



Geophysical Prospecting

**Integrated Geophysical and Hydromechanical Assessment
for CO₂ Storage: Shallow Low Permeable - Reservoir
Sandstones**

Journal:	<i>Geophysical Prospecting</i>
Manuscript ID	GP-2015-0062.R2
Manuscript Type:	Special Issue
Date Submitted by the Author:	19-Jan-2016
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Keyword:	Electrical resistivity, Seismic, Permeability

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7 **Integrated Geophysical and Hydromechanical**
8 **Assessment for CO₂ Storage: Shallow Low Permeable -**
9 **Reservoir Sandstones**
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39 Manuscript submitted to:

40 **Geophysical Prospecting**
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45 *Special Issue "Advances in Rock Physics"*
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48 Date: August, 2015
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Abstract

Geological reservoirs can be structurally complex and can respond to CO₂ injection both geochemically and geomechanically. Hence, predicting reservoir formation behaviour in response to CO₂ injection, and assessing the resulting hazards are important pre-requisites for safe geological CO₂ storage. This requires a detailed study of thermal-hydro-mechanical-chemical coupled phenomena that can be triggered in the reservoir formation, most readily achieved through laboratory simulations of CO₂ injection into typical reservoir formations. Here, we present the first results from a new experimental apparatus of a steady state drainage flooding test conducted through a synthetic sandstone sample, simulating real CO₂ storage reservoir conditions in a shallow (~1 km), low permeability ~1mD, 26% porosity sandstone formation. The injected pore fluid comprised brine with CO₂ saturation increasing in steps of 20% brine/CO₂ partial flow rates up to 100% CO₂ flow. At each pore fluid stage, an unload/loading cycle of effective pressure was imposed to study the response of the rock under different geomechanical scenarios. The monitoring included axial strains and relative permeability in a continuous mode (hydromechanical assessment), and related geophysical signatures (ultrasonic P- and S-wave velocities, V_p and V_s respectively, and attenuations, Q_p^{-1} and Q_s^{-1} respectively, and electrical resistivity, ER). On average, the results showed V_p and V_s dropped ~7% and ~4% respectively during the test, while Q_p^{-1} increased ~55% and Q_s^{-1} decreased by ~25%. From the electrical resistivity data, we estimated a maximum CO₂ saturation of ~0.5, while relative permeability curves were adjusted for both fluids. Comparing the experimental results to theoretical predictions, we found that Gassmann's equations explain V_p at high and very low CO₂ saturations, while bulk modulus yields results consistent with White and Dutta-Odé model

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7 predictions. This is interpreted as an heterogeneous distribution of the two pore fluid
8 phases, corroborated by electrical resistivity tomography images. The integration of
9 laboratory geophysical and hydromechanical observations on representative shallow
10 low permeable - sandstone reservoir allowed us to distinguish between pure
11 geomechanical responses and those associated with the pore fluid distribution. This
12 is a key aspect in the understanding of CO₂ injection effects in deep geological
13 reservoirs associated with carbon capture and storage practices.
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25 **Key words:** seismic velocity, attenuation, electrical resistivity, permeability, CO₂
26 injection, reservoir geophysics.
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1 Introduction

Atmospheric concentrations of carbon dioxide (CO₂) have increased dramatically since the end of the nineteenth century, owing chiefly to increased burning of fossil fuels by humans, but also to steel works, cement factories and chemical plants. It is well accepted that, to date, the most realistic leading mitigation strategy is Carbon Capture and Storage (CCS, IPCC 2005). CCS technologies involve the sequestration of CO₂ into deep brine saturated geological formations, specially selected for such massive-scale storage. While the concept is a promising one, uncertainties and risks remain a matter of concern, especially with regard to public acceptance related to induced overpressure from CO₂ injection, such as seismicity (earthquakes and landslides) or leakages to shallower aquifers. So, it is of crucial importance to develop methods and monitoring tools to accurately map the subsurface CO₂ plume evolution (Shi *et al.* 2007).

Geological reservoirs are commonly complex both structurally and stratigraphically, and can respond to CO₂ injection both geochemically (inducing dissolution/precipitation reactions especially important in the presence of carbonates which can lead to porosity reduction (Gaus 2010)) and geomechanically (induced pore pressure build-up resulting in dilatations in loose sand formations, or fractures in tight reservoir formations (Rutqvist 2012)). When injected, CO₂ (whether gas, liquid or supercritical) may trigger various physical phenomena as a result of pressure and temperature gradients, and chemical disequilibria (Gaus 2010). These coupled phenomena, so called Thermal-Hydro-Mechanical-Chemical coupled processes (THMCs), need to be modelled accurately using 3D earth models of storage sites constrained by field and laboratory data, as stated in the European Directive 2009/31/EC. In this regards, many laboratory tests have been carried out

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7 since the early days of CCS using representative rock samples of the reservoir site
8 target formation to further analyse and interpret the different trapping mechanisms
9 related to injection-storage issues (Burnside and Naylor 2014). Because seismic
10 surveys represent the most widespread technique to map and monitor the advance
11 of the subsurface CO₂ plume (Chadwick *et al.* 2010; Chadwick *et al.* 2004; Xue and
12 Lei 2006), the experimental works focus preferentially on the analysis of seismic
13 properties of reservoirs from elastic wave propagation through water-CO₂ saturated
14 rock samples (Chen *et al.* 2013; Lei and Xue 2009; Shi *et al.* 2007; Siggins *et al.*
15 2010; Xue and Lei 2006; Xue and Ohsumi 2004), but also on the hydrodynamic
16 behaviour of the two phase solution (Akbarabadi and Piri 2013; Krevor *et al.* 2012;
17 Levine *et al.* 2013; Perrin and Benson 2010) to analyse CO₂ spreading efficiency.
18 However, few experimental works combining simultaneous analysis of different
19 properties have been performed, such as linking geophysical properties with
20 physico-chemical reactions (Canal *et al.* 2013; Hangx *et al.* 2010), hydrodynamic
21 behaviour (Alemu *et al.* 2013; Berg *et al.* 2013; Nakatsuka *et al.* 2010), or
22 mechanical responses associated with water-CO₂-rock interactions (Hangx *et al.*
23 2013; Hangx *et al.* 2010; Vialle and Vanorio 2011; Zemke *et al.* 2010).

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40 CO₂ storage simulations in the laboratory are commonly addressed by pore
41 fluid substitution tests under real reservoir conditions (i.e., temperature, and
42 confining and pore pressures) with the aid of special high pressure, high temperature
43 rigs. The experimental rigs combine diverse instrumentation to monitor pore fluid
44 evolution with other systems to measure changes in the original properties of the
45 rock (e.g., permeability or porosity). The most usual case of study is an originally
46 brine saturated rock sample that is subjected to CO₂ or CO₂-brine mixture flow. This
47 sort of flooding test can be carried out using either steady or unsteady state flow
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7 techniques (Müller 2011). X-ray computed tomography (CT) is the preferred tool to
8 accurately measure CO₂ partial saturations (Berg *et al.* 2013; Krevor *et al.* 2012;
9 Nakagawa *et al.* 2013; Oh *et al.* 2013; Perrin and Benson 2010), which is crucial to
10 fit relative permeability curves or calibrate poroelastic numerical models (Mavko *et*
11 *al.* 2009). However, electrical resistivity tomography (ERT) appears as an alternative
12 method to calculate CO₂-brine saturations (Alemu *et al.* 2013; Wang *et al.* 2009)
13 because of the great electrical conductivity difference between CO₂ and brine. In
14 contrast to CT, electrical resistivity has been successfully applied *in situ* at real CO₂
15 sites (Carrigan *et al.* 2013).
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24 To address changes in reservoirs based on the THMC concept, we have
25 designed a unique multidisciplinary plan which combines experimental testing of rock
26 samples exposed to CO₂ injection under realistic environmental and geological
27 conditions with state-of-the-art data analysis and interpretation. The experimental
28 procedure is based on coupling geophysical and hydromechanical monitoring,
29 controlling real P-T (pressure-temperature) reservoir conditions and fluid
30 composition. Here, we present a demonstration of our novel experimental rig by
31 simulating variable pore pressure conditions in shallow, low permeable reservoirs
32 (e.g., the sort of Svalbard pilot-scale CCS project (Farokhpoor *et al.* 2014; Senger *et*
33 *al.* 2015)) at target depths of c. 900 m and a porosity of 25.9%. Accordingly, we
34 conduct a drainage flooding test through synthetic brine saturated sandstone
35 changing the brine-CO₂ fraction of pore fluid while simultaneously evaluating
36 ultrasonic wave velocity and attenuation, electrical resistivity, axial strains and
37 relative permeability, under variable injection induced pore pressure scenarios.
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2 Materials and Methods

2.1 Sample core plug

Synthetic sandstones represent ideal reservoir samples that are unaffected by micro-structural damage associated with the unloading process that any core suffers when extracted from deep geological formations (Lei and Xue 2009). Hence, we used in this study a 5 cm diameter, 2 cm length, brine-saturated synthetic silica-cemented sandstone. The sample was made according to the manufacturing process presented by Tillotson *et al.* (2012). The mineralogy estimated via X-ray diffraction for this process is >90% silica (Tillotson *et al.* 2012). The porosity was determined by helium injection porosimetry resulting in 0.259, which converted into a pore volume of $\sim 10 \text{ cm}^3$.

2.2 Experimental rig

Figure 1 shows a schematic diagram of our experimental rig. It is an upgraded version for two fluids injection of the rig presented by Falcon-Suarez *et al.* (2014). The rig is designed around a triaxial cell core holder that allows rock samples to be subjected to confining pressures up to 69 MPa. The confining fluid (mineral oil) is delivered from a dual pumping controller Teledyne ISCO model EX100D, configured in a non-coupled mode to independently set axial and radial target confining stresses. Inside the triaxial cell, the rubber sleeve that isolates the core plug from the confining fluid is equipped with 16 electrodes for electrical resistivity tomography measurements (ERT, see North *et al.* (2013)); strain gauges (350 Ohm) are also added on the sleeve-wall to measure axial and lateral strains during the test. Both signals are extracted via feedthrough connectors. The sample is indirectly in contact with the ultrasonic pulse-echo instrumentation (two buffer rods in between, see below) for measuring the ultrasonic velocity and the ultrasonic attenuation (Best *et*

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7 *al.* 2007). As explained on the inner cell zoom drawing of Figure 1, the core plug is
8 isolated from the rest of the rig and the ultrasonic transducer by two acrylic buffer
9 roads. These buffer rods have well defined acoustic impedance and low energy loss,
10 providing a reliable delay path to enable the identification of top/base sample
11 reflections for calculating wave velocities and attenuations. The buffer rods
12 implement pathways (inlet and outlet ports) to conduct the pore fluid through the
13 sample. The surfaces of the buffer rods in contact with the sample are specially
14 designed with circumferential fluid flow pathways to facilitate the fluid spreading
15 upstream and the collection downstream.
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24 Likewise the confining pressure, pore fluids are transferred using a dual
25 pumping controller Teledyne ISCO model EX100D configured in an independent
26 mode. An additional single controller Teledyne ISCO model ED100 is used to
27 provide back pressure to accurately control the pore pressure downstream of the
28 sample. The pore fluids are indirectly supplied via fluid transfer vessels (FTVs). The
29 aim of these vessels is twofold: firstly, to prevent potential damage to the controller
30 from the high corrosiveness of brine and CO₂; and secondly, to heat the fluids to
31 target conditions by direct immersion of the FTVs in a thermal bath (up to 100 °C).
32 As a result, three vessels are connected to three pumping controllers: two for
33 transferring brine and CO₂ into the sample, and one for receiving the resulting fluid
34 while setting the pore pressure constant. To further quantify the pore pressure, two
35 piezo-resistive pressure transmitters (Keller model PA-33X) are inserted up- and
36 downstream in the hydraulic system to accurately measure pressure, but also the
37 temperature changes during the test. The remaining part of the experimental rig
38 outside the thermal bath is enclosed in a commercial greenhouse cover system
39 dotted with heat fans and automatic controllers to keep constant the temperature of
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7 the rig. Additionally, the triaxial cell is also wrapped with an electrical rope heater
8 connected to a PID (proportional/integral/derivative) controller (maximum
9 temperature 150 °C; accuracy ± 0.5 °C) which set the target temperature in the
10 vessel according to the information received from an inner-vessel thermocouple.
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14 15 2.3 Test methodology

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17 The experiment was a steady state drainage test (Müller 2011), in which an
18 electrical resistivity method was used to observe the fluid distribution in the porous
19 medium. The sample was saturated first in degassed 35 g L⁻¹ NaCl-brine via water
20 imbibition in a vacuum vessel. The assumption of saturation was based on the fact
21 that the porosity according to water-weight was less than 0.6% lower than the one
22 obtained from helium porosimetry.
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29 Once inside the cell, the sample was hydrostatically confined (σ_c) at 16.5 MPa
30 (simulating ~900 m depth “shallow” reservoir conditions) and the pore pressure (P_p)
31 set at 8.2 MPa using the backpressure pumping controller downstream. The pore
32 fluid was injected from the bottom end of the sample to remove remaining air
33 bubbles from the pore space, at a constant flow rate. To avoid alterations of sample
34 properties due to overloading while reaching the initial conditions of the test, the
35 confining and pore pressures were simultaneously increased keeping the effective
36 pressure ($P_{eff} = \sigma_c - P_p$) lower than the test-starting target conditions (i.e., $P_{eff} = 8.3$
37 MPa). The temperature was set at 35 °C to be above the critical point of CO₂ (i.e.,
38 31°C, 7.39 MPa). After 4 days of compaction and settlement (geomechanical
39 stabilisation), the sample was subjected to a 0.25 ml min⁻¹ brine-flow, which is an
40 accepted value for field flow rates (Berg *et al.* 2013), while setting the pressure
41 downstream to 8.2 MPa. Thereafter, an unload/loading sequence of effective
42 pressure was reproduced by increasing the pore pressure downstream 1.2 MPa
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7 stepwise from 8.2 MPa to 11.8 MPa and back to 8.2 MPa, while keeping the
8 confining pressure constant at 16.5 MPa. For each step, time was taken to allow a
9 minimum of one pore volume (PV) to circulate through the sample while pore
10 pressure upstream and strains were continuously recorded. Additionally, at the end
11 of each step, electrical resistivity and ultrasonic measurements were collected.
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17 After the first flooding sequence, the initial 100% brine solution was replaced by
18 brine-CO₂. The new solution injected into the sample was obtained by mixing
19 variable volumetric flow rates of brine and CO₂ (differential fractional flows)
20 independently set by the corresponding controller. The resulting flow (Q) was kept
21 constant at 0.25 ml min⁻¹ for all the fractional brine:CO₂ flows that were used (i.e., Q
22 = $Q_w + Q_{CO_2}$). The methodology is similar to that proposed in previous water-CO₂
23 flooding studies (Akbarabadi and Piri 2013; Krevor *et al.* 2012; Perrin and Benson
24 2010): both fluids meet at certain point in the high pressure hydraulic network,
25 flowing together thereafter along 12 m length, 1.6 mm-ID-pipe (which takes a time of
26 ~2 h at the experimental flow rate) before reaching the sample. The assumption of
27 brine-CO₂ equilibrium is based on the results shown by (El-Maghraby *et al.* 2012).
28 They demonstrated that for a one litre vessel containing brine-CO₂ at similar P-T
29 conditions to those used in the present study (CO₂ at supercritical state), the time
30 needed to achieve the equilibrium between both phases was approximately one
31 hour. We assume this time should be enough to equilibrate the fluid in our pipe-
32 reservoir system, since the cross section, and also the total volume displaced, are at
33 least two orders of magnitude lower than those used by El-Maghraby *et al.* (2012).
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50 This experimental procedure was repeated six times, varying 20% stepwise the
51 fractional brine:CO₂ flow up to 100% CO₂ (final stage). Between two consecutive
52 unload/loading sequences, the existing pore fluid was replaced by the new solution.
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7 So, before starting the measurements, the new solution was forced to flow through
8 the sample at the initial P_p of the sequence (8.2 MPa) until the measured electrical
9 resistivity stabilised (in all cases, no less than 4 PVs).
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12 13 2.4 Ultrasonic measurements

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15 Ultrasonic P- and S-wave velocities (V_p and V_s) and attenuations (inverse
16 quality factors Q_p^{-1} and Q_s^{-1}) were measured using the pulse-echo technique
17 (McCann and Sothcott 1992), which provides useable frequencies between 300 -
18 1000 kHz with absolute accuracies of $\pm 0.3\%$ for velocity and $\pm 0.1 \text{ dB cm}^{-1}$ for
19 attenuations (for 2.54 cm diameter single mode transducers); here we used dual P/S
20 wave transducers which give accuracies of $\pm 0.3\%$ for velocity and $\pm 0.2 \text{ dB cm}^{-1}$ for
21 attenuation (Best 1992). P- and S-wave velocities and attenuations were measured
22 air dry, brine saturated, and for increasingly CO_2 saturated brine solutions
23 corresponding to the pore fluid composition imposed by each fractional brine: CO_2
24 flow. For every single fluid composition, the monitoring parameters covered each
25 unload/loading effective pressure step.
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36 37 2.5 Electrical resistivity, partial saturation and pore fluid

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39 Electrical resistivity was measured with a 16 electrodes, tetra-polar electrode
40 configuration radially distributed in two rings around the plug, capable of assessing
41 heterogeneity and anisotropy without previous information of sample properties
42 (North *et al.* 2013). Under typical operating conditions the resistivity measurement
43 error is $\sim 5\%$ (at frequencies 1 - 500 Hz) for samples in the electrical resistivity range
44 1-100 $\Omega \text{ m}$. Detailed information about system configuration and the data processing
45 can be found in North *et al.* (2013) and North and Best (2014). The data acquisition
46 was systematically performed immediately after each ultrasonic measurement,
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7 obtaining comparable values between ultrasonic parameters and electrical resistivity.

