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# Acoustic response of reservoir sandstones during injection of supercritical CO<sub>2</sub>

M. Lebedev<sup>a\*</sup>, S. Iglauer<sup>b</sup>, V. Mikhaltsevich<sup>a</sup>

<sup>a</sup>Department of Exploration Geophysics, Curtin University, 26 Dick Perry Avenue, 6151 Kensington, Australia

<sup>b</sup>Department of Petroleum Engineering, Curtin University, 26 Dick Perry Avenue, 6151 Kensington, Australia

## Abstract

We report experimental results for acoustic response measurements conducted during injection of supercritical carbon dioxide into a brine saturated sandstone plug. We measured P- and S-wave velocities ( $V_p$ ,  $V_s$ ) as a function of effective stress and CO<sub>2</sub> saturation in a sandstone plug. We demonstrate that Gassmann's fluid substitution procedure matches the experimental results well for this sample. A 3.5% reduction of P-wave velocity after injection of two pore volumes of CO<sub>2</sub> into brine-saturated sample was measured. We conclude that measurement of  $V_p$  can be used to estimate CO<sub>2</sub> saturations in rock. In addition, x-ray computer tomography (CT) images were acquired at reservoir conditions with a resolution of 33  $\mu\text{m}$ , which provided more detailed information about CO<sub>2</sub> saturations and distributions in the rock. It is envisaged that these techniques (seismic and CT) can be combined in the future to enable a more holistic understanding of how fluid-fluid displacement processes are coupled with the acoustic response characteristics of the rock.

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## 1. Introduction

Successful monitoring of immiscible or partially miscible fluid-fluid displacement in the subsurface is key to many technological applications such as oil and gas recovery [1], water management, contaminant transport [2], or

\* Corresponding author. Tel.: +618 9266 2330; fax: +61 8 9266 3407.

E-mail address: [m.lebedev@curtim.edu.au](mailto:m.lebedev@curtim.edu.au)

geological greenhouse gas sequestration [3]. At the state-of-the-art, seismic measurements methods are the most favorable monitoring method; this includes time lapse (4D) monitoring, where subsurface features are imaged in 3D over time periods spanning years to decades [4]. Such seismic methods are based on sending elastic waves into the formation and on measuring associated arrivals times and amplitudes of the reflected and/or transmitted seismic waves [5]. This seismic wave propagation strongly depends on the mechanical properties of the porous medium. Consequently the replacement of one fluid inside the pore space of the rock by a second fluid will change the response of the seismic waves as the effective mechanical properties of the reservoir are changed. This relationship enables monitoring of the fluid plume evolution.

One important example – on which we focus here – and where these mechanisms play an important role are industrial-scale carbon geo-storage (CGS) projects, where CO<sub>2</sub> is injected deep underground for storage [3]. CGS projects specific to Australia are the Otway (Otway basin, Victoria), South West Hub (Harvey, Western Australia), CTSCo (Surat basin, Queensland), and CarbonNet (Victoria) projects; each project requires 4D monitoring of the CO<sub>2</sub> plume migration for risk assessments and public assurance [6]. Seismic monitoring provides information about the dimensional characteristics of the plume and an estimation of CO<sub>2</sub> saturation. However, despite many years of research efforts, both in the areas of exploration geophysics and multiphase flow through porous media, the effect of fluid-fluid displacement on mechanical rock properties is poorly understood. As a result, the accuracy of seismic monitoring techniques is limited. In this context a limited number of experiments, where the impact of CO<sub>2</sub> injection on elastic rock properties was studied, were reported; a reduction of the P-wave velocity caused by partial replacement of brine with carbon dioxide was observed by several groups [7-13], moreover in some experiments [13] a violation of Gassmann's fluid substitution theory [14] was found. We note that the mechanism, which causes this violation, is currently debated, and samples anisotropy, squirt flow [15], as well as distribution of multiple fluids [16], and/or reaction of the rocks with the fluids have been suggested to explain such violation.. In conclusion, for an accurate measurement of CO<sub>2</sub> saturation and monitoring of plume size and plume migration it is vital to further understand how the underlying multi-phase fluid dynamics are coupled with the acoustic response patterns. We thus investigate here experimentally the effects of supercritical carbon dioxide (scCO<sub>2</sub>) injection on the acoustic properties of a reservoir sandstone sample.

