves were extracted by history matching the unsteady state experiments conducted with mutually saturated CO<sub>2</sub> and brine. As a reference and for comparison, decane–brine primary drainage was conducted on the same sample. The CO<sub>2</sub>–brine relative permeability was found to be different from the decane–brine relative permeability (which had been validated against steady-state experiments on twin-samples), reflecting the change in the wetting state from water-wet decane–brine/Berea to the rather intermediate-wet behavior of CO<sub>2</sub>–brine/Berea, which is in agreement with literature data on contact-angles for the two cases. However, the CO<sub>2</sub> brine data are somewhat different from data on the same rock type as reported by Perrin and Benson (2010) which is likely a consequence of sample heterogeneity.

Aspects of the mass transfer between the CO<sub>2</sub> and the brine phase were studied by drainage and imbibition with unsaturated phases. When comparing saturated and unsaturated CO<sub>2</sub>–brine primary drainage, the mass transfer due to mutual solubility leads to two effects: (1) evaporation near the inlet due to water dissolving in CO<sub>2</sub> and (2) a diminished displacement of brine by CO<sub>2</sub> due to CO<sub>2</sub> dissolving in brine. In addition, an imbibition experiment was conducted where unsaturated brine was injected into rock filled with mutually saturated CO<sub>2</sub> and brine phase at near-residual CO<sub>2</sub> saturation. After the CO<sub>2</sub>-saturated brine had been miscibly displaced by unsaturated brine, dissolution of the trapped CO<sub>2</sub> in the injected brine was subsequently observed. These experiments represent the transition from residual trapping to solubility trapping and indicate the time and length scales involved.

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### 1. Introduction

Geological sequestration of carbon dioxide (CO<sub>2</sub>), originating from sources like fossil-fueled power plants and contaminated gas production, is presently seen as a large scale disposal option to reduce greenhouse-gas emission (IPCC, 2005). Deep saline aquifers and depleted oil and gas fields are potential candidates for CO<sub>2</sub> subsurface deposits (IPCC, 2005; Bachu et al., 1994; Bachu and Gunter, 2004). Trapping mechanisms and the degree of trapping are important factors for assessing the storage capacity for sequestration in formation rock (Suekane et al., 2008).

The migration of the CO<sub>2</sub> plume and the pore-space utilization by capillary trapping are two aspects of the displacement process in the subsurface that are relevant for assessing the safety aspects of

geological sequestration (Esposito and Benson, 2010). Single phase fluid flow in porous media, in general, is described by the porosity ( $\phi$ ) and permeability ( $K$ ) of the formation rock, and the fluid viscosity. When considering multiphase flow (immiscible displacement), e.g. the CO<sub>2</sub>–brine system, relative permeability ( $k_r$ ) and capillary pressure ( $p_c$ ) saturation functions are important factors as well. The latter two parameters largely influence the plume migration (Egermann et al., 2006), the CO<sub>2</sub> saturation of the pore space, and the residually trapped volume after re-imbibition of the aquifer.

Then availability of relative permeability data for CO<sub>2</sub>–brine drainage is currently seen as one of the critical subsurface issues in CO<sub>2</sub> storage (Imbus et al., 2006). In petroleum engineering, these parameters are commonly measured in either steady state (SS) or unsteady state (USS) core flood experiments. While these measurements already require substantial resources (rock samples, instrumentation, time), the special requirements of CO<sub>2</sub> in particular set high demands on materials and experimental procedures (Bennion and Bachu, 2005). As a consequence, there have so far

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been only a few attempts to determine data such as the relative permeability of CO<sub>2</sub>–brine systems.

Systematic studies were conducted by Bennion and Bachu (2005, 2008) who determined CO<sub>2</sub>–brine and H<sub>2</sub>S–brine relative permeability in sandstone and carbonate samples from the Wabamun and Zama area in central Alberta, Canada. Drainage and imbibition experiments were carried out in the unsteady state with mutually saturated fluid phases on representative single cores or multiple plug stacks. The relative permeability curves were extracted from the total brine and CO<sub>2</sub> flow rates and the pressure drop during the core flood. Since no in situ saturation monitoring e.g. by imaging was used, i.e. no in situ saturation profiles in the sample were accessible, the CO<sub>2</sub> endpoint relative permeability and the capillary end effect were determined by multi-rate end point floods. Relative permeability saturation functions were determined from production data and pressure drop over the core using numerical regression methods (Bennion and Bachu, 2005). The residual fluid saturations were determined ex situ by Dean-Stark extraction after depressurization.

Egermann et al. (2006) also used unsteady-state core flooding experiments to determine relative permeability data for homogeneous limestone rock with the aim of using that data for an integrated modeling approach.

Perrin et al. (2009); Perrin and Benson (2010); Kuo et al. (2010) measured CO<sub>2</sub>–brine relative permeability on Berea sandstone and on a sample originating from a storage site in southwest Australia in steady-state (SS) experiments. All samples had a substantial degree of heterogeneity. The authors studied the influence of that heterogeneity on fluid flow and CO<sub>2</sub> distribution in the core. Furthermore, both experiments showed a flow rate dependence of the relative permeability curves obtained.