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9 The raw data for every single acquisition resulted in a total of 208 measurements,
10 from which an electrical resistivity average value and the inversion model were
11 calculated. While tomography images from the inversion were used to visualise
12 distribution patterns of pore fluid across the sample, the electrical resistivity average
13 was used to compare results against other variables and also to determine the
14 partial saturation of brine:CO₂ in the sample.
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20 The degree of saturation of brine (S_w) is related to the bulk electrical resistivity
21 (ER_b) of a rock through the connectivity of the porous medium and the electrical
22 resistivity of the pore fluid (ER_w) according to Archie's law (Archie 1942):
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$$ER_b = \frac{ER_w}{\varphi^m S_w^n} a \quad (1)$$

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27 where φ is the porosity, a is a proportionality constant, m is the cementation factor of
28 the material and n the saturation exponent. The latter three parameters are
29 empirically adjusted or assumed for a particular type of rock. In our experiment, the
30 initial stage is 100% brine saturation so that the initial bulk electrical resistivity (ER_0)
31 can be expressed as:
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$$ER_0 = \frac{ER_w}{\varphi^m} a \quad (2)$$

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41 The experimental test is a two-phase solution system, hence the evolution of the
42 brine or CO₂ saturation (i.e., $S_w = 1 - S_{CO_2}$) can be obtained dividing equation (1) by
43 equation (2) (Carrigan *et al.* 2013; Nakatsuka *et al.* 2010):
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$$S_w = \left(\frac{ER_0}{ER_b} \right)^{1/n} \quad (3)$$

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7 In our unload/loading sequence test ER_b varies by the effect of compaction. So,
8 we calculated ER_0 for each effective pressure of each brine-saturation stage,
9 adopting the corresponding values thereafter to solve equation (3). Nonetheless, it is
10 worth noting that this expression is only valid if ER_w remain constant with time, which
11 is the case of a chemically non-reactive scenario. Otherwise, $S_w - ER_b$ relations
12 should be addressed using equation (1). In this regard, the pore fluid was sampled at
13 the end of each fractional brine:CO₂ flow stage, and ER_w determined with a
14 commercial conductivity metre (up to 19.99 mS cm⁻¹). The results were surprisingly
15 constant and close to the original brine (0.189 Ω m): $ER_w = 0.191 \pm 0.009$ Ω m.
16 Since no preferential trends were identified in the ER_w , we assumed the flooding test
17 was chemically stable and adopted equation (3) to calculate the changes in the
18 degree of saturation. The latter assumption is supported by the fact that no porosity
19 changes were identified after the test.
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32 *2.6 Absolute and relative permeability*

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34 By changing the fractional brine:CO₂ flow, we determined the relative
35 permeability curves for both phases at the simulated reservoir conditions. We used
36 the steady state relative permeability method of Müller (2011). As a first step, the
37 sample permeability to brine was calculated for every P_{eff} step at the initial brine:CO₂
38 stage (100:0), using the well-known steady state flow method based on Darcy's law:
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$$44 \quad k = \frac{\mu L Q}{\Delta P A} \quad (4)$$

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47 where k is permeability, Q the volumetric flow rate, ΔP the pressure drop across the
48 sample, A the cross sectional area, L the length, and μ the dynamic viscosity of the
49 fluid. We found that the equilibrium was achieved rapidly after a flow ~ 0.1 PVs. This
50 represents an advantage of our experimental setup related to the sample size (~ 2 cm
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length), respect to the rest of experimental configurations found in the relevant literature (for instance Akbarabadi et al. 2013 and Berg et al. 2013, use ~15 cm length samples; Krevor et al. 2012, Alemu et al. 2013, Kitamura et al. 2014, ~10 cm; Canal et al. 2013 and El-Maghraby and Blunt 2013, ~7.5 cm length; Chen et al. 2013, ~7 cm length; and Chang et al. 2013, ~ 5 cm length samples). Despite this, the minimum volume circulated during every step of the test was 1 PV.

Assuming an homogenous contribution of the whole cross sectional area to the flow through the entire core, the above expression can be modified for our two-phase brine-CO₂ system in which the relative permeability of each phase ($k_{r,i}$) is a function of the partial contributions to the total saturation (Krevor *et al.* 2012; Levine *et al.* 2013):

$$k_{r,i}(S_i) = \frac{\mu_i L Q_i}{\Delta P A k_{abs}} \quad (5)$$

Note in the above expression that subscript i refers to each phase. Since permeability clearly depends on the effective pressure (Nguyen *et al.* 2013), in the present work we used the maximum permeability measured during the first effective pressure sequence as the absolute permeability (i.e., $k_{abs} = 1.01$ mD), in order to compare the magnitude of these changes. For permeability calculations, CO₂ viscosity was determined according to Heidaryan *et al.* (2011) while brine viscosity was taken from Mavko *et al.* (2009).

Relative permeability curves with the Brooks-Corey model (Brooks and Corey 1964) for a brine-CO₂ two phase system ($k_{r,w}$ and k_{r,CO_2}) and the fractional flow curve (f_{CO_2}) were performed as presented in Krevor *et al.* (2012):

$$k_{r,w} = (S_w^*)^{N_w} \quad (6)$$

$$k_{r,CO_2} = k_{r,CO_2}(S_{wi})(1-S_w^*)^2(1-(S_w^*)^{N_{CO_2}}) \quad (7)$$

$$S_w^* = \frac{S_w - S_{wr}}{1 - S_{wr}} \quad (8)$$

$$f_{CO_2}(S_{CO_2}) = \frac{Q_{CO_2}^{T,P}}{Q_{CO_2}^{T,P} + Q_w^{T,P}} \quad (9)$$

where N_w and N_{CO_2} are the Corey fitting-components for brine and CO_2 . In absence of data of irreducible water saturation (S_{wr}), this parameter was considered an additional fitting parameter in the modelling.

2.7 Strains

Sample deformation was monitored from electrical strain gauges added to the wall of the rubber sleeve within the triaxial vessel, and additionally from the axial piston displacement proportional to the confining fluid injected from the servo-controller volumetric pump (Figure 1). Radial strain gauges were likewise added on the sleeve, but unfortunately the signal recorded was inaccurate due to the high compressibility of the thick rubber sleeve. Before and after the test, the results were first calibrated with a standard (5 cm diameter, 2 cm height aluminium sample). Then, a more recent modification of our triaxial cell allowed us to repeat an additional post-experiment calibration by directly adding strain gauges on the wall-side of the sample (water-saturated), covering the complete sequence of P_{eff} . In Figure 2 we show the correlation between the deformation recorded by the gauges added on the sample (G_{sa}) and those added on the sleeve (G_{sl}).

During the flooding test, axial strains were measured with a sampling frequency of 0.08 Hz during brine: CO_2 stages, covering the whole range of P_{eff} and compositional steps. Then, only axial strains (ε_{ax}) were considered and processed

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7 (the correction factor of Figure 2 was applied) in terms of relative longitudinal
8 deformation according to equation (10):
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$$\varepsilon_{ax} = \frac{(L_0 - L)}{L_0} \cdot 100 \quad (10)$$

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15 where L_0 and L are the original and measured sample length, respectively. In the
16 absence of radial strains, relative changes of porosity during the test were neglected.
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18 19 **3 Experimental results**

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21 The effective experiment extended ~50 hours. During this period, 67 PVs at
22 different brine:CO₂ fractions circulated through the sample. Figure 3 shows the raw
23 data obtained during this drainage test. The evolution of P- and S-wave velocities
24 and their corresponding attenuations (frequency of 600 kHz obtained from Fourier
25 analysis of broad band signals), axial strain, relative permeability of each fluid phase
26 and the electrical resistivity of the rock sample is plotted together for each of the six
27 brine:CO₂ unload/loading effective pressure sequences. For all of these measured
28 parameters, Figure 3 shows that pore fluid effects are greater than effective pressure
29 effects since transitions between sequences lead to more significant changes than
30 those observed within a single sequence.
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41 During the transition stages between two consecutive brine/CO₂ episodes
42 (indicated by grey vertical bands), both electrical resistivity and ultrasonic wave
43 velocities were computed for every 1 PV flushed through the sample. Although these
44 data are not presented in this study, they were used to select the starting point of a
45 new fluid stage upon data stabilisation. Even though this stabilisation occurred very
46 quickly in some cases, we imposed a minimum flow of 4 PVs before concluding the
47 transition. This would be enough to reach an homogeneous fluid distribution in a 2
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7 cm length core sample, based on the results presented by (Oh *et al.* 2013) for a
8 similar flooding test.
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10 11 3.1 Geophysical signatures

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13 Figure 3 shows P-wave velocity (V_p) gradually decreases with the CO₂
14 saturation, by ~7% at the end of the test. The highest drop (~4%) rapidly occurs after
15 the first fluid change (brine:CO₂ = 80:20), which agrees with the data reported by
16 other authors (Alemu *et al.* 2013; Kitamura *et al.* 2014; Lei and Xue 2009; Shi *et al.*
17 2007; Xue and Ohsumi 2004). At higher CO₂ contents, V_p becomes less affected by
18 fluid changes, coinciding with the results presented by (Alemu *et al.* 2013). The
19 same effect can be seen for the S-wave velocity (V_s), but after the first drop (~4%)
20 the velocity becomes steady and mechanical variations become more significant. P-
21 and S-wave attenuations (expressed by the inverse quality factors Q_p^{-1} and Q_s^{-1} ,
22 respectively) show the opposite behaviour: Q_p^{-1} increases with CO₂ content (up to
23 ~55%) evolving inversely comparing to V_p ; Q_s^{-1} decreases (~25%) with an inverse
24 trend compared to V_s caused by P_{eff} unload/loading variations. Hence, according to
25 the results, V_s and Q_s^{-1} are better geomechanical indicators, while V_p and Q_p^{-1} can
26 provide useful information about pore fluid compositional changes.
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41 The electrical resistivity (ER) increases from ~12 Ω m for pure brine to ~45 Ω m
42 during the last stage, where the highest CO₂ saturation was achieved. While Figure
43 3 shows average resistivities, Figure 4 shows 3D electrical resistivity tomography
44 images of the sample, corresponding to the minimum P_{eff} step (i.e., 4.7 MPa) of each
45 brine:CO₂ stage. We only show one P_{eff} step per brine:CO₂ stage because changes
46 between steps within a single stage are visually negligible. The stacked images
47 correspond to sections at different heights through the sample, centred on the one in
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7 the middle. The radial distribution of electrical resistivity can be seen to become
8 increasingly heterogeneous with increasing CO₂ saturation.
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11 Joint interpretation of acoustic and electrical properties is a fundamental step
12 towards identifying and understanding reservoir changes and improving prediction
13 models (Han *et al.* 2011; Han *et al.* 2015). We compare the ultrasonic wave
14 velocities and electrical resistivity data of our drainage test in Figure 5. V_p decreases
15 with ER , which is in line with reported observations (Wang *et al.* 2009), while V_s
16 initially drops but then stabilizes. This is clearly showing the differential repercussion
17 the fluid has on V_p . Plotting velocities versus electrical resistivity smooths the effect
18 of the effective pressure on the data to better assess changes in the fluid
19 composition, which is intensified through the V_p/V_s ratio (Figure 6). Likewise, the
20 variations of the P- and S-wave attenuations with the electrical resistivity are shown
21 in Figure 7 and Figure 8, respectively. Q_p^{-1} increases with the electrical resistivity, the
22 opposite to Q_s^{-1} , making the Q_s/Q_p ratio less diagnostic than V_p/V_s ratio for
23 interpreting pore fluid changes. Nevertheless, this ratio provides useful information
24 regarding the conversion of low resistivity values into brine/CO₂ saturation because
25 Q_s/Q_p greater than unity means $S_w < 0.9$ (Amalokwu *et al.* 2014; Murphy 1982),
26 which occurs at resistivity values above 18 Ω m.
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42 3.2 Sample deformation

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44 In Figure 3, the axial strain evolves within each effective pressure sequence as
45 expected, i.e., recovering initial deformation during unloading to be newly gained
46 while loading, oscillating less than 0.04% for every single sequence. The
47 deformation carries hysteresis, in so far as after the loading ramp the final value is
48 slightly higher than the original one for a single sequence. From the stage brine:CO₂
49 60:40 onwards, the hysteresis effect is stronger, which is also reflected in the V_p
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7 values. Nonetheless, the sample behaves quasi-elastically for a single sequence of
8 effective pressure with a slight degree of cumulative damage, but reaching ~0.06%
9 of axial deformation after flow times of 60 PVs. The data agree reasonably well with
10 the values reported by Xue and Ohsumi (2004) and Lei and Xue (2009) for similar
11 experimental conditions, and Hangx *et al.* (2013) who showed strains approximately
12 one order of magnitude higher, but from poorly consolidated carbonate- and quartz-
13 cemented samples.
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20 21 3.3 Relative permeability

22 The relative permeability of both phases naturally increases with their
23 respective partial saturations. Permeability and axial strain show opposite trends for
24 a single brine:CO₂ stage, indicating that the connectivity of the pores varies with
25 effective pressure, with the exception of the last stage of brine:CO₂ as mentioned
26 above. The variation of pore connectivity is also reflected in the electrical resistivity
27 of the rock for a single unload/loading effective pressure sequence; however, this
28 parameter is primarily dependent on the electrical resistivity of the pore fluid, so that
29 mechanical effects are very small compared to fluid changes. Nevertheless, to
30 minimise the mechanical effect and emphasise the fluid dynamics, the permeability
31 measured at each P_{eff} step during the initial pure brine flooding stage was used as
32 the particular absolute permeability for the same steps at the subsequent brine:CO₂
33 stages. Resistivity measurements carry an error ~5%. This has only a small impact
34 on the calculation of the degree of saturation if brine saturation is high, but increases
35 with CO₂ saturation up to 5%.
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50 Brine saturation (i.e., $S_w = 1 - S_{CO_2}$) was computed from the average value of
51 electrical resistivity, using equation (3). Accordingly, the maximum CO₂ saturation
52 achieved during the experiment was $S_{CO_2} \sim 0.5$, which is likewise indicative of the
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7 brine that CO₂ is able to displace under the simulated test conditions. In other
8 words, this value indicated the maximum CO₂ storage capacity of an idealised
9 reservoir (Kitamura *et al.* 2014). The capillary number $N_c = v\mu/\sigma$ (where v is Darcy
10 velocity in m s^{-1} , μ viscosity in Pa s, and σ the interfacial tension in N m^{-1}), was
11 calculated to obtain the magnitude of the relation between viscous and pore scale
12 capillary forces. In our case $N_c \sim 10^{-10}$, but it has been proposed that N_c only affects
13 relative permeability estimations if $>10^{-6}$ (Akbarabadi and Piri 2013; Krevor *et al.*
14 2012). Figure 9 shows the relative permeability curve fits to our experimental data.
15 Since electrical resistivity slightly varies as a result of P_{eff} changes, we address the
16 study of the relative permeability from a global plot without considering the
17 differential mechanical effects. The best-fit Corey exponents for brine and CO₂ were
18 found to be $N_w = 7$ and $N_{CO_2} = 0.5$, and $S_{wr} = 0.15$, while the maximum $k_{r,CO_2} = 0.05$
19 (at $S_{CO_2} = 0.5$). The value obtained for k_{r,CO_2} might be underestimated because,
20 according to (Krevor *et al.* 2012), when 100% CO₂ is injected the steady state flow
21 method stabilises and the observed pressure gradient is related to the capillary
22 pressure rather than to the permeability. Pini *et al.* (2012) presented an experimental
23 method based on changes of flow rates to obtain the permeability through the slope
24 of the flow rate versus the pressure gradient to calculate the permeability at this
25 point. We did not apply it during this test, but it will be carried out in future
26 experiments.