## **2. Experimental Methodology**

We selected a core from an actual proposed injection site in Australia and measured the acoustic response characteristics of the core plug for various saturation states at reservoir conditions (high pressure, elevated temperature): dry, fully brine saturated, and during CO<sub>2</sub> injection; this mimicked the processes in a storage formation, when CO<sub>2</sub> is injected, and the CO<sub>2</sub> plume evolution is monitored with seismic techniques. In a separate experiment, we imaged the sandstone plug at analogue flooding conditions (high pressure, but only slightly elevated room temperature) with an x-ray tomograph in order to better understand the associated fluid dynamics in the rock specimen.

### *2.1. Sample*

We selected a core from the Harvey-1 well (depth 915 m, upper Leuser sandstone (Yalgorup member) of Perth basin, [17]), and drilled a smaller, standard sized cylindrical plug from the core (diameter = 38mm; length = 80mm). The composition of a "sister" plug (drilled next to the plug used in the flooding experiments) was measured by XRD and indicated a content of 82wt% quartz, 12wt% of K-feldspar; 5wt% of kaolinite, and 1wt% calcite [17]. The brine permeability and porosity of the sample at 10 MPa confining stress was measured as  $2.57 \times 10^{-14} \text{ m}^2$  (26 mD) and 19.1%, respectively.

## 2.2. Ultrasonic measurements

For the acoustic measurements the plug was mounted into a tri-axial rig, which was connected to core flooding equipment. The P- and S-wave velocities of the dry plug were then measured as a function of effective stress (4–36 MPa, Table 1) in axial direction using ultrasonic transducers (V153, Panametrics) at 0.5 MHz central frequencies.

The dry plug was then vacuumed for more than 24 hours to remove all air from the pore space. This was followed by injection of 3 pore volumes (PV) of brine at low pressure; then the pore pressure was increased to 6.5 MPa by a high precision syringe pump and another 3 PV of brine were injected. This procedure guaranteed full saturation of the plug. Again P- and S-wave velocities were measured as a function of effective stress. Subsequently CO<sub>2</sub> was injected with a high precision syringe pumps (D260, Teledyne – Isco) at a flow rate of 1mL/min and 0.5 MPa capillary pressure, Table 1; the CO<sub>2</sub> saturation in the plug was measured by mass balance, and the acoustic response was continuously measured during CO<sub>2</sub> flooding. All components of the rig were heated and maintained at isothermal conditions throughout the whole experiment (temperature 318K). The sample was positioned vertically (the Bond number was estimated to be 0.01, and we therefore do not expect that gravitational forces significantly influence the flow pattern [18]); and all fluids were injected into the bottom face of the plug, while production occurred from the top. The detailed experimental protocol is described elsewhere [13]; we note that the experiment was designed in such a way that it allowed us to measure acoustic velocities of the samples for all possible CO<sub>2</sub> saturations (from 0 to 100%).

## 2.3. X-ray Computer Tomography (CT) imaging

For the CT imaging, the specimen was housed in an x-ray transparent high pressure elevated temperature (HPET) polyetheretherketone (PEEK) core holder (Figure 1). Two piezoelectric transducers were attached to the outside walls of the HPET cell with which the P-waves can be generated and measured during CO<sub>2</sub> flooding; this setup will be used in the future to simultaneously acquire acoustic and CT data.

