# Visualization of CO<sub>2</sub> and oil immiscible and miscible flow processes in porous media using NMR micro-imaging

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**Abstract:**  $CO_2$  flooding is considered not only one of the most effective enhanced oil recovery (EOR) methods, but also an important alternative for geological  $CO_2$  storage. In this paper, the visualization of  $CO_2$  flooding was studied using a 400 MHz NMR micro-imaging system. For gaseous  $CO_2$  immiscible displacement, it was found that  $CO_2$  channeling or fingering occurred due to the difference of fluid viscosity and density. Thus, the sweep efficiency was small and the final residual oil saturation was 53.1%. For supercritical  $CO_2$  miscible displacement, the results showed that piston-like displacement occurred, viscous fingering and the gravity override caused by the low viscosity and density of the gas was effectively restrained, and the velocity of  $CO_2$  front was uniform. The sweep efficiency was so high that the final residual oil saturation was 33.9%, which indicated  $CO_2$  miscible displacement could enhance oil recovery more than  $CO_2$  immiscible displacement. In addition, the average velocity of  $CO_2$  front was evaluated through analyzing the oil saturation profile. A special core analysis method has been applied to in-situ oil saturation data to directly evaluate the local Darcy phase velocities and capillary dispersion rate.

Key words: NMR micro-imaging, porous media, CO<sub>2</sub> flooding, enhanced oil recovery, saturation

# **1** Introduction

 $CO_2$  is a major contributor to the greenhouse effect leading to global warming. Enhanced Oil Recovery (EOR) using  $CO_2$  has been an important alternative for geological  $CO_2$ storage.  $CO_2$  flooding is considered one of the most effective tertiary recovery processes in light/medium oil reservoirs and has been widely used.  $CO_2$  is injected into a reservoir to increase production by reducing oil viscosity and providing miscible or immiscible displacement of the oil. However, the complicated displacement mechanisms have not been fully understood.

Traditionally, core analysts have been forced to assume that those objects were homogeneous black boxes. Volume and composition of fluids that were injected and recovered could be measured, but how the fluids were distributed and moving inside the core could only be inferred. With techniques such as noninvasive neutron radiography (Brunner and Mardock, 1946), X-ray (Morgan et al, 1950; Laird and Putuan, 1951), gamma ray, and microwave absorption, it became possible to obtain one-dimensional fluid saturation

\*Corresponding author. email: songyc@dlut.edu.cn. Received October 16, 2010 and solute distribution data. The development of X-ray computerized tomography (CT) allowed for determination of two-dimensional and three-dimensional rock densities, saturations of fluids, and concentrations of solutes (Wellington and Vinegar, 1985, 1987; Hunt and Bajsarowicz, 1988) in the core. Recently X-ray CT had been carried out to investigate CO<sub>2</sub> foam flow in a consolidated Bentheimer sandstone core saturated with surfactant solution (Carretero-Carralero et al, 2007; Du et al, 2007; 2008). However, since this method is sensitive to mass density contrast, experiments usually have to be carried out with large tracer concentrations leading to unwanted fingering (Wooding, 1969; Perkins and Johnston, 1969) and change of the fluid properties (Kantzas, 1995). It is because the X-ray attenuation is principally determined by the atomic number of the sample nuclei and is, therefore, dominated by the rock matrix rather than the  $CO_2$  or oil.

NMR imaging is similar to X-ray CT in that twodimensional and three-dimensional images can be obtained from selected regions of an object. However, X-ray CT can only image electron density and atomic number, while NMR imaging, besides spin density of a variety of nuclei (<sup>1</sup>H, <sup>19</sup>F, <sup>23</sup>Na, <sup>31</sup>P, etc.), also measures a number of other quantities such as relaxation times and chemical shifts (Vinegar, 1986; Blackband et al, 1986). It is inherently more informative than radiographic techniques. It is also much less hazardous. Special features of many NMR imaging techniques include the ability to measure the fluid flow velocities and to distinguish different liquids using their differences in intrinsic NMR properties. This type of information would be difficult to obtain by other methods. NMR imaging is a powerful analytical tool for noninvasive multidimensional visualization of flow and transport in porous media (Callaghan, 1994). NMR imaging can provide unprecedented quantitative information about fluid-phase distributions in porous media during displacement processes, as well as information about rock structure corresponding to local regions within porous media. Such information can significantly advance our understanding of the storage and transport of multiphase fluids in porous media (Chen et al, 1992; 1993). So in the past two decades, there have been many studies of visualization of flow and transport in porous media using NMR imaging techniques, for example, the transfer and fluid flow behavior during water flooding in fractured rock or rock of various wettability was experimentally investigated using NMR imaging technique (Fernø et al, 2007; Brautaset et al, 2008a; Aspenes et al, 2008). Some research was done on visualization of water flooding and polymer flooding through unconsolidated porous media using the unique centric scan SPRITE NMR imaging technique and to quantify in-situ fluid saturation distribution in rocks (Romero-Zeron et al. 2009; 2010). The distribution of remaining oil and NMR relaxation characteristic following a CO<sub>2</sub> miscible displacement of crude oil in San Andres crystalline dolomite cores were investigated using NMR imaging (Hazlett et al, 1993). Restricted by the condition of the experimental high-pressure core holder, it could not monitor dynamic fluid-phase distributions in porous media during displacement processes. Until recently, by using the high-pressure core holder, only few studies were done on the behavior of two-phase flow in porous media at high pressure using NMR imaging. Some research was done on the behavior of CO<sub>2</sub> and water two-phase flow in porous media (Suekane et al, 2005; 2006; 2009). The fluid saturation distributions and the flow characteristics in-situ were investigated and monitored during waterfloods and subsequent injection of either liquid or supercritical  $CO_2$  in four Portland Chalk core samples at different wettabilities using NMR imaging (Brautaset et al, 2008b).