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45 Furthermore, the inner plot in Figure 9 shows the fractional flow to CO₂
46 saturation. The experimental data points fit well the theoretical estimation.
47 Nevertheless, for low fractional flow stages the observed S_{CO_2} is lower than
48 predicted. This indicates the low influence of CO₂ on the resistivity at such high brine
49 saturations, an issue that will be discussed in the following section. In this regard, we
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have included one unique cross error in Figure 9 to facilitate the visualization: horizontally, we show the error associated with the electrical resistivity transferred to the calculated degree of saturation (also both horizontal and vertical error bars in the inner plot); vertically, the error regarding the influence of the neglected effect of dissolved CO₂ in the relative permeability. The latter error is calculated based on the data presented by Berg et al. (2013) for very similar experimental conditions to the present work. According to that work, the solubility of CO₂ in water is about 1.79mol%, i.e., up to 9 vol% dissolved CO₂ into the aqueous phase. So, our CO₂ flow rate would be likewise overestimated and therefore the relative permeability.

4 Discussion

4.1 Comparison to predictions by Gassmann's model

The theoretical modelling of changes in our ultrasonic data as a result of fluid substitutions was primarily addressed using the Gassmann equation (Gassmann, 1951). Based on the concept of poroelasticity, Gassmann's model predicts variations in elastic wave velocities V_p and V_s in saturated porous media as follow:

$$V_p = \left(\frac{K_b + \frac{4}{3}G_b}{\rho_b} \right)^{1/2} \quad (11)$$

$$K_b = K_d + \frac{(1 - K_d / K_m)^2}{\varphi / K_f + (1 - \varphi) / K_m - K_d / K_m^2} \quad (12)$$

where K_b and G_b the bulk modulus and the shear modulus of the rock of bulk density ρ_b and porosity φ . As shown in equation (12) K_b is related to the dry rock frame modulus, solid mineral bulk modulus and the fluid bulk modulus (subscripts d , m and f , respectively). As G_b is unaffected by the fluid saturation, then $G_b = G_d = \rho_b V_s$. In

our two phase system, the bulk density is expressed as follows to address the effect of each fluid on the total ρ_b value:

$$\rho_b = (1 - \varphi) \rho_m + \varphi \rho_f \quad (13)$$

$$\rho_f = \rho_w (1 - S_{CO_2}) + \rho_{CO_2} S_{CO_2} \quad (14)$$

Note that bulk modulus subscript w and CO_2 refers to brine and CO_2 (i.e., $S_w = 1 - S_{CO_2}$). Likewise, as described in Kitamura *et al.* (2014) or Chen *et al.* (2013) and based on Woods' fluid mixing law, the bulk modulus of the brine- CO_2 solution is calculated as follow:

$$K_f^{-1} = \left(\frac{K_{CO_2}}{S_{CO_2}} \right)^{-1} + \left(\frac{K_w}{S_w} \right)^{-1} \quad (15)$$

In the equation above, the modulus of each component was calculated from the relations presented by Batzle and Wang (1992), for each of the P-T experimental conditions. See details of calculation parameters on Table 1.

Figure 10 and Figure 11 show experimental data and Gassmann estimates for P- and S-wave velocity. Initial experimental velocities obtained during pure brine flow (i.e., 100% brine saturation) are higher than the Gassmann predictions, an effect that can be attributed to additional dispersion mechanisms due wave-induced local fluid flow mechanisms not considered in Gassmann's model (Winkler 1985; Chapman *et al.* 2002; Sarout 2012). Thereafter, Gassmann predictions fits better to V_p data at high and very low CO_2 saturations; between 10 - 20% CO_2 saturation, the model underestimates V_p by ~3.5% on average. While no clear $V_p - P_{eff}$ relations are observed between experimental data and predictions, in the case of V_s , the same trend is observed for every single effective pressure, although Gassmann's model

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7 overestimates V_s by $\sim 2\%$ in all cases. Since V_s is relatively unaffected by the fluid, it
8 is a good indicator of the geomechanical state of the rock, although we observe V_s
9 drops 6% between brine saturated and 20% CO_2 saturation, possibly related to local
10 flow fluid mechanisms which is corroborated by V_p data (e.g., Winkler and Nur 1982).
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12 Nonetheless, V_p depends on the bulk modulus of the rock, and also the fluid bulk
13 modulus. Hence, the dispersion of the data may suggest non-homogeneous
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15 distribution of the fluid in the pores of the rock sample.
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20 4.2 Comparison to predictions by White and Dutta-Ode' model

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22 To investigate the effect of sub-core scale heterogeneities on the fluid
23 distribution pattern (patchy partial saturation), we used the model of White and
24 Dutta-Ode' (see Mavko *et al.* (2009)). The model is based on the assumption that in a
25 partially saturated porous medium, the pore space is occupied by a single fluid which
26 hosts spherical regions of a secondary fluid with contrasting bulk compressibility, i.e.,
27 in the present case brine hosting CO_2 . The idealised patchy spheres (each) have a
28 radius denoted b and a for brine and CO_2 spheres respectively, giving $S_{\text{CO}_2} = (a/b)^3$.
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30 Furthermore, to analyse the two fluid saturation states it is also necessary to
31 introduce the critical diffusion length parameter $L_c = (kK_w/\lambda\mu)^{1/2}$, where k is the
32 permeability, λ is the ultrasonic frequency, and K_w and μ the fluid bulk modulus and
33 viscosity of the brine, respectively. L_c is an indicator of whether the two phases in the
34 porous medium are well distributed ($L_c \sim 0.01$ cm at our experimental conditions).
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36 That is, homogenous saturation exists if the fluids are mixed at a scale length $< L_c$
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38 and a heterogeneous (or patchy) saturation if $> L_c$.
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50 The effect of fluid modulus reduction on the overall rock-fluid bulk modulus
51 during increasing CO_2 saturation is analysed in Figure 12a - d, for each of the four
52 effective pressures conditions. The model with $a = 0.01$ cm was found to yield results
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7 consistent with the experimental bulk modulus, which means that b varies between
8 0.3 - 0.07 cm (decreasing with the CO₂ saturation), and hence $L_c \ll$ patch size. At
9 this point, it is worth mentioning that the selected frequency (600 kHz) lies between
10 the cut-off frequencies between which the White and Dutta-Odé model is valid (see
11 Lei and Xue (2009) for details): i) b is at least 10 times lower than either the P- or S-
12 wave length (5 and 3 mm, respectively); and ii) the so called transition frequency
13 (upper limit), the frequency up to which Darcy flow is justified, varies in our case
14 between 900 - 1200 kHz.
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22 In this respect, the results can be linked to the heterogeneities observed from
23 electrical resistivity tomography. Likely, these are associated with higher capillary
24 pressure regions (predominantly larger than the diffusion length at the experimental
25 conditions, but small enough to affect the relative permeability, i.e., $N_c \sim 10^{-10}$)
26 preferentially occupied by the original brine solution (Shi *et al.* 2007; Xue and Lei
27 2006); while CO₂ (of lower density and higher compressibility) would replace brine
28 from higher pore size regions. Furthermore, a closer view of Figure 12a - c reveals
29 additional details about pore fluid heterogeneous distribution. Notice how the data
30 related to unloading (solid circles) are slightly closer to patchy saturation than those
31 of loading (open circles). This could be indicating a slight tendency to homogeneous
32 saturation during loading, maybe related to fluid movements between pores by
33 squeezing.
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45 The P-wave attenuation (inverse quality factor) observations shown in Figure
46 13a - d are much larger than the model predicted values, which agrees with results
47 previously reported (Amalokwu *et al.* 2014; Nakagawa *et al.* 2013). Figure 13a - d
48 show Q_p^{-1} experimental data with White and Dutta-Odé predictions for the whole
49 saturation range. The model predicts a rapid increase in P-wave attenuation with a
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7 peak at ~25% of CO₂, followed by a gradual decrease towards higher CO₂
8 saturation. Our data lie far above the predicted values both under unloading or
9 loading, but follow an increasing trend from initial CO₂ saturation states up to peak
10 values at maximum experimental CO₂ saturations; unlike the model predictions,
11 there is no evidence for a drop off at higher CO₂ saturations.
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17 As expected, the replacement of the initial brine by brine-CO₂ solution had a
18 striking effect on both the ultrasonic wave velocity and attenuation. Although several
19 damping mechanisms have been proposed to describe these phenomena (Müller *et*
20 *al.* 2010; Lei and Xue 2009), the homogeneity of our synthetic sample, the low
21 deformation observed during the test, and the invariable porosity and pore fluid
22 electrical conductivity (i.e., no geochemical effects), suggest that viscous fluid flow
23 mechanisms are the cause. Attenuation observations are especially important to
24 explain changes above 20% CO₂ saturation, when P-waves become less sensitive to
25 fluid changes. The White and Dutta-Odé model could explain in part the observed
26 attenuations, at least the trend of one limb of the model curve, but the higher
27 measured attenuations suggest the presence of other loss mechanisms, as yet
28 unidentified. However, bulk modulus appears to be a good indicator of the rock-fluid
29 state, even showing some sort of squeezing phenomenon related to loading, as
30 previously mentioned, that could help to identify potential overpressure reservoir
31 conditions in real CO₂ storage sites.
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45 46 4.3 Ultrasonic data quality assessment 47

48 Regarding the ultrasonic attenuation mechanisms mentioned above, it is
49 worth showing the ultrasonic frequency spectra to check data quality, as in Figures
50 13 a – d, and to confirm the valid frequency ranges for observation of fluid flow loss
51 mechanisms. According to Best (1992), the ultrasonic pulse-echo system as
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7 configured in this work (using a dual P/S wave transducer of effective radius 0.89
8 cm) gives usable P-wave results above 580 kHz for a rock V_p of 4 km s⁻¹ or lower,
9 and above 330 kHz for S-waves for a rock V_s of 2.5 km s⁻¹ or lower. Accuracies are ±
10 0.3% for velocity and ± 0.2 dB cm⁻¹ for attenuation coefficient (about ±10% for $Q =$
11 50; see Best *et al.* (1994)). The upper frequency limits for P- and S-waves are
12 determined by signal to noise ratio, often caused by the onset of Rayleigh scattering
13 as the wavelength approaches the size of any heterogeneities such as sand grains,
14 or in this case, liquid/gas patches. In our case, this is no relevant since we used fine
15 sand to make the synthetic sample, which means the grains (< 75 μm) were over ten
16 times smaller than the wavelength.
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26 In Figure 14a, V_p decreases smoothly (although non-monotonically) with
27 frequency by c. 0.25% between 500 – 800 kHz for all saturations states, but V_s
28 increases up to about 700 kHz (by c. 0.6% from 400 – 700 kHz) before dropping off
29 at higher frequencies for all saturation states (Figure 14b). As the percentage
30 change is small compared to the absolute velocities (but nevertheless significant
31 relative to the measurement accuracy), we will not try to further interpret the
32 underlying causes here. However, as noted above, the attenuation values in Figure
33 13a - d suggest other attenuation mechanisms are present at 600 kHz apart from the
34 White-Dutta-Odé mechanism (at least, as modelled); they could be related to the
35 negative V_p dispersion seen in Figure 13a - d. However, V_s shows “normal” positive
36 dispersion (Figure 14b) as we would expect from squirt flow dispersion mechanisms;
37 the high frequency drop-off is possibly due to poor signal-to-noise ratio.
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50 P-wave attenuation Q_p^{-1} in Figure 14c shows an almost linear increase with
51 frequency between about 500 – 800 kHz; we can ignore values below about 400 kHz
52 as they are probably affected by beam spreading/sidewall reflections. However, S-
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7 wave attenuation Q_s^{-1} is variable in the useable bandwidth (Figure 14d), which is
8 unexplained.
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11 While providing the velocity and attenuation spectra may increase uncertainty
12 in the ultrasonic observations, in fact these data provide a rich source of information
13 on possible loss mechanisms which might be helpful for future interpretations. What
14 is clear is that the ultrasonic velocity and attenuation results are consistent between
15 saturation states, and hence provide confidence in the observed relative changes
16 between saturation states at 600 kHz. The selected frequency of 600 kHz in Figures
17 2, 4 – 7 and 9 – 11 is within the valid measurement range for the pulse-echo system
18 as implemented here.
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26 27 *4.4 Resistivity, saturation degree and relative permeability*

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29 The central part of the sample is in all cases less resistive than the edges,
30 suggesting preferential flow paths or a heterogeneous pore size distribution, but
31 likely a combination of both. Chang et al. (2013) interpreted similar heterogeneities
32 evident from transient dissolved CO₂ concentrations during CO₂ core flooding as
33 areas where local non-equilibrium dissolution is occurring due to differential pore
34 size distribution: smaller pores are occupied by water while larger pores, with lower
35 capillary pressure, are dominated by CO₂. However, since longitudinally the
36 resistivity remains approximately constant, this idea reduces to very local areas.
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38 Similar longitudinal homogeneity of radial heterogeneities were observed by Perrin
39 and Benson (2010) who suggested this indicates the absence of gravity or capillary
40 end effects.
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51 Transformation of electrical resistivity into degree of saturation is crucial to
52 properly couple geophysical signatures and hydromechanical responses during our
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7 experiment. We observed anomalous low resistivity values related to the second
8 brine:CO₂ flow regime (i.e., 80:20 stage; Figure 3), which particularly affect the
9 relative permeability curves and P-wave velocity prediction (Figure 9 and Figure 10).
10 Likewise, the fractional flow represented in the inner plot of Figure 9 shows
11 experimental CO₂ saturations lower than predicted below $S_{CO_2} = 0.2$. We interpret
12 this lack of CO₂ as a dissolution effect. According to Börner *et al.* (2013), CO₂ would
13 increase the electrical conductivity of the brine during dissolution, due to pressure-
14 dependent dissociation processes; however, high conductivity brines as the one we
15 use in this work would mask the dissociation phenomena. Furthermore, instead of
16 increasing, the dissolved CO₂ might decrease the electrical resistivity due to either
17 transformation into carbonic acid or mineral dissolution which would increase the
18 concentration of ionic material especially relevant in the presence of carbonates
19 (Dethlefsen *et al.* 2013). However, our sample is carbonates-free and it remained
20 chemically stable according to the stability of the electrical conductivity of the pore
21 fluid.
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36 Hence, the electrical resistivity record is neglecting the CO₂ dissolved fraction,
37 and the total volume of CO₂ in the pore space is likewise underestimated. This
38 particularly affects the interpretation of the initial stages of the injection but is
39 progressively attenuated with the CO₂ saturation. So for instance, if we focus on the
40 brine relative permeability in Figure 9, the estimations will improve with the
41 displacement of the high brine saturation points to lower values. According to this
42 observation, the electrical resistance method is useful for mapping the CO₂ plume
43 movement in the sub-surface, but it is limited to high concentrations, neglecting the
44 dissolution trapping storage mechanism.
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7 Our results regarding the relative permeability coincide with observations by
8 Perrin and Benson (2010) and later by Krevor *et al.* (2012); despite the absence of
9 any observed capillary end effects or gravity forces, the maximum S_{CO_2} stays below
10 0.5. This threshold is likely to be imposed by the capillary forces of the porous
11 medium, indicating the limit of the storage capacity of the sample. Further, because
12 this study primarily aims at demonstrating the feasibility of the new experimental
13 setup, we decided to remove the sample from the vessel after the 100% CO_2
14 injection stage had finished in order to measure the final brine saturation via wet-dry
15 weight difference, to assess our results. The value obtained was $S_{w,final} = 0.39$, which
16 is ~11% lower than the estimated value of 0.50. This difference could be attributed to
17 gravitational leakage during unloading and disassembly of the apparatus. For future
18 works, we plan to estimate the residual trapping by injecting 100% brine after the last
19 step, to obtain the residual S_{CO_2} , i.e., the remaining CO_2 immobilised in the pore
20 space. This parameter is very important since it gives the remaining CO_2 in the
21 reservoir in a catastrophic reservoir-collapse scenario (Burnside and Naylor 2014).
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35 36 **5 Conclusions**