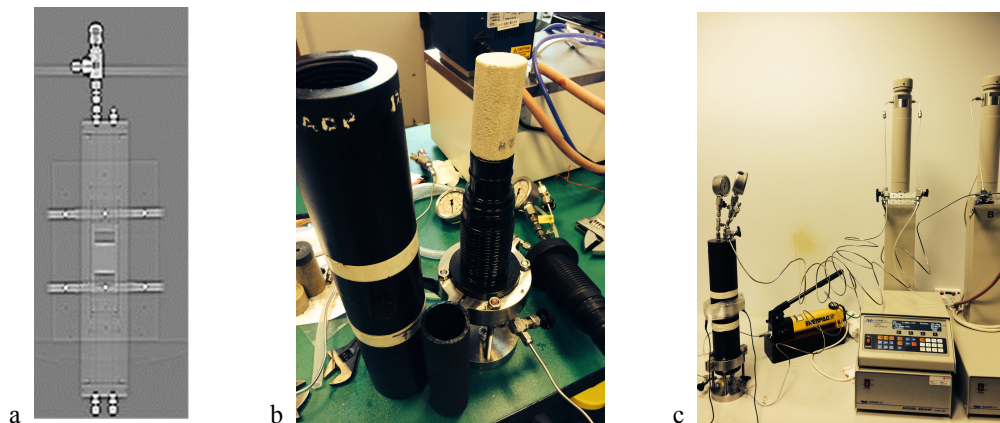


Fig. 1. (a) Radiograph of the x-ray transparent core holder used for the CT experiments. (b) Preparation of the experiment: 38mm diameter sandstone plug is placed in the middle of the core holder; (c) Core holder connected to syringe pumps.

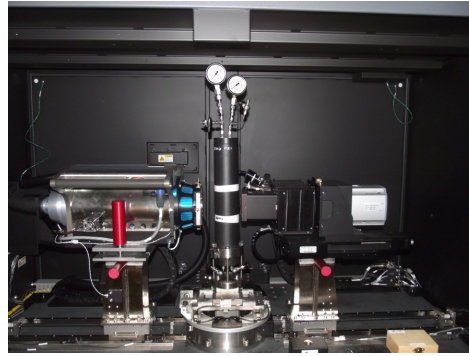


Fig. 2. Setup of experiment inside the micro CT scanner: the x-ray transparent core holder is oriented vertically.

Once the plug was mounted in the core holder, a confining pressure of 8 MPa was applied, and the sample was imaged in a dry state with a micro-computed tomograph (Versa XRM-500, Xradia –Zeiss) at a nominal voxel resolution of  $(33.3\mu\text{m})^3$ . The arrangement of the HPET cell inside the microCT scanner is shown in the Figure 2. The same image resolution was used throughout the whole workflow, and at this resolution some of the larger individual pores and their saturation states could be observed. In the images of the dry sample, air is black (as it has the lowest relative radiodensity, [19]), quartz is dark grey and minerals with high x-ray absorption (i.e. calcite and K-feldspar) are white or light grey. The dry sample was then vacuumed for at least 24 hours to remove all air from the pores. Subsequently 3 PV of brine were injected at low pressure, and another 3 PV at 6.5 MPa backpressure and 8 MPa effective stress; this procedure fully saturated the plug with brine. The sample was then imaged again at full water saturation. As a next step,  $\text{CO}_2$  was injected at a constant flow rate of 1 mL/min and the volumes of injected and produced fluids were measured. In addition, the pressures at the inlet and outlet of the plugs were measured during the experiments; note that the pressure at the outlet was kept constant at 6.5 MPa by a high accuracy syringe pump. After injecting 2 pore volumes of  $\text{CO}_2$ , injection was stopped while the pore and confining pressures were kept constant by closing valves at the relevant inlet and outlet ports of the HPET cell. The plugs – now at initial  $\text{CO}_2$  saturation, i.e. partially saturated with  $\text{CO}_2$  – were imaged again. CT scan time for each image amounted to 30 min. The flow parameters for both experiments are summarized in Table 1.

Table 1: Flow parameters associated with the acoustic and CT experiments. \*Evaluation of influence of effective stress for dry and wet sample. \*\*Acoustic measurement during  $\text{CO}_2$  injection.