Due to the complicated displacement mechanisms involved during  $CO_2$  injection, the need for in-situ data is of great importance in order to fully understand the displacement process. In this study, tests have been conducted on gaseous  $CO_2$  immiscible displacement and supercritical  $CO_2$  miscible displacement in high-permeability sand packs. NMR imaging has been utilized to qualitatively monitor the displacement processes. This article addresses the application of the NMR imaging technique to qualitatively monitor fluid propagation and dynamic oil saturation profile during  $CO_2$  flooding in sand pack core models.

# **2** Experimental

#### 2.1 Experimental apparatus

A simplified schematic diagram of the experimental setup is shown in Fig. 1. The experimental setup consisted of two circuits, namely, the displacement line and the temperature control line. In the displacement line, the liquid  $CO_2$  was forced into a transfer vessel in a thermostatic chamber by a  $CO_2$  pump, and then oil and  $CO_2$  was forced into the sandpack cell by a syringe pump. The flow rate and back pressure were controlled by pumps and a back pressure regulator. The pressure drop through the sand pack was measured with a low differential pressure transmitter.

In the temperature control line, fluorinert FC-40 was used to control the temperature of the sand-pack cell because fluorinert contains no hydrogen atoms, thus it is not imaged, and its low dielectric properties minimize radio frequency (RF) losses (Suekane et al, 2005; Baldwin et al, 2009; Ersland et al, 2010). This fluid was maintained at a constant flow rate while being circulated through the thermostatic bath system



Fig. 1 A simplified schematic of the experimental setup

and around the sand-pack cell by a recirculation pump.

In this study, a new high-pressure sand-pack cell was designed and constructed for the Varian NMR Systems with a RF probe of 40 mm inner diameter. The new high-pressure sand-pack cell and its cross-sectional diagram are shown in Fig. 2. The cell was designed and constructed for NMR imaging measurements. It was inserted vertically into the NMR imaging system as shown in Fig. 1. The maximum working pressure and temperature of the sand-pack cell were 15 MPa and 70 °C, respectively. The cell consisted of water fittings (1), end cap (2), sealing O-rings (3, 4, 6, 10, and 11), end pieces (5), filter screen (7), high-pressure polyimide tube (8), and normal-pressure polyimide sleeve (9). The high-pressure polyimide tube was 200 mm long and 15 mm in inner diameter. The tube material was nonmagnetic and hence should not interfere with the RF signals used in the experiments. Temperatures at the inlet and exit of the sand-pack cell were measured with thermocouples.



Fig. 2 The high-pressure NMR imaging sand-pack cell (unit: mm)1-Water fittings; 2-End cap; 3, 4, 6, 10, 11-Sealing O-rings; 5-End pieces;7-Filter screen; 8-High-pressure polyimide tube; 9-Normal-pressure polyimide sleeve

## 2.2 Experimental materials

Unconsolidated, high-permeability sand packs were used to evaluate oil displacement in porous media. The sand packs were prepared by packing 0.177-0.250 mm Soda-glass beads (BZ02, made in Japan) into the cylindrical cell.  $CO_2$  (99% purity) was used as the gas phase, and *n*-Decane was used as the oil phase. Fluid properties, including densities and viscosities at relevant temperatures and pressures, are listed in Table 1. The glass bead-packed models had a porosity of 35.2%, which was calculated from a traditional gravimetric method, and an absolute permeability to water of 13.5 D.