37 We have conducted a drainage, steady state type, flooding test through a
38 synthetic sandstone core plug under variable effective pressure, in order to simulate
39 a realistic shallow, low permeable CO_2 storage reservoir scenario. Ultrasonic P- and
40 S-wave velocities and attenuations, electrical resistivity, axial strains and relative
41 permeability were simultaneously monitored during the test.
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48 On average, the injection of increasingly CO_2 saturated brine resulted in V_p and
49 V_s drops ~7% and ~4% respectively, while Q_p^{-1} increased ~55% and Q_s^{-1} decreases
50 ~25%. The Gassmann model predictions for V_p are supported by the experimental
51 data at low and very high CO_2 saturations; V_s is ~2% overestimated although
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7 following the predicted trends. The bulk modulus is in good agreement with the
8 White and Dutta-Ode' model. Despite Q_p^{-1} showing higher values than predicted, we
9 can conclude that the observed velocity changes with CO₂ saturation and energy
10 loss are associated with heterogeneous fluid distribution within the sandstone pores
11 medium rather than with internal discontinuities in the sandstone solid skeleton.
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16 Fluid distribution patterns in the core were analysed through electrical resistivity
17 tomography. The mean value was adopted to transform electrical resistivity into
18 water saturation. Heterogeneities observed on tomography images at scales greater
19 than the diffusion length for our experimental conditions corroborate the good
20 agreement to the White and Dutta-Ode' model. However, it is worth taking into
21 consideration that during early stages of CO₂ injection, the electrical resistivity is not
22 reflecting the volume of CO₂ dissolved in the brine, leading to an underestimation of
23 the partial CO₂ saturation.
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32 The steady state method was applied successfully to determine relative
33 permeability during the test. The electrical resistivity allowed us to relate relative
34 permeability to partial saturation, and hence the application of theoretical curves.
35 Additionally, relative permeability followed axial strain trends for every single
36 unloading/loading sequence of effective pressure. Although clearly reflected on P-
37 and S- wave velocities and their respective attenuations, or on the electrical
38 resistivity, geomechanical effects played a secondary role in the present study due to
39 the robust nature of the synthetic sandstone and the shallow conditions of the
40 simulated reservoir. Instead, the experiment was dominated by changes in the pore
41 fluid.
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52 The novel experimental setup presented in this study has allowed us to
53 successfully relate geophysical signatures to hydro-mechanical responses of a
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7 sandstone sample subjected to the injection of CO₂. The increasing CO₂ saturation
8 stages and the oscillating effective pressure have simulated a wide range of
9 scenarios that might occur during CO₂ storage in a shallow, low permeable reservoir.
10 We plan to develop future works covering real conditions of other reservoir types
11 using a similar experimental protocol to facilitate the comparison of results between
12 CO₂ storage sites.
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18 **Acknowledgments**

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20 This work was carried out as part of the DiSECCS project with funding from the
21 United Kingdom's Engineering and Physical Sciences Research Council (EPSRC)
22 (grant EP/K035878/1) and the Natural Environment Research Council (NERC). We
23 also thank Anna Lichtschlag (NOC) for help with pore fluid analysis, Jordi Delgado
24 (University of La Coruña, Spain) for his valuable advices during the design stage of
25 the experimental rig, and Anne-Kari Furre and Joel Sarout for their thorough review
26 and valuable comments that have significantly improved the manuscript.
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Table 1. Modelling parameters: physical properties of the synthetic sandstone and fluids, for the different effective pressures of the test (note that confining pressure is kept constant at 16.5 MPa)

<i>Rock</i>	Effective pressure (MPa)				<i>Reference</i>
	8.3	7.1	5.9	4.7	
K_m (GPa)		36.32			Estimated from XRD
K_d (GPa)	12.56	12.45	12.40	12.43	From dry V_p and V_s
G_d (GPa)	11.02	10.97	10.87	10.72	From V_s
ρ_s (kg m ⁻³)		2592.8			From ρ_d
ϕ		0.259			Helium porosimetry
k (mD)*	0.59	0.63	0.68	0.78	From Darcy's law
<i>Fluid</i>					
K_{CO_2} (GPa)	0.015	0.022	0.031	0.042	Batzle and Wang (1992)
ρ_{CO_2} (kg m ⁻³)	542.8	686.5	732.5	762.9	Batzle and Wang (1992)
μ_{CO_2} (Pa s) · 10 ⁻⁵	3.11	4.15	4.99	5.68	Batzle and Wang (1992)
K_w (GPa)	2.503	2.509	2.516	2.523	Batzle and Wang (1992)
ρ_w (kg m ⁻³)	1021.1	1021.6	1022.1	1022.6	Batzle and Wang (1992)
μ_w (Pa s)		7.4 · 10 ⁻⁴			Batzle and Wang (1992)

* Average of unload/loading values. No hysteresis effects were considered

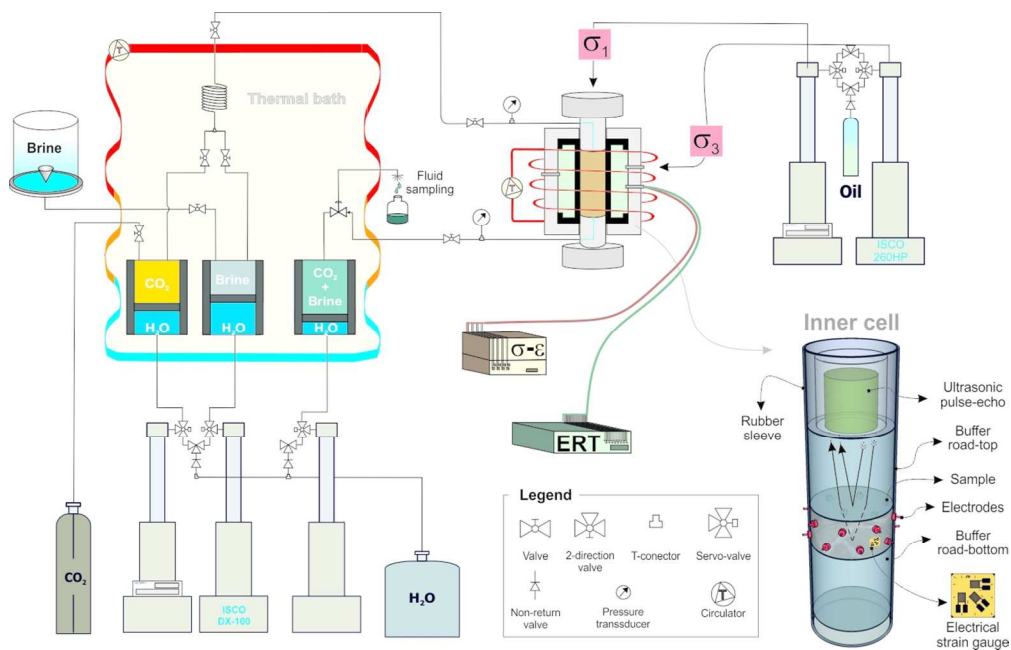


Figure 1. Experimental rig for CO₂ storage simulations. Bottom right, a schematic inner cell view highlights the distribution of the different geophysical tools and strain gauges.
241x153mm (150 x 150 DPI)

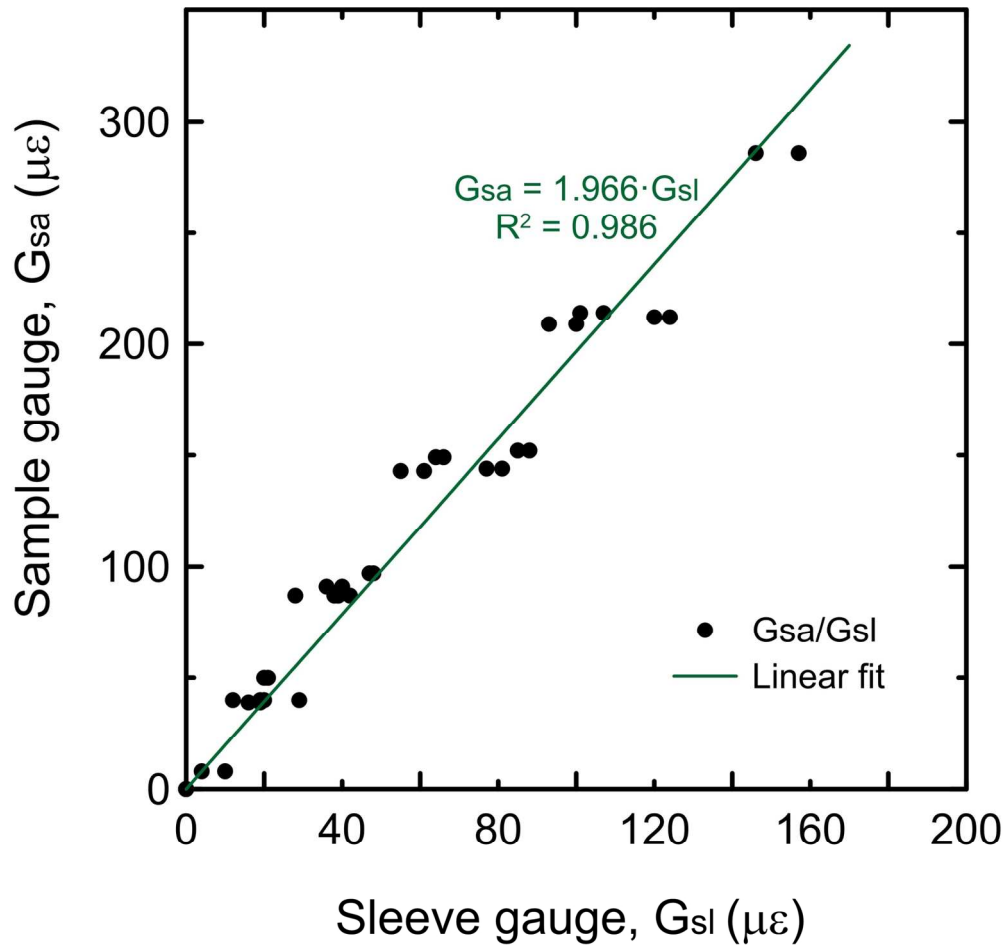


Figure 2. Axial strain calibration.
152x142mm (300 x 300 DPI)

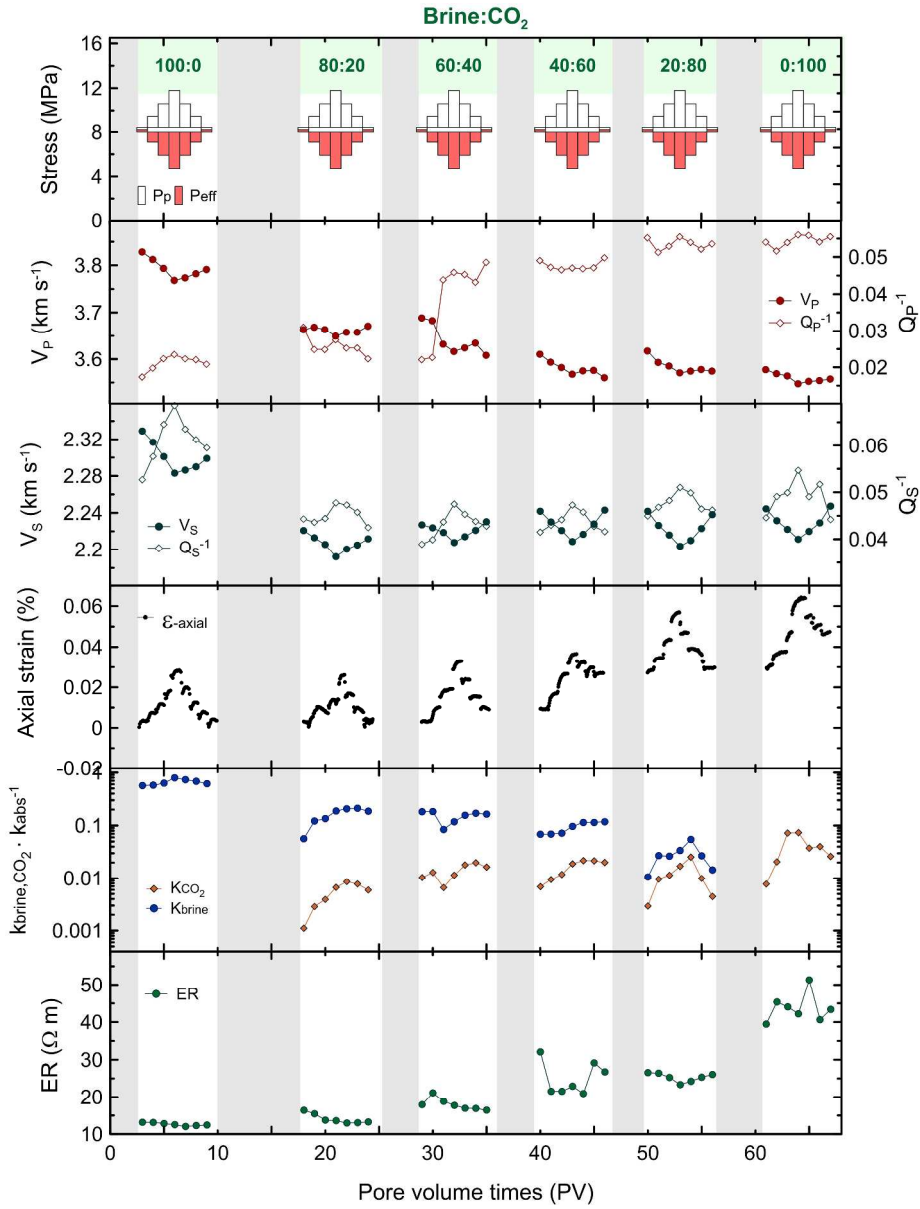


Figure 3. Six fractional flows brine:CO₂ flooding test. P- and S-wave velocities (V_p , V_s) and attenuation (by the inverse quality factors Q_D^{-1} and Q_S^{-1} , respectively), axial strains (ϵ_{axial}), relative permeability (k_{brine,CO_2}) and electrical resistivity (ER) against pore volume times (PV ~ 10 cm³). Geomechanical conditions defined by effective pressure, P_{eff} , and pore pressure, P_p .
417x544mm (300 x 300 DPI)