	Acoustic measurement*	Acoustic measurement**	CT measurement
Effective stress [MPa]	4-36	8	8
Brine pressure [MPa]	6.5	6.5	6.5
$\text{CO}_2$ injection rate [mL/min]	---	1	1
$\text{CO}_2$ injection pressure [MPa]	---	7.5	7.5
PV of $\text{CO}_2$ injected	---	2	2
Temperature [K]	318	318	296

### 3. Results and discussion

#### 3.1. Ultrasonic measurements

Results of measurement of P- and S-wave velocities ( $V_p$ ,  $V_s$ ) as a function of applied stress (defined as confining stress minus pore pressure) for dry and brine saturated sample are shown in the Figure 3. Both P and S velocities are increasing with increasing the stress.

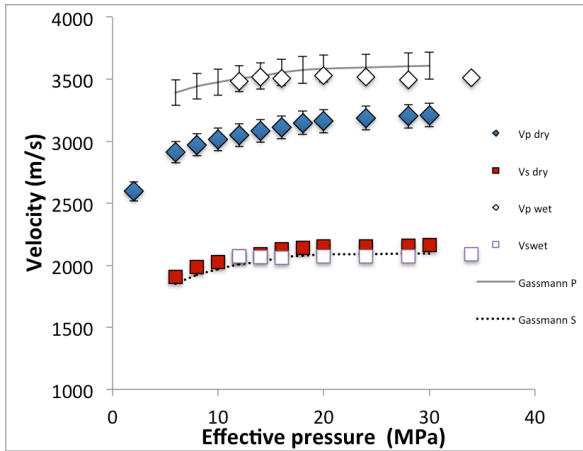


Fig. 3. P- and S-wave velocities  $V_p$  and  $V_s$  as a function of effective stress pressure for dry and brine saturated sandstone (Harvey 12-H) at 318K. Pore pressure was 6.5 MPa. Theoretical predictions of  $V_p$  and  $V_s$  velocities based on Gassmann’s fluid substitution procedure are shown as solid and dashed lines.

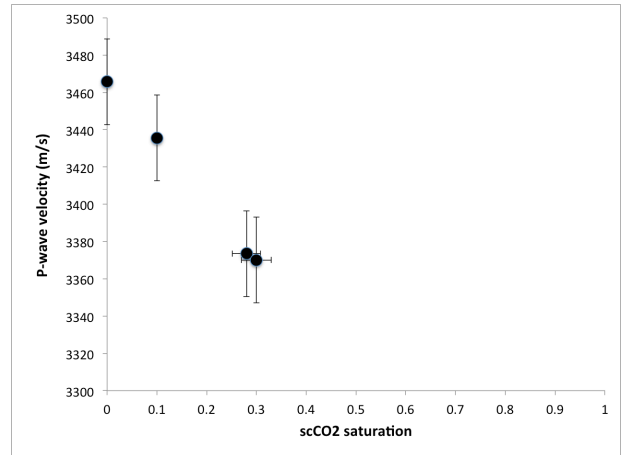


Fig. 4. P-wave velocity dependence on  $\text{CO}_2$  saturation (temperature = 318K, Confining pressure 15 MPa, brine pressure = 6.5 MPa).

The experimental results match very well those predicted with Gassmann’s fluid substitution procedure (equation 1, [14]):

$$K_{sat} = K_{dry} + \frac{\left(1 - \frac{K_{dry}}{K_0}\right)^2}{\frac{\phi}{K_{fl}} + \frac{1-\phi}{K_0} - \frac{K_{dry}}{K_0^2}} \quad (1)$$

where  $\phi$  is the rock porosity and  $K_{sat}$ ,  $K_{dry}$ ,  $K_0$  and  $K_{fl}$  are the bulk moduli of the saturated rock, dry rock, the minerals forming the rock, and the pore fluids, respectively.