In this study, we report on unsteady state (USS) experiments combined with X-ray computed tomography (CT) to determine relative permeability  $k_r$  and capillary pressure  $p_c$  for primary drainage of brine by supercritical CO<sub>2</sub> in a nearly homogeneous Berea sandstone at conditions representative for a realistic field case. Berea sandstone is an outcrop rock which is widely used as a model and reference rock. We use the USS method because it has a large degree of analogy with the actual field process for CO<sub>2</sub> injection. USS experiments typically cover the range where fluids are mobile; this is also the range that is relevant for plume migration in the field.

### 1.1. Experiments conducted in this study

A set of four different unsteady-state experiments were conducted on the same Berea sample starting with a primary drainage experiment where brine is displaced by decane. This experiment served as a reference experiment. On a very similar Berea rock sample from the same block, steady-state experiments were conducted using decane–brine/glycol for comparison with the unsteady-state data.

In the second experiment, brine not equilibrated with CO<sub>2</sub> (referred to as unsaturated brine) was displaced by supercritical CO<sub>2</sub> not equilibrated with brine (referred to as unsaturated CO<sub>2</sub>) and the effects of mass transfer between the non-equilibrated phases were observed. With respect to CO<sub>2</sub> injection in the field, this case is representative for the flood front where fluids have not yet fully equilibrated.

In experiment three, saturated brine was displaced by saturated CO<sub>2</sub>. Here, no mass transfer between the fluids is expected and the data is directly compared with the decane–brine reference case. With respect to CO<sub>2</sub> injection in the field, this case is representative for regions where fluids are already fully equilibrated, e.g. behind the flood front and in the center of the plume.

**Table 1**

Overview of the experiments, flow velocity and capillary numbers  $N_{cap}$ .

Exp.	System parameters	Flow velocity (ft/day)	$N_{cap} (\times 10^{-8})$
<i>Drainage</i>			
1	Decane → brine	1.2	6.5
	Decane → brine/glycol (SS)	1.8–88	29–1469
2	Unsaturated: CO <sub>2</sub> → brine	2.3	0.88
3	Saturated: CO <sub>2</sub> → brine	2.0	0.79
<i>Imbibition</i>			
4	Unsaturated brine → trapped CO <sub>2</sub>	1.2	9.3

Experiment four started at the end of experiment three: unsaturated brine was injected in order to “simulate” the re-imbibition of the aquifer after CO<sub>2</sub> injection was stopped.

The experiments conducted in this study are listed in Table 1.

## 2. Materials and methods

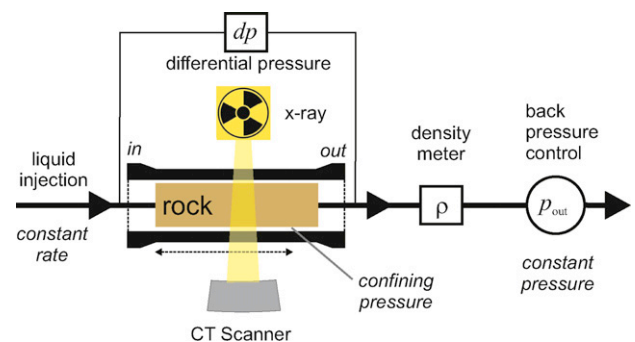
### 2.1. Flow setup

A cylindrical rock sample (“core”) contained in a metal sleeve with a Teflon liner is enclosed in a (X-ray transparent) carbon fiber epoxy core holder (Coenen, 2009). The core was oriented in the horizontal direction (same as main flow direction in the field). Fluids were injected through a distribution plate and a highly permeable porous metal disk into the inlet face of the cylindrical sample. The same configuration was used at the outlet as well.

The sleeve annulus is pressurized to 150 bar in order to press the sleeve on the core and avoid bypassing of fluids. A sketch of the setup, which has been described in more detail in Berg et al. (2010), is shown in Fig. 1.

N-decane, CO<sub>2</sub> and brine are injected at a constant rate into the core via a high pressure syringe pump (Teledyne ISCO, Lincoln NE, USA). For equilibration of CO<sub>2</sub> with water and during CO<sub>2</sub> injection, the pumps were kept at 25 °C and the CO<sub>2</sub> was heated to experimental temperature (45 °C) in the injection lines. This results in a slight under-saturation of the CO<sub>2</sub> ensuring the injection of a single phase, and increases the total available CO<sub>2</sub> volume at experimental conditions. At the outlet, a constant pressure is maintained either by a dual cylinder piston pump (Quizix, Vindum Engineering, INC., San Ramon CA, USA) or by two Tescom back pressure controllers which are operated in series. All seals were made from Teflon, highly fluorinated rubber or Viton-A (hardness: shore 90) to avoid the risk of explosive decompression of CO<sub>2</sub>.

Injection rates ranged between 0.25 and 0.5 ml/min which corresponds to flow velocities of 1.2–2.3 ft/day which are often considered to be typical field flow rates (can be estimated from Figs. 3–17 in Lake, 1989). The associated capillary numbers  $N_{cap} = \mu v / \sigma \approx 10^{-8} - 10^{-7}$  ( $\mu$  is the viscosity of the displacing fluid,  $v$  the velocity, the interfacial tension  $\sigma$  for decane–brine is 47.9 mN/m



**Fig. 1.** Experimental setup.

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