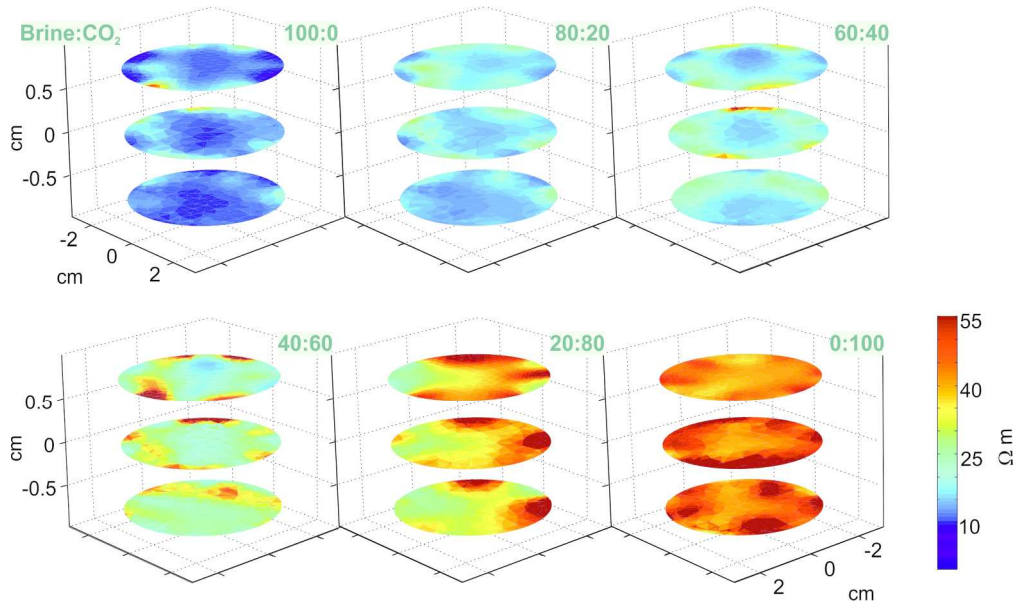


Figure 4. Electrical resistivity tomography for each fractional flow brine:CO₂ stage.
197x118mm (300 x 300 DPI)

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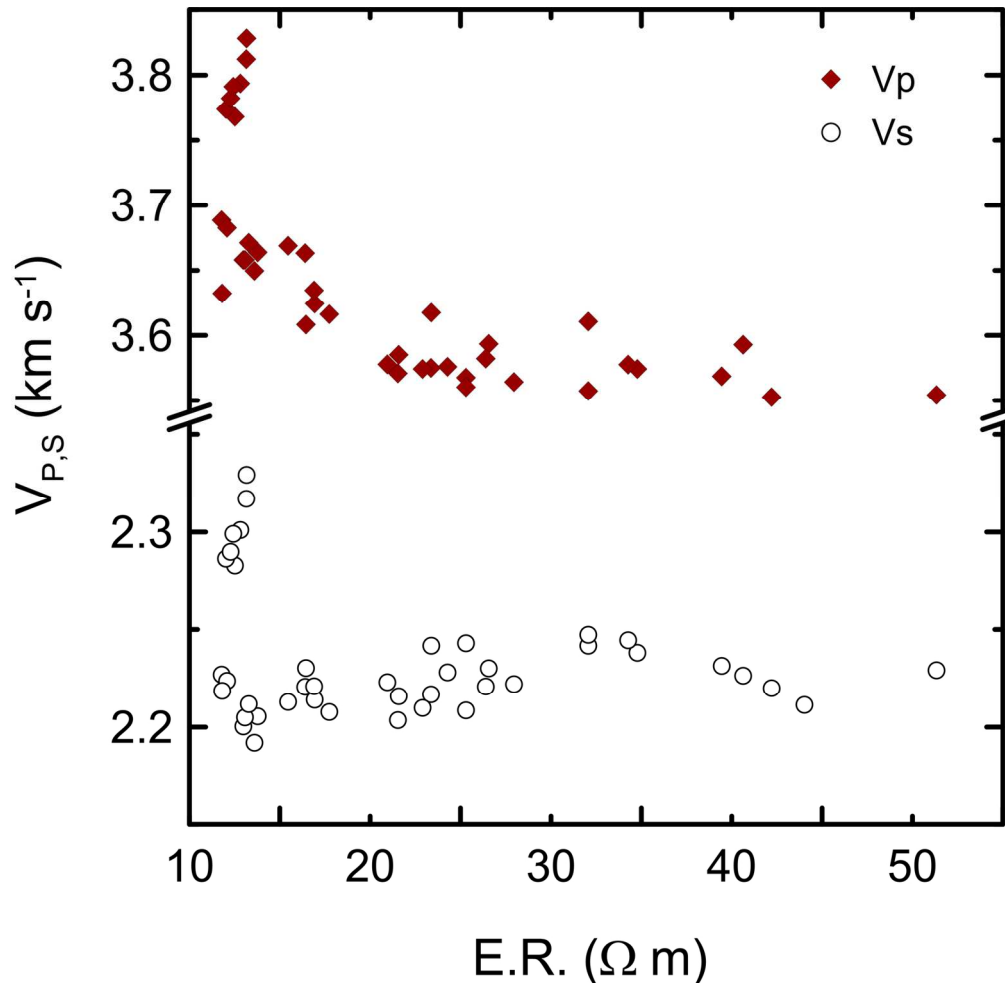


Figure 5. Acoustic wave velocities versus electrical resistivity. Experimental results. 152x149mm (300 x 300 DPI)

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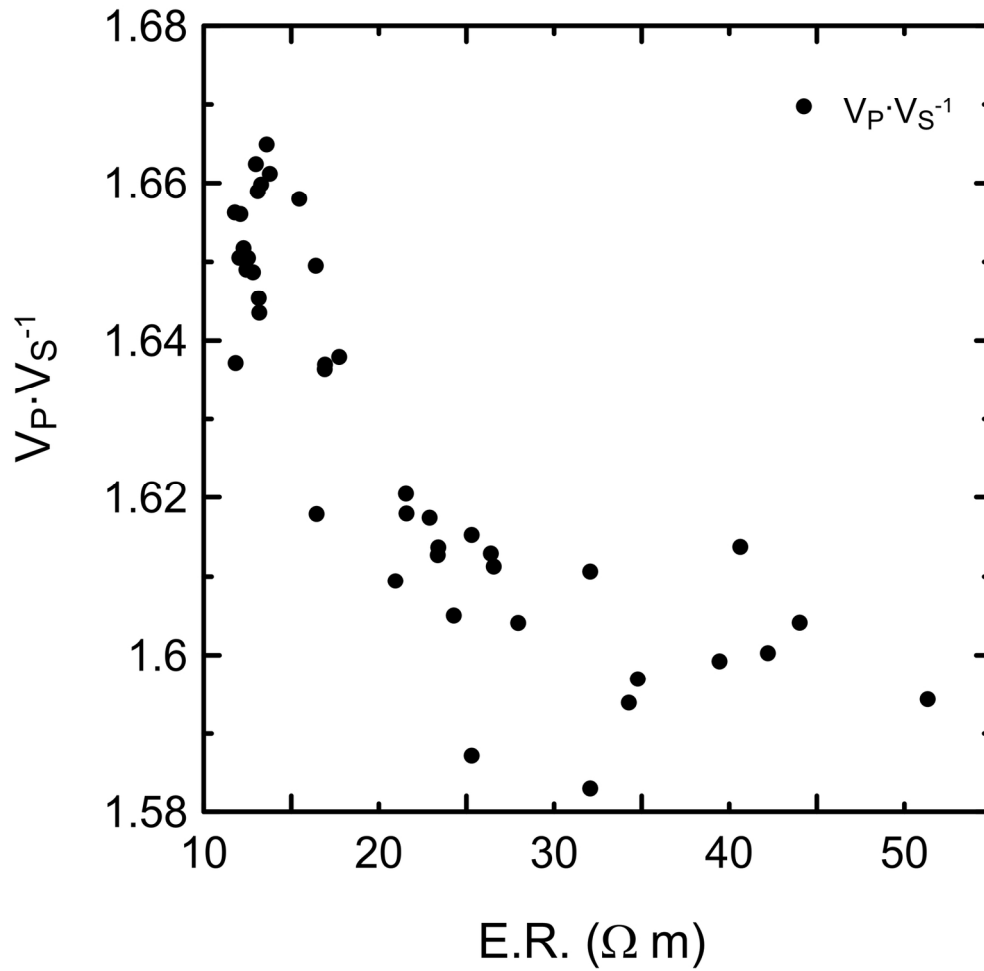


Figure 6. Acoustic wave velocity ratio $V_p \cdot V_s^{-1}$ versus electrical resistivity. Experimental results. 156x150mm (300 x 300 DPI)

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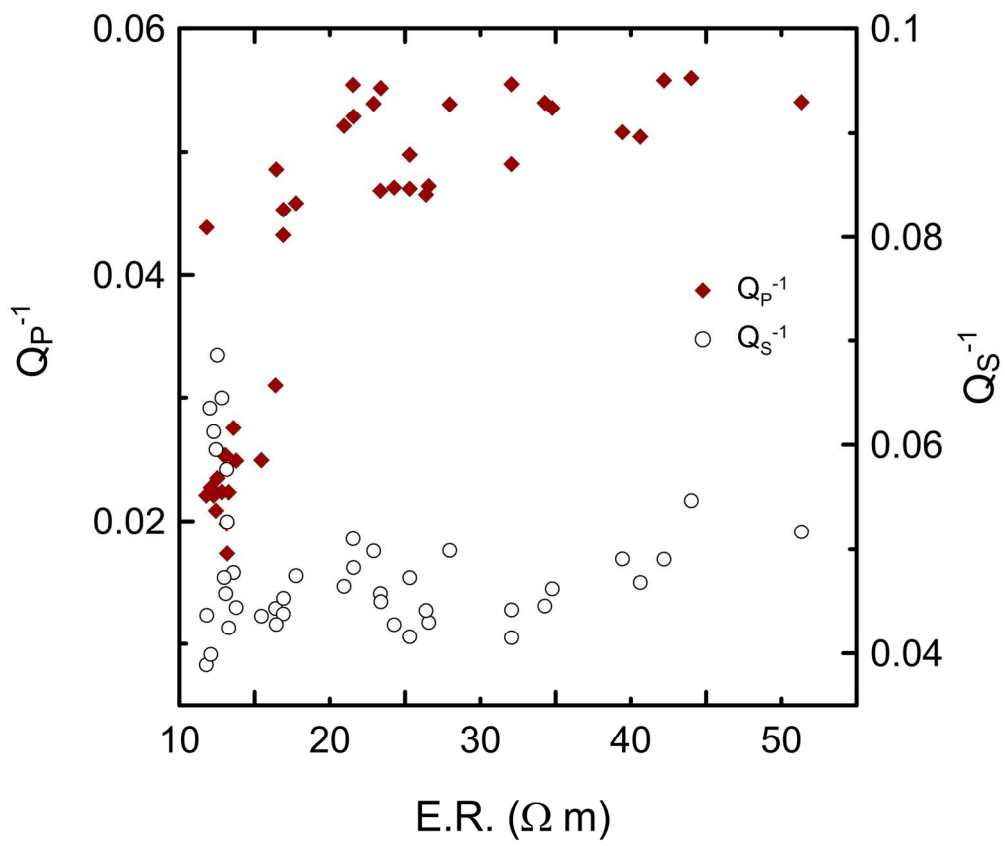


Figure 7. Acoustic wave attenuation factors versus electrical resistivity. Experimental results.
156x129mm (300 x 300 DPI)

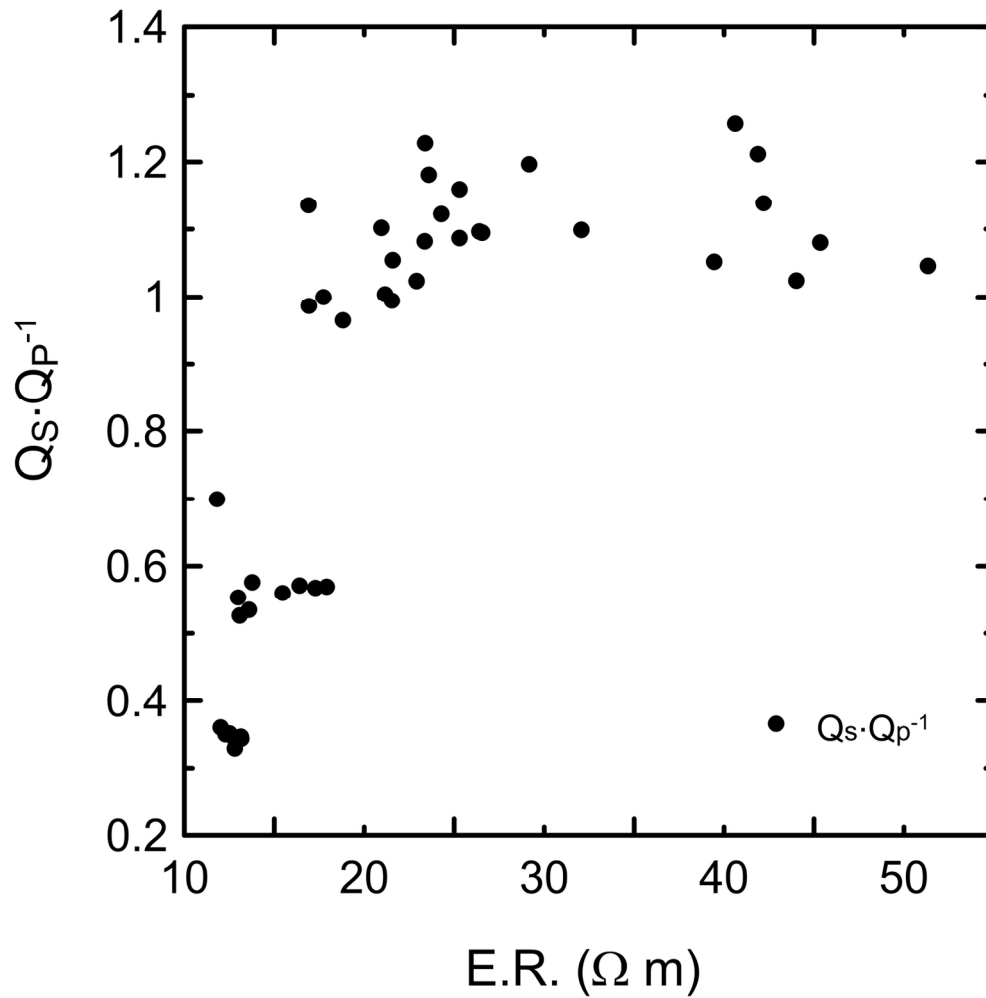


Figure 8. Acoustic wave attenuation ratio $Q_s \cdot Q_p^{-1}$ versus electrical resistivity. Experimental results.
156x155mm (300 x 300 DPI)

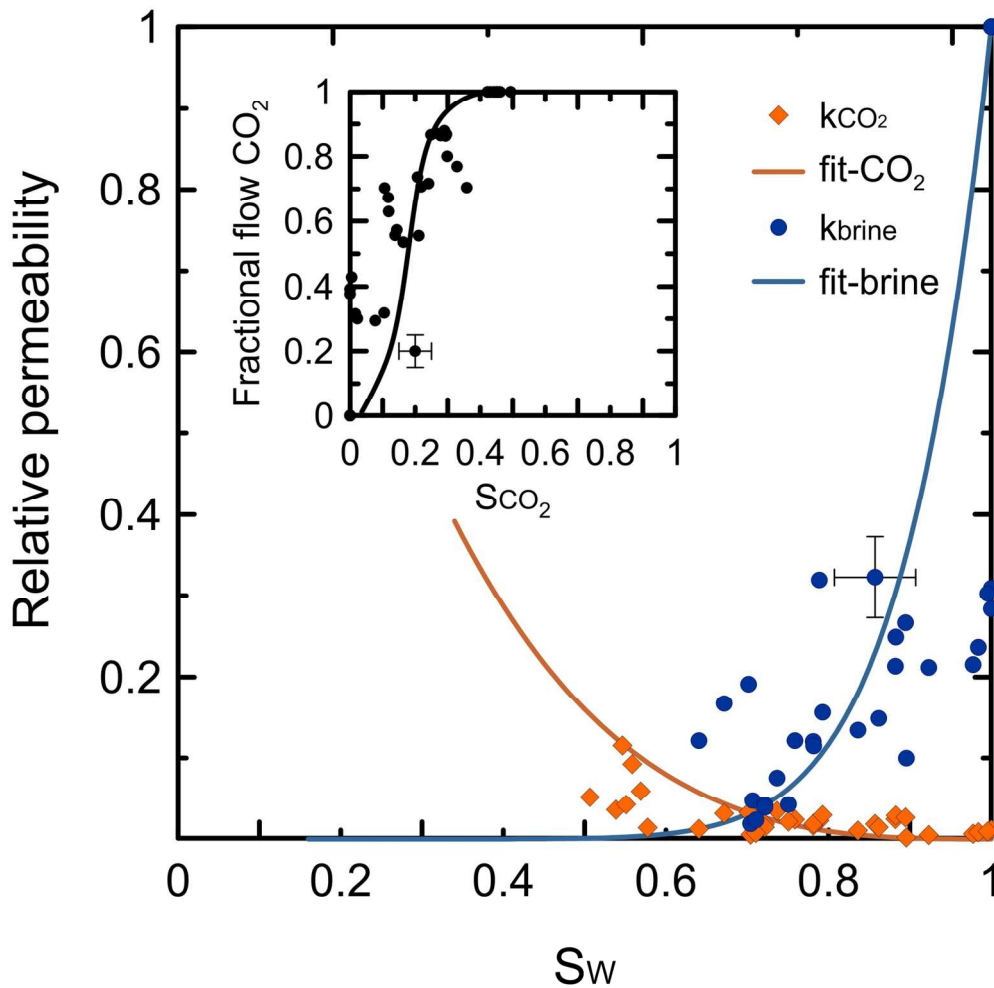


Figure 9. Brine-CO₂ relative permeability. Corey fitting: $S_{wr} = 0.15$, $N_w = 7$ and $N_{CO_2} = 0.5$; inner plot represents fractional flow evolution with CO₂ saturation. 155x154mm (300 x 300 DPI)