$K_0$  is chosen to be equal to 37 GPa, the bulk modulus of quartz. The bulk modulus of a dry sandstone is calculated with equation (2), from dry compressional  $V_p$ , and shear,  $V_s$ , velocities and density,  $\rho$ , as follows:

$$K_{dry} = \rho V_p^2 - \frac{4}{3} \rho V_s^2 \quad (2)$$

where  $\rho$  is the rock density. As Gassmann’s fluid substitution procedure assumes that the shear moduli of the saturated and the dry sample ( $\mu_{sat}$ ,  $\mu_{dry}$ ) are identical and we obtain:

$$\mu_{sat} = \mu_{dry} = \rho V_s^2 \quad (3)$$

Compressional and shear velocities of saturated sandstones then can be obtained as

$$V_p = \left[ \left( K_{sat} + \frac{4}{3} \mu_{dry} \right) (\rho + \phi \rho_{fl})^{-1} \right]^{\frac{1}{2}} \quad (4)$$

and

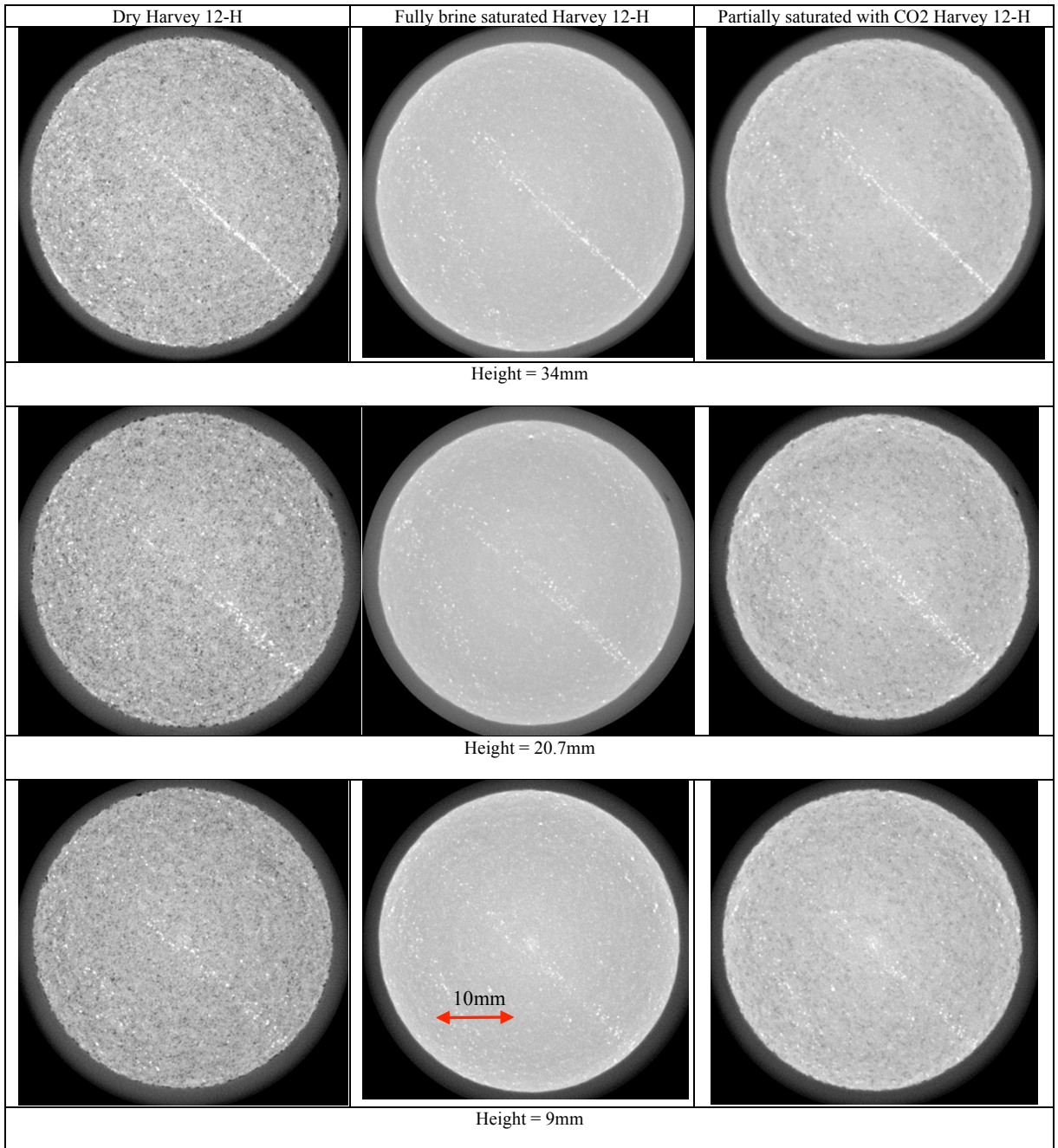


Fig. 5. Set of selected axial slices through sandstone: (left) dry sandstone; (middle) fully brine saturated sandstone; (right) partially saturated with CO<sub>2</sub>. The height from the plug's bottom face is written below each slice. The diameter of the shown circles is 38.1mm. The height of the plug was 84mm.