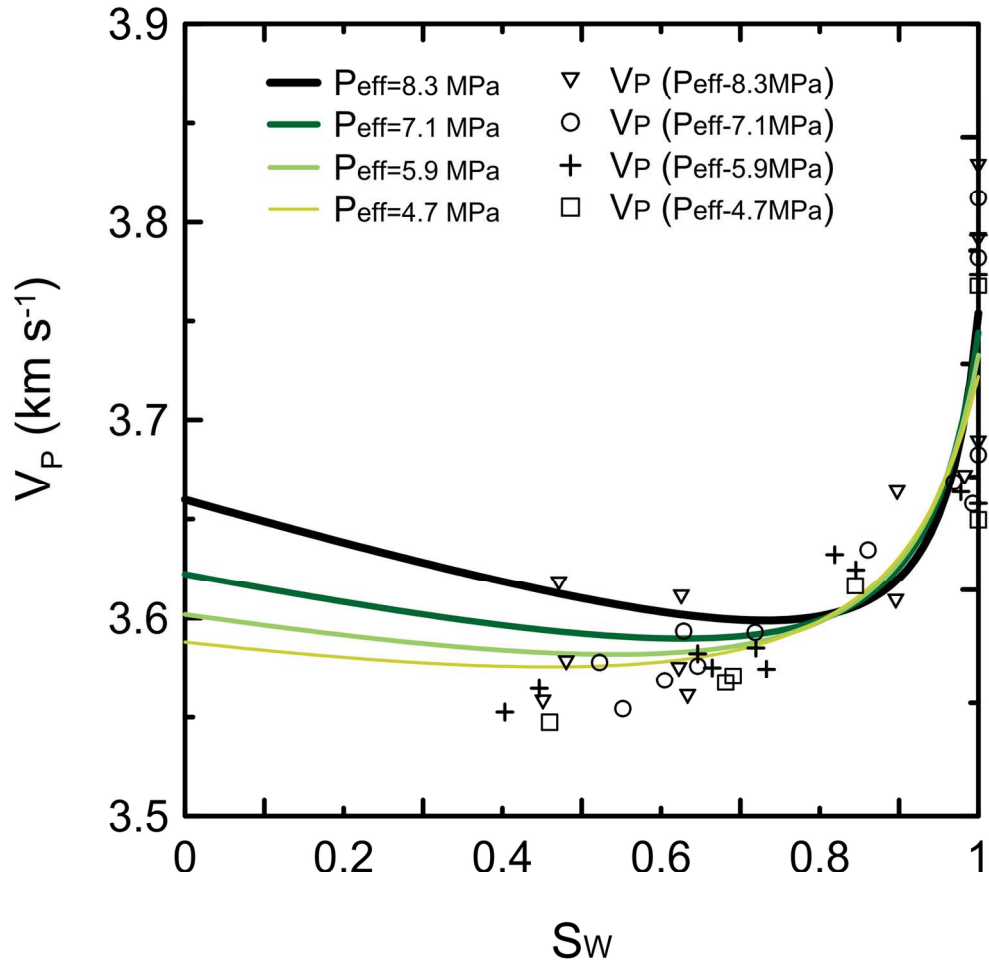


Figure 10. Gassmann's model prediction and observed V_p for the four effective pressure steps of the test. 155x150mm (300 x 300 DPI)

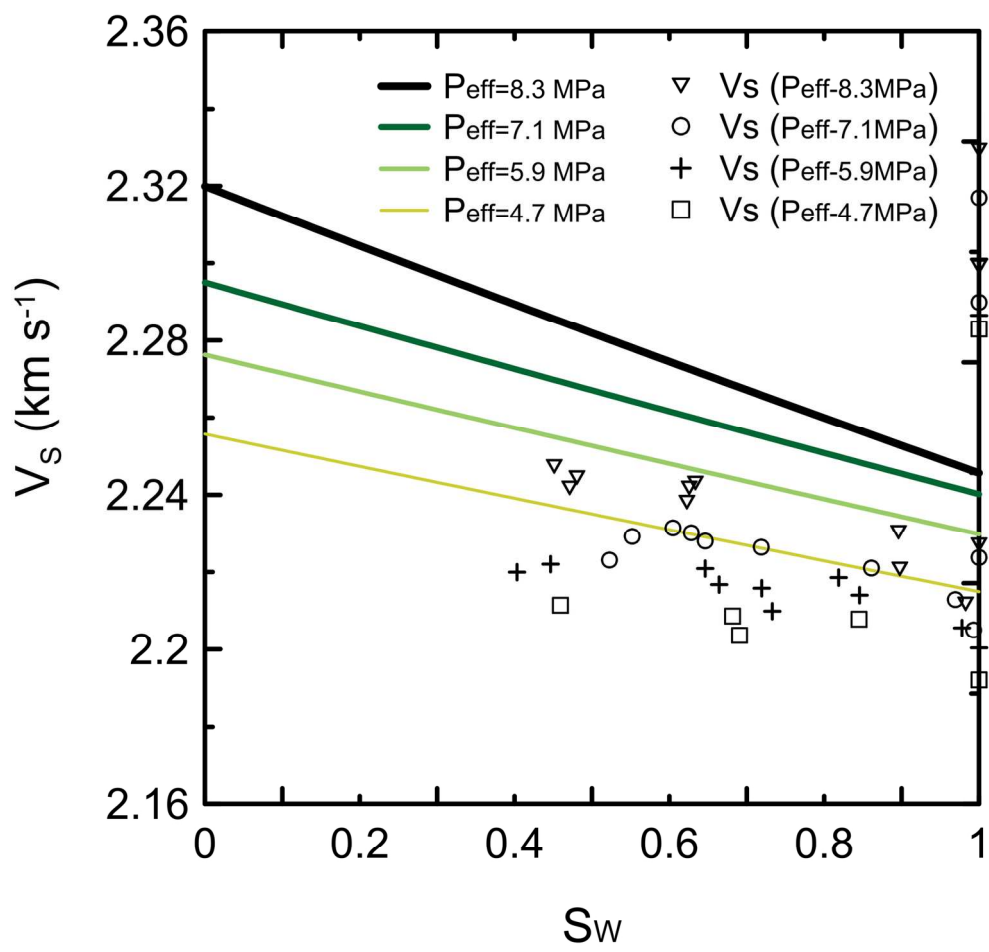


Figure 11. Gassmann's model prediction and observed V_s for the four effective pressure steps of the test. 155x146mm (300 x 300 DPI)

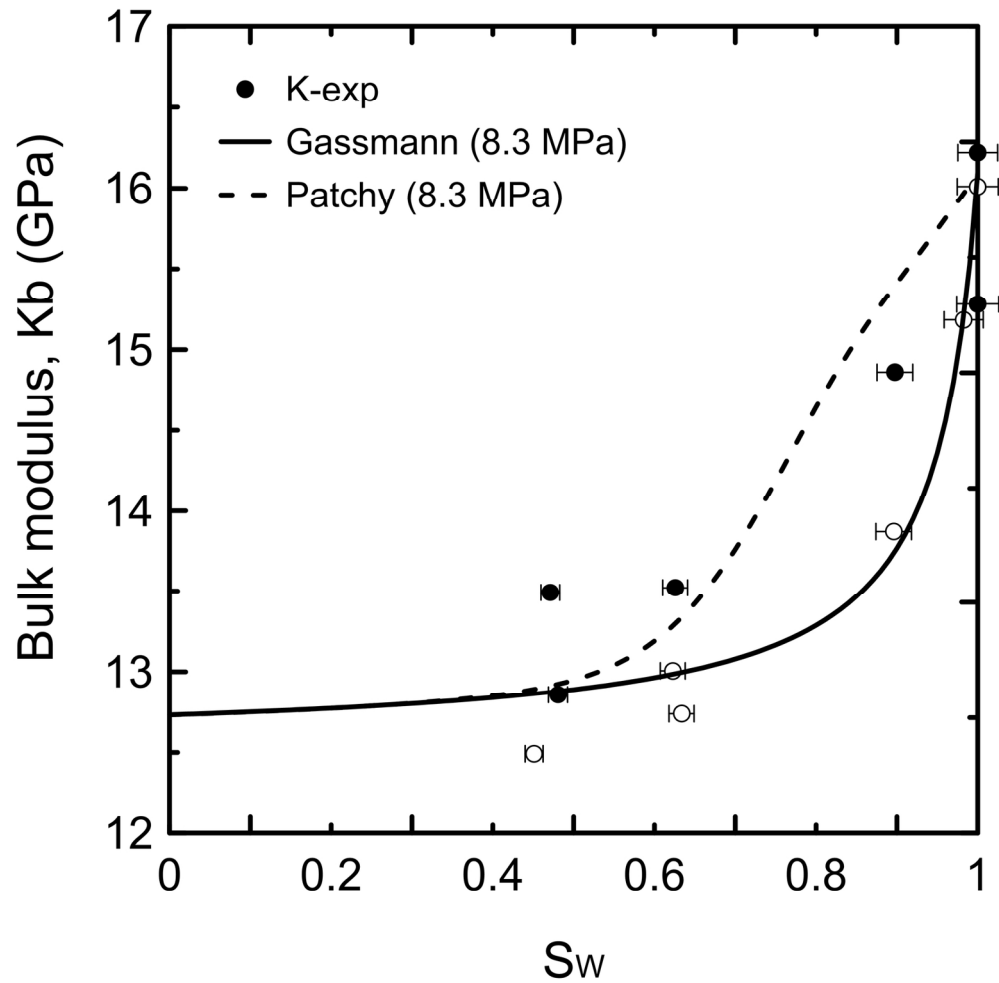


Figure 12a. Bulk modulus K_b validation by Gassmann and Patchy models the four effective pressure of the test $P_{\text{eff}} = 8.3$ MPa
 155x153mm (300 x 300 DPI)

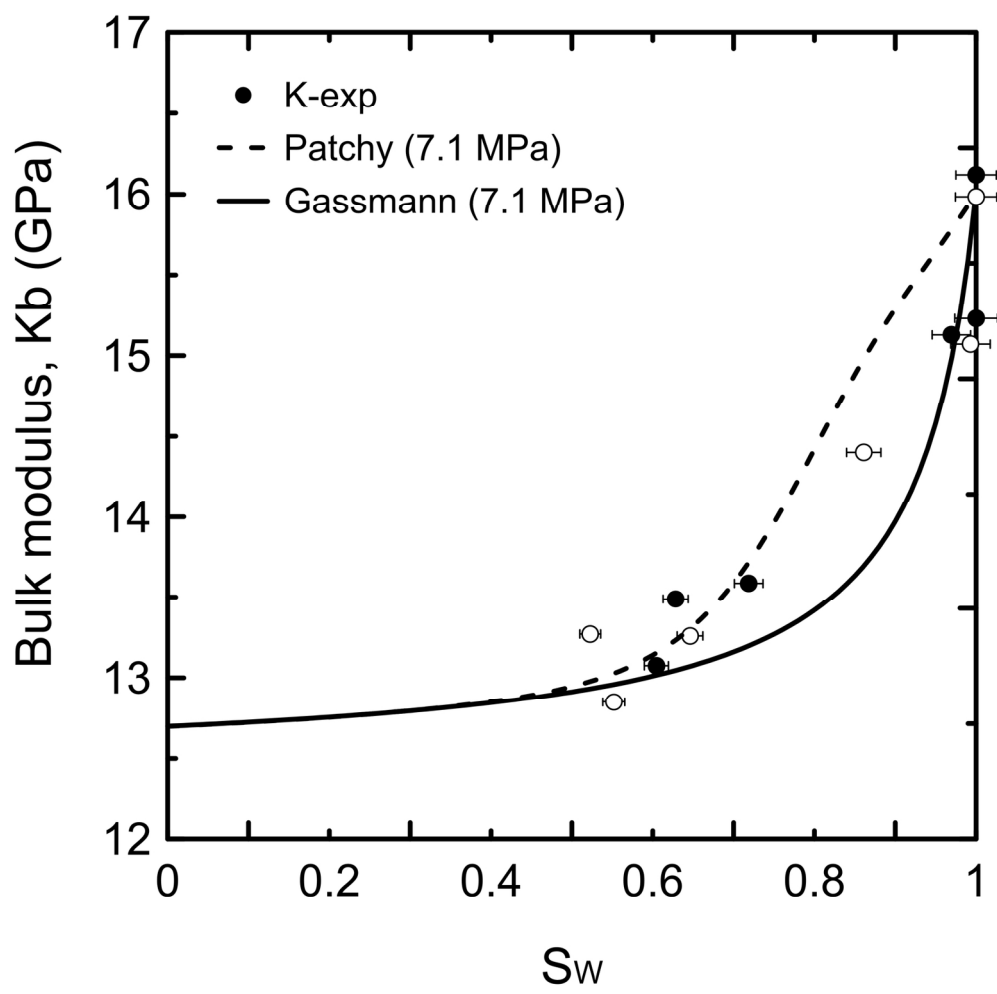


Figure 12b. Bulk modulus K_b validation by Gassmann and Patchy models the four effective pressure of the test $P_{eff} = 7.1$ MPa
155x153mm (300 x 300 DPI)

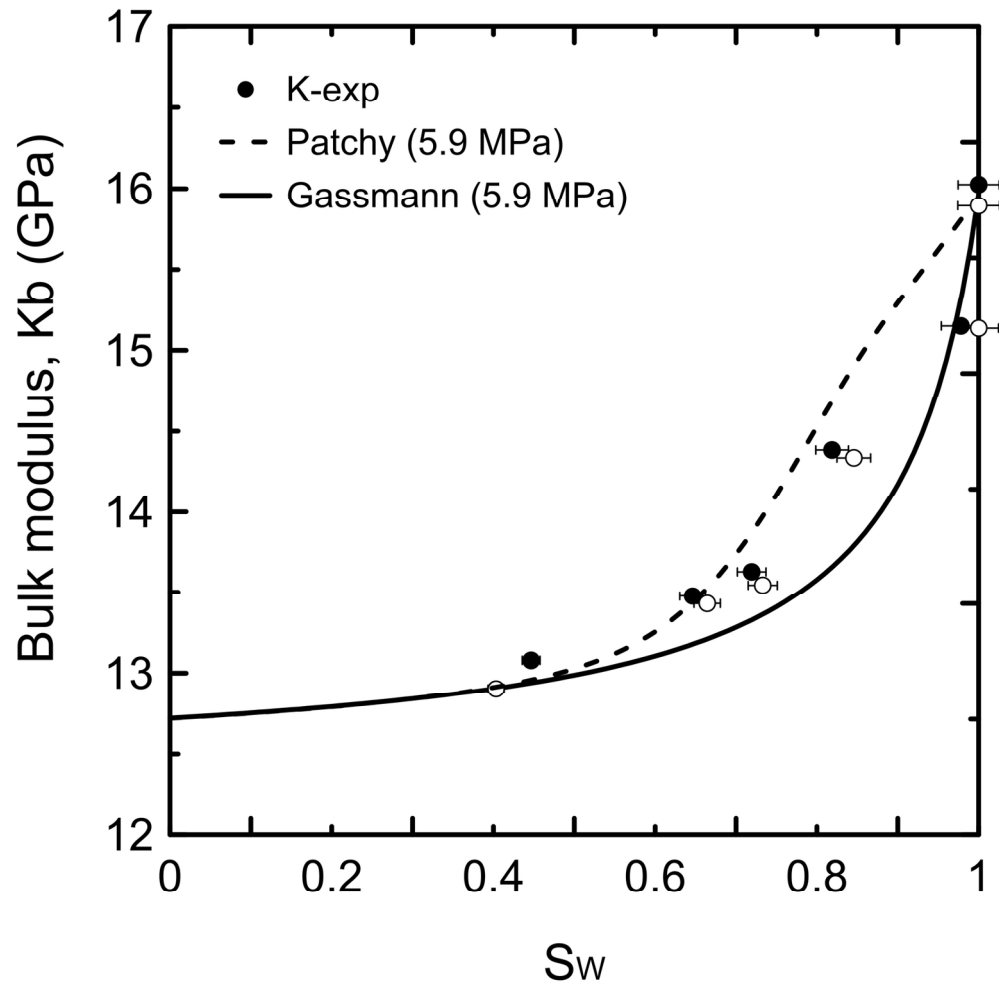


Figure 12c. Bulk modulus K_b validation by Gassmann and Patchy models the four effective pressure of the test $P_{\text{eff}} = 5.9$ MPa
155x153mm (300 x 300 DPI)

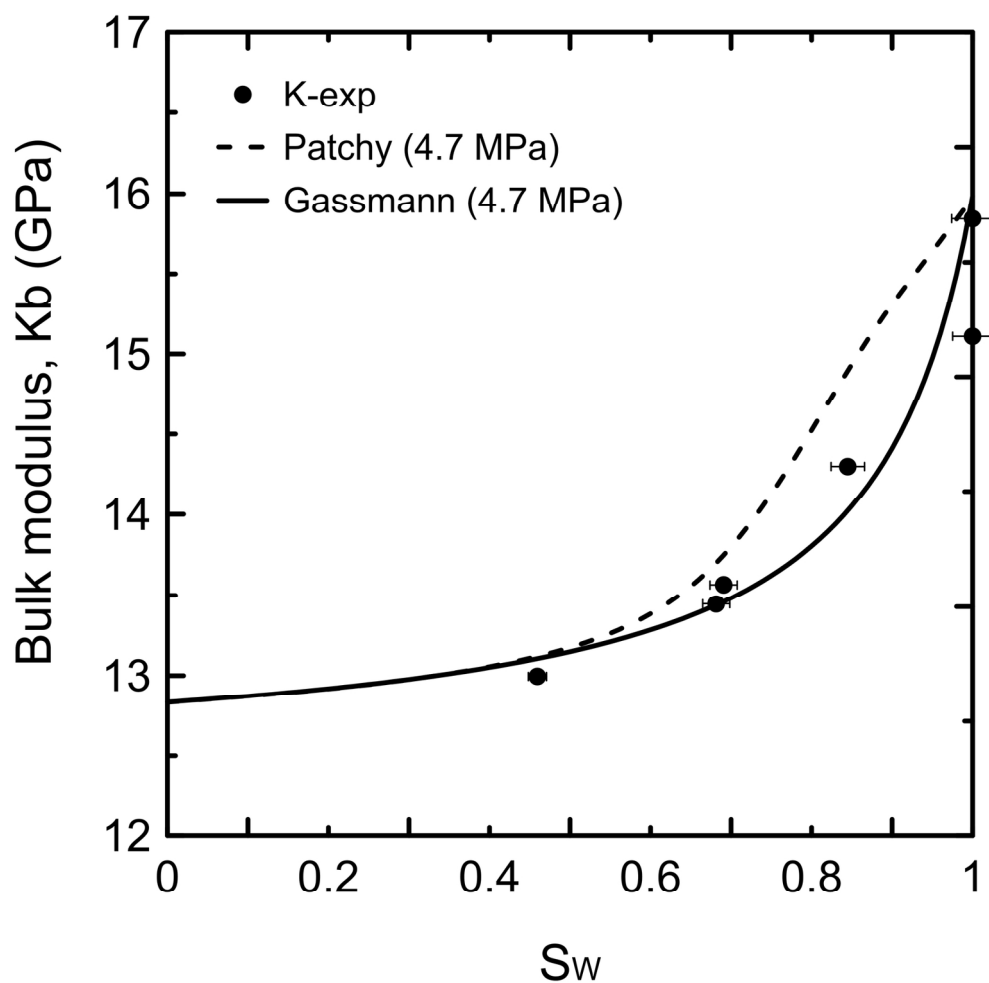


Figure 12d. Bulk modulus K_b validation by Gassmann and Patchy models the four effective pressure of the test $P_{eff} = 4.7$ MPa
155x152mm (300 x 300 DPI)

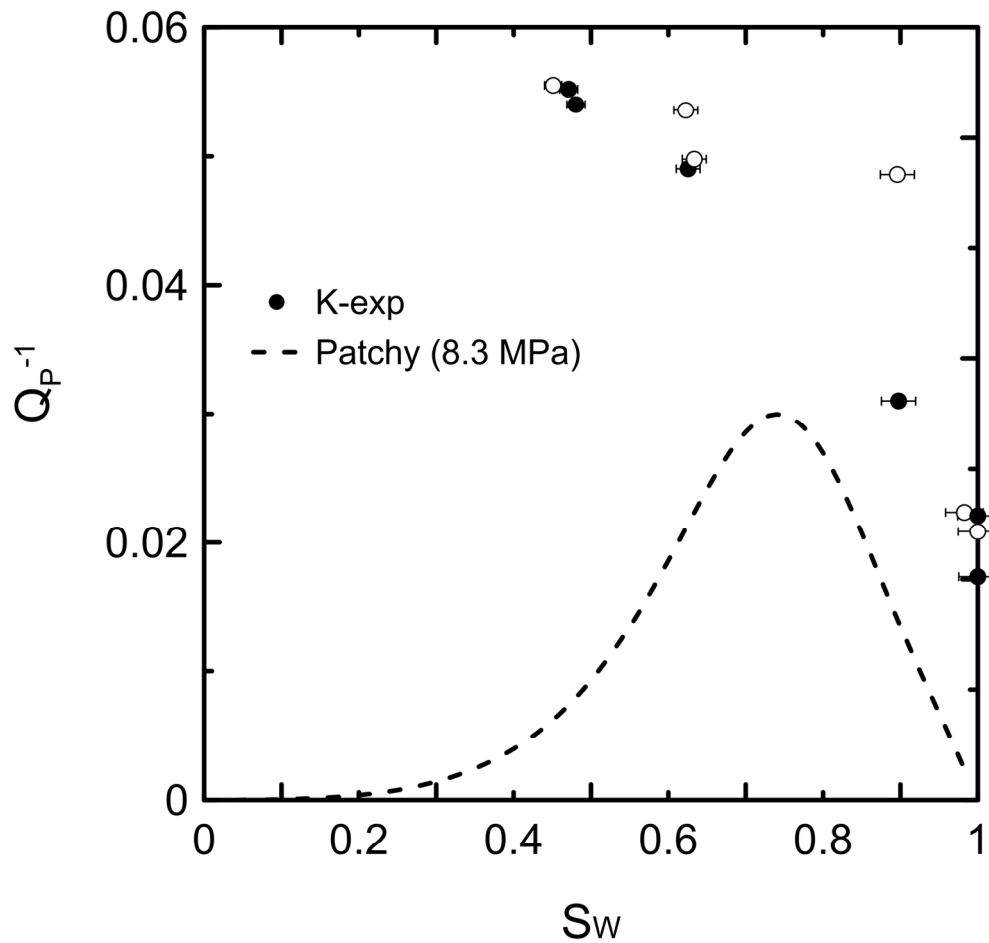


Figure 13a. P-wave attenuation Q_p^{-1} versus brine saturation for the four effective pressure of the test, at $P_{eff} = 8.3$ MPa. Experimental data shown by open circles (loading) and solid circles (unloading).
155x146mm (300 x 300 DPI)

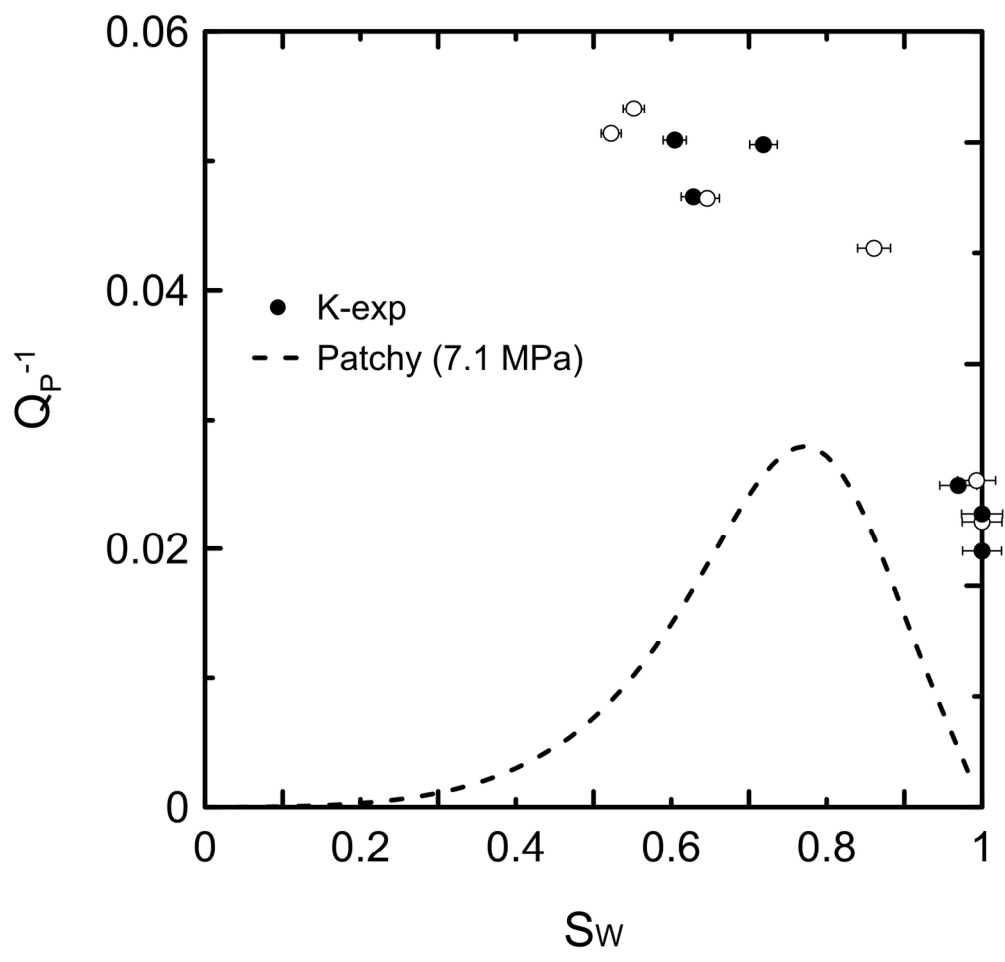


Figure 13b. P-wave attenuation Q_p^{-1} versus brine saturation for the four effective pressure of the test, at $P_{eff} = 7.1$ MPa. Experimental data shown by open circles (loading) and solid circles (unloading).
155x147mm (300 x 300 DPI)

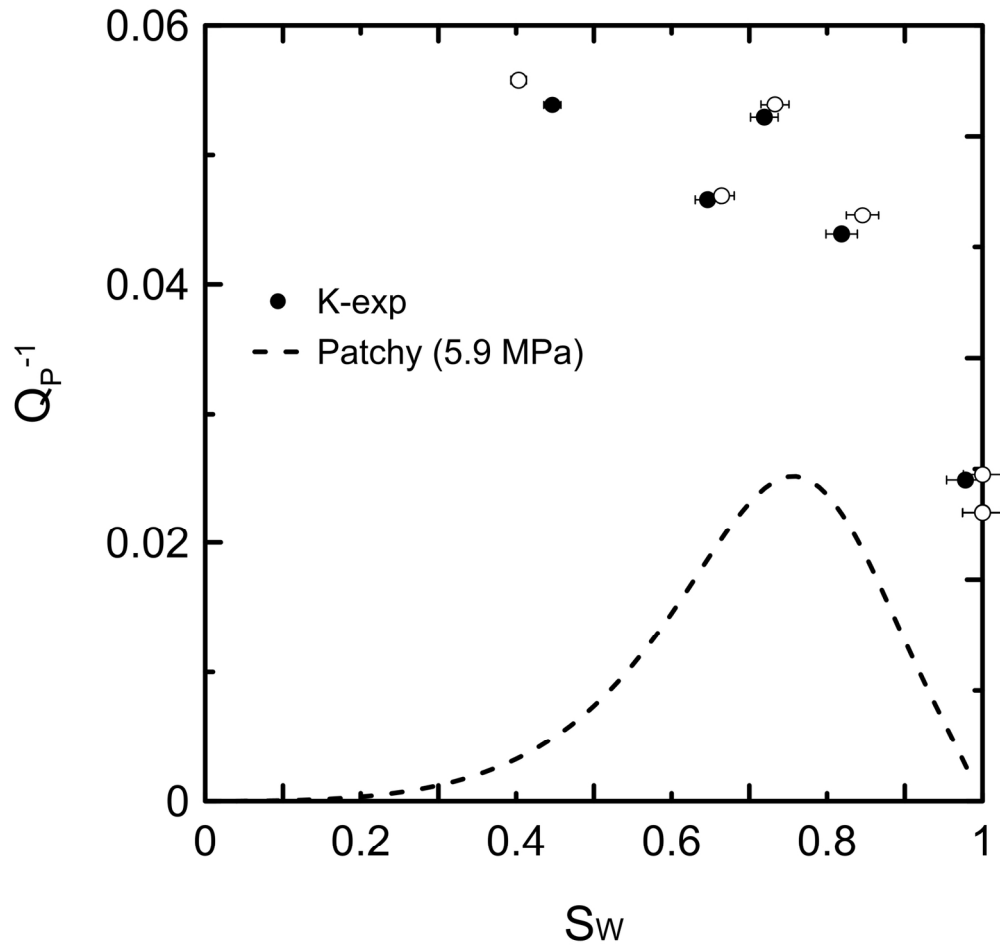


Figure 13c. P-wave attenuation Q_p^{-1} versus brine saturation for the four effective pressure of the test, at $P_{\text{eff}} = 5.9$ MPa. Experimental data shown by open circles (loading) and solid circles (unloading).
155x147mm (300 x 300 DPI)