$$V_s = \left[ \mu_{dry} (\rho + \phi \rho_f)^{-1} \right]^{\frac{1}{2}}, \quad (5)$$

where  $\rho_f$  is density of the saturating fluid

The introduction of dense CO<sub>2</sub> into the pore space significantly changed  $V_p$ , Figure 4.  $V_p$  dropped continuously from ~3470 m/s at 0% CO<sub>2</sub> saturation to ~3370 m/s at ~30% CO<sub>2</sub> saturation. A change in the mechanical response behaviour is expected as dense CO<sub>2</sub> has a lower bulk modulus than brine [13], thus the effective bulk modulus of the rock sample decreases.

We note that in the ultrasonic experiment CO<sub>2</sub> breakthrough was detected on the production side after approximately 0.24PV (4.1mL) of brine were produced. This corresponds to an estimated CO<sub>2</sub> saturation of 24% at breakthrough. In total 6 mL of brine were produced after injection of 2PV of CO<sub>2</sub>, which implies an initial CO<sub>2</sub> saturation of 34% under these conditions. We note that the initial CO<sub>2</sub> saturation depends on the applied capillary pressure [20, 21], and that the residual CO<sub>2</sub> saturation is a function of the initial saturation [20, 22], porosity [23] and wettability [24–26].

### 3.2. X-ray computer tomography measurements

Figure 5 illustrates a set of selected axial slices through the dry, fully (100% brine) and partially saturated (i.e. plugs containing CO<sub>2</sub> and brine) plug; the nominal resolution of the images is 33.3  $\mu\text{m}$ . Darker greyscale tones relate to matter with lower relative radiodensities, i.e. air (dry core) or CO<sub>2</sub>. The brine (we used doped brine, 10wt% NaI in deionized water, to enhance CT contrast) had approximately the same CT number as quartz and could consequently not be distinguished from quartz. Moreover, minerals with high x-ray absorption, which are light grey or white were identified (the XRD analysis suggests that these minerals are calcite and K-feldspar). CO<sub>2</sub> swept through the plug in a piston-like displacement process, leading to a homogeneously distributed initial CO<sub>2</sub> saturation. However, a bedding layer consisting of minerals with high x-ray absorption was identified, but this bedding layer had no apparent influence on the CO<sub>2</sub> spreading characteristics. The initial CO<sub>2</sub> saturation after CO<sub>2</sub>-flooding estimated from the CT images was ~37%, which is consistent with the mass balance measurements.

## Conclusions

We measured ultrasonic P- and S-wave velocities ( $V_p$ ,  $V_s$ ) as a function of effective stress and scCO<sub>2</sub> saturation in a sandstone plug. We demonstrate that Gassmann's fluid substitution procedure matches the experimental results well for this sample extracted from a potential CCS site. A 3.5% reduction of P-wave velocity after injection of two pore volumes of CO<sub>2</sub> into brine-saturated sample was measured. We conclude that measurements of  $V_p$  can be used to estimate CO<sub>2</sub> saturations in seismic monitoring tools. The X-ray CT images demonstrated that the CO<sub>2</sub> advanced in a "piston like" displacement in this core. The plug contained a bedding layer, which, however, had no apparent influence on the CO<sub>2</sub> spreading characteristics. An initial CO<sub>2</sub> saturation after injecting 2PV of CO<sub>2</sub> at ~0.5 MPa capillary pressure was estimated as 32% (acoustic measurements) -37% (CT scanning); thus these techniques reached consistent saturation values; the CO<sub>2</sub> breakthrough saturation was estimated as 24%.

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