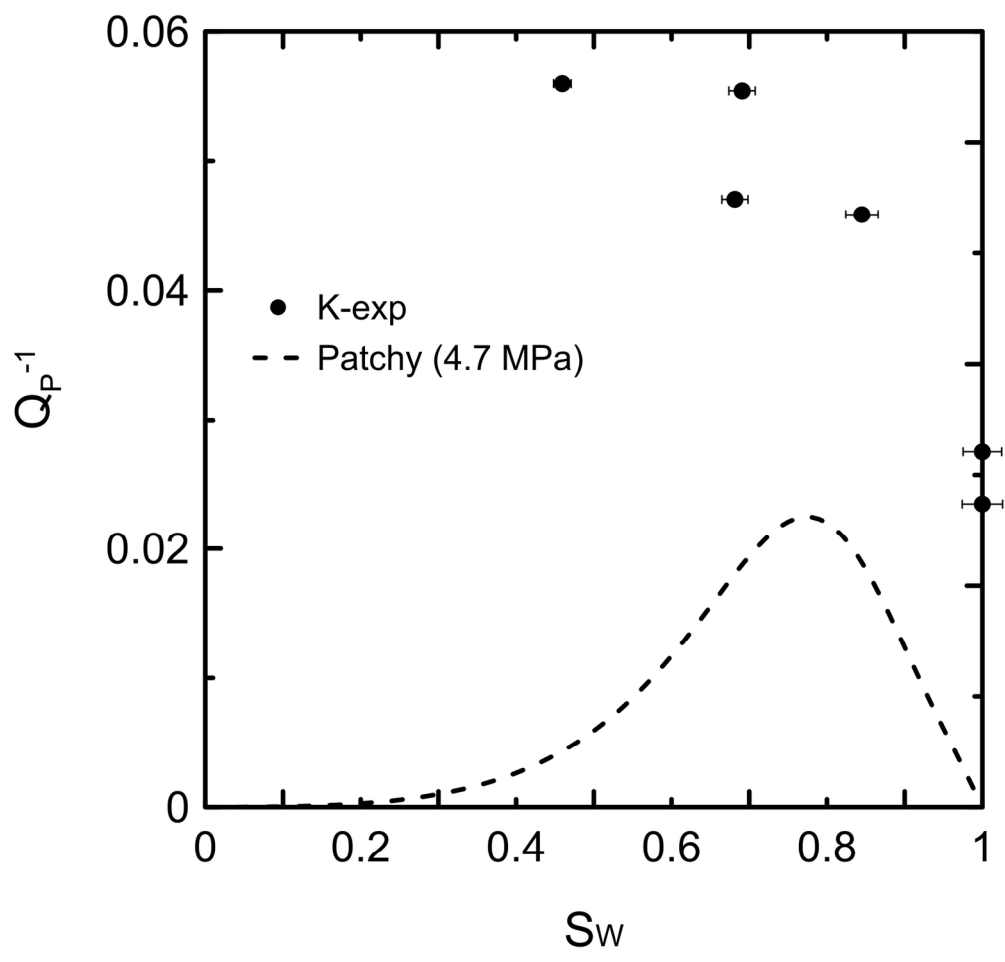


Figure 13d. P-wave attenuation Q_p^{-1} versus brine saturation for the four effective pressure of the test, at $P_{eff} = 4.7$ MPa. Experimental data shown by open circles (loading) and solid circles (unloading).
155x147mm (300 x 300 DPI)

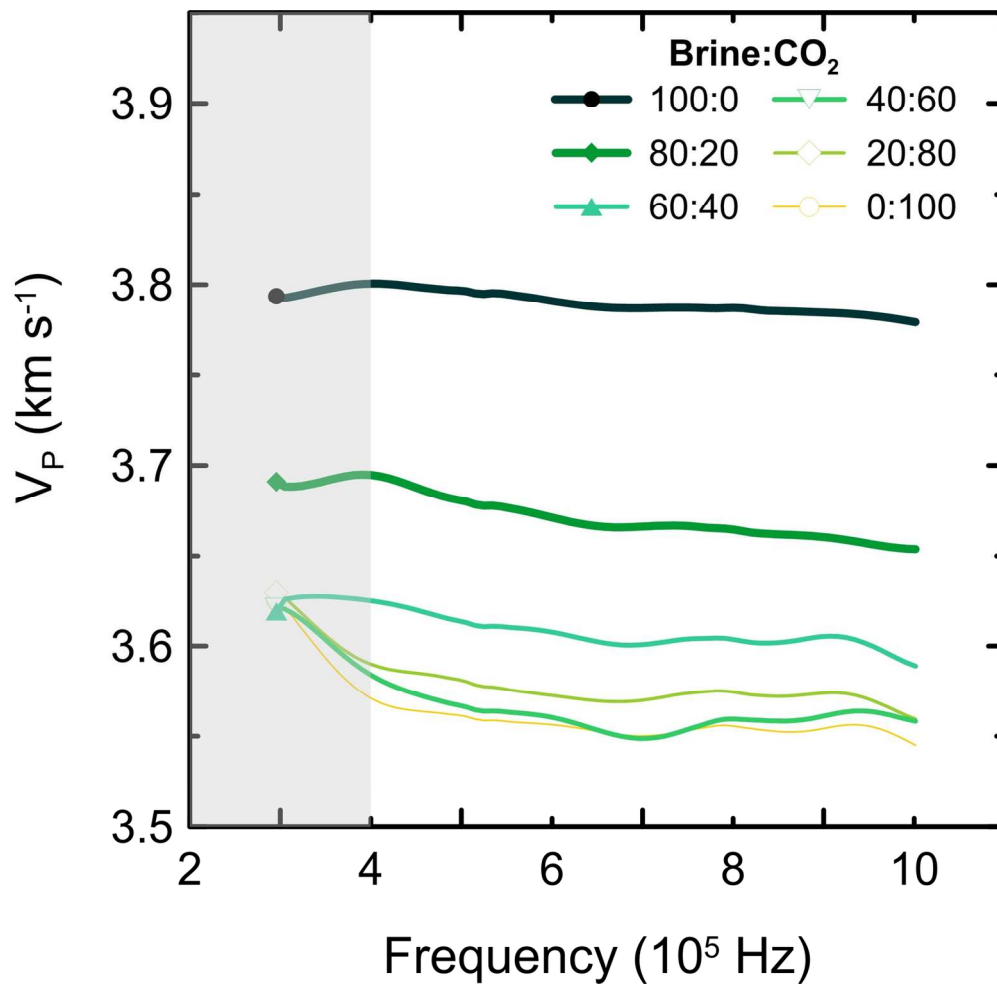


Figure 14a. P-wave velocity in the frequency range 3-10 kHz, for the six fractional flows brine:CO₂. The examples correspond to the experimental data at P_{eff} = 4.7 MPa.
151x147mm (300 x 300 DPI)

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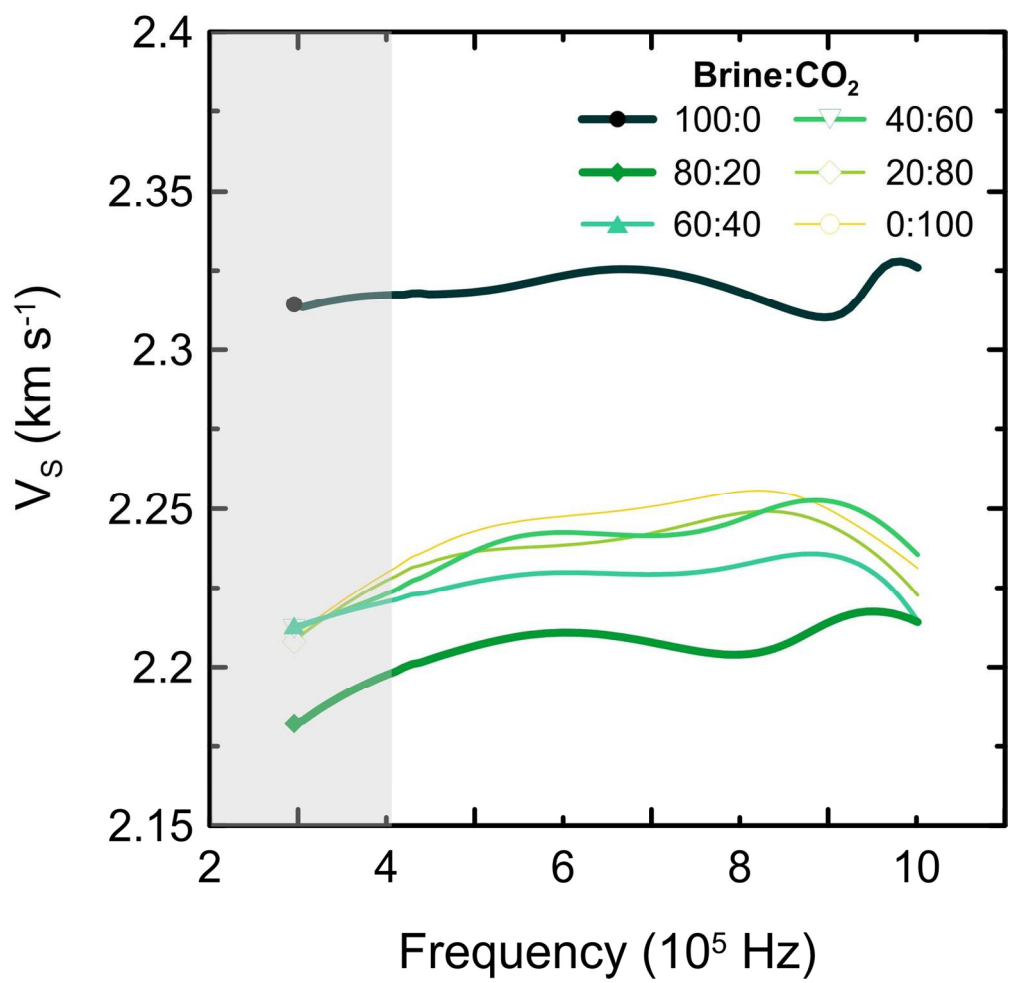


Figure 14b. S-wave velocity in the frequency range 3-10 kHz, for the six fractional flows brine:CO₂. The examples correspond to the experimental data at $P_{eff} = 4.7$ MPa.
155x150mm (300 x 300 DPI)

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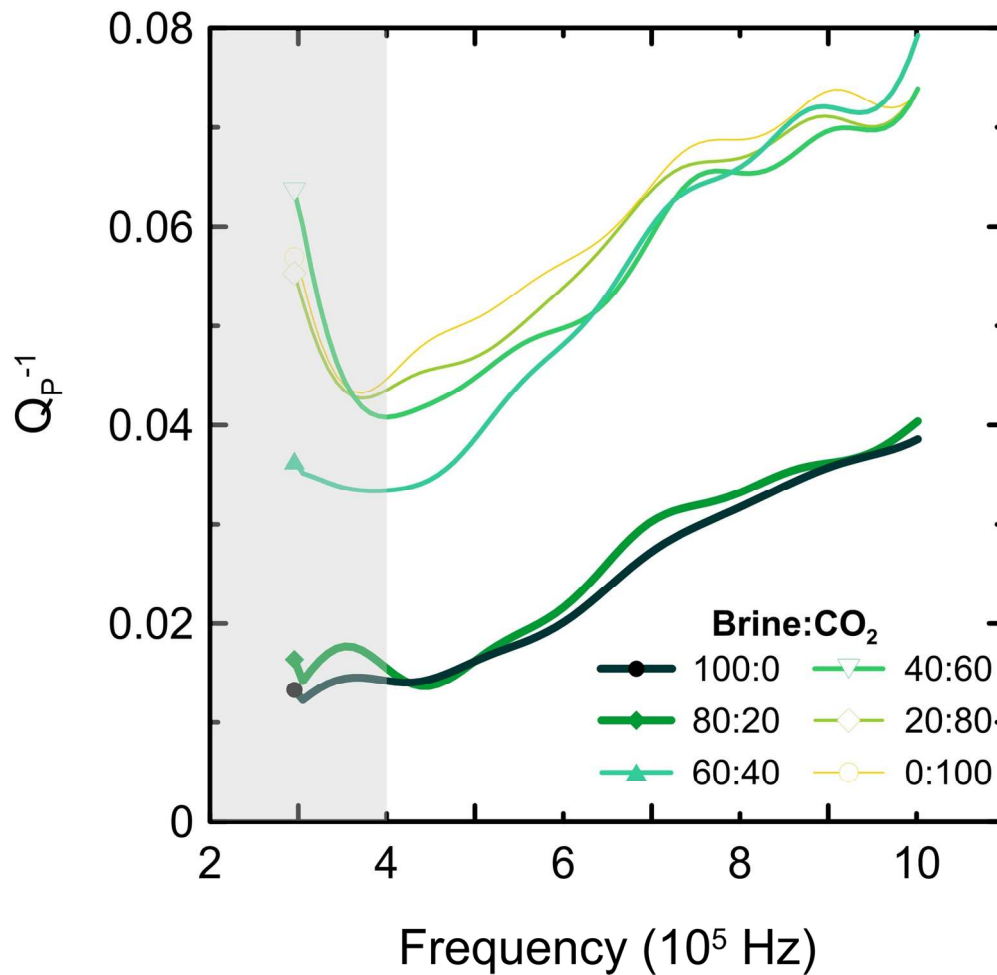


Figure 14c. P-wave attenuation in the frequency range 3-10 kHz, for the six fractional flows brine:CO₂. The examples correspond to the experimental data at $P_{eff} = 4.7$ MPa.
155x150mm (300 x 300 DPI)

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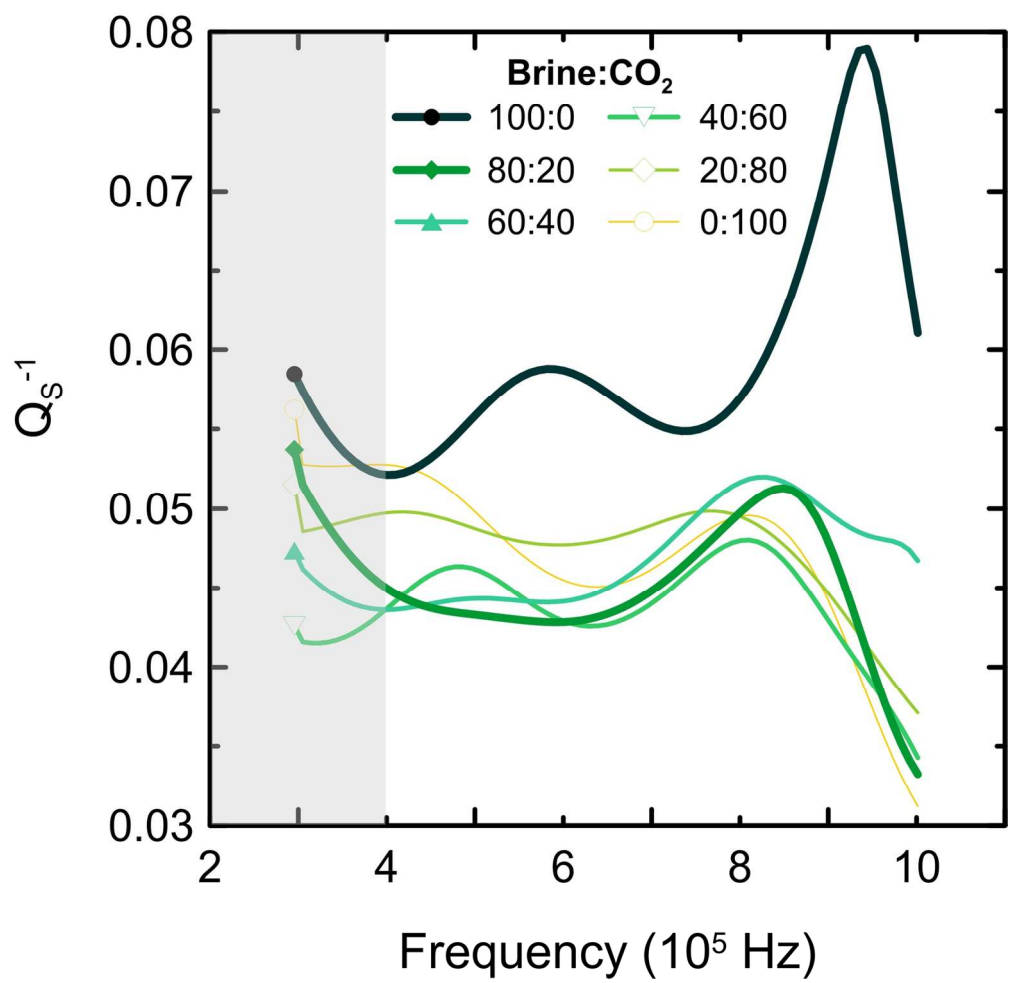


Figure 14d. S-wave attenuation in the frequency range 3-10 kHz, for the six fractional flows brine:CO₂. The examples correspond to the experimental data at $P_{eff} = 4.7$ MPa.
155x150mm (300 x 300 DPI)

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