Table 1	Fluid	properties
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Fluid	Pressure MPa	Temperature °C	Density g/cm <sup>3</sup>	Viscosity cP
CO <sub>2</sub>	5.4	40	0.127	0.017
	8.1	40	0.29	0.023
n-Decane	5.4	40	0.72	0.74
	8.1	40	0.72	0.76

The critical point of CO<sub>2</sub> is at 31.1 °C and 7,398 kPa. The minimum miscible pressure (MMP) for the *n*-Decane/CO<sub>2</sub> system has been determined at 35 °C and 7,329 kPa (Asghari and Torabi, 2008) and at 37.8 °C and 7894 kPa (Ayirala et al, 2006). In the tests, the temperature of 40 °C and pressure of 5.4 MPa were selected to ensure gas properties of CO<sub>2</sub> for the CO<sub>2</sub> (gas) immiscible displacement test. The temperature of 40 °C and pressure of 8.1 MPa were selected to ensure the supercritical properties for the supercritical CO<sub>2</sub> miscible displacement test.

## 2.3 NMR imaging technique

All NMR imaging measurements were performed on a Varian NMR system with 9.4 Tesla, wide-bore (89 mm in diameter), vertical superconducting magnet. A <sup>1</sup>H 40 mm Millipede vertical micro-imaging probe was used and the gradient coils provided a maximum gradient strength of 50 G/cm. The NMR imaging was conducted by the fast spin echo multi-slice pulse sequence (FSEMS) during flooding and the experimental conditions are as follows: echo time (TE) 2.66 ms, repetition time (TR) 1 s, image data matrix  $192 \times 192$ , field of view (FOV) 40mm×40mm with 1 mm

thickness, number of slices 15 (the position of slices is shown in Fig. 3), spatial resolution  $0.21 \times 0.21 \times 1 \text{ mm}^3$ , number of images for averaging 1, acquisition time 24 s. The bulk relaxation times were measured with the CPMG method (Carr and Purcell, 1954; Meiboom and Gill, 1958). When the sand pack was 100% saturated with oil, the spin-spin relaxation time (T<sub>2</sub>) was 55 ms. When the sand pack was 33% saturated with oil, T<sub>2</sub> was 22 ms. T<sub>2</sub> changed with oil saturation, but TE used in this study was about eight times smaller than the bulk T<sub>2</sub>. The sand pack did not have large T<sub>2</sub> distribution due to fairly uniform diameter of the glass beads used, the inhomogeneity of the magnetic field caused by the susceptibility gradients would be largely refocused by the 180 pulse. So we treated the images as spin density images and the quantitative analysis of saturation was true. Oil contained in the sand pack was visualized in planes along the flow direction. However, these traditional NMR imaging methods fail in actual rocks with paramagnetic impurities, since the transverse signal lifetimes of these rocks are too short to be detected. Quantitative information is the ultimate goal for rock core analysis. The single-point ramped imaging with  $T_1$  enhancement (SPRITE) imaging technique has proven to be a very robust and flexible method for the study of a wide range of systems with short signal lifetimes. As a pure phase-encoding technique, SPRITE is largely immune to image distortions generated by susceptibility variations, chemical shift, and paramagnetic impurities, unlike clinical magnetic resonance imaging methods. It enables systems with transverse lifetimes as short as tens of microseconds to be successfully visualized (Chen et al, 2006).



Fig. 3 Sketch map for the position of slices

# **3** Results and discussion

## 3.1 Immiscible displacement by gaseous CO<sub>2</sub>

# 3.1.1 NMR image analysis

In the immiscible displacement process, gaseous  $CO_2$  was injected vertically upward into the sand pack saturated with oil at 5.4 MPa and 40 °C. The injection rate of  $CO_2$  was maintained at 0.1 mL/min. Fig. 4 shows a series of NMR images that illustrate oil saturation after different pore volumes (PV) of  $CO_2$  were injected into the sand pack. The  $CO_2$  moved rapidly upwards due to low fluid viscosity and high buoyancy, and then the  $CO_2$  broke through the sand pack in the field of view (FOV). Because the sand pack was not perfectly homogenous, the injected  $CO_2$  tended to channel

through the high-permeable zones. Therefore, some thin channels were formed and  $CO_2$  ran through the channels vertically in a short period according to the images (b) and (c) in Fig. 4. Once these channels were being formed, the secondary oil desaturation started,  $CO_2$  would continuously run through these channels as shown in the images (d) and (e) in Fig. 4, bypassing most of the residual oil in the matrix. The oil saturation decreased gradually and the residual oil tended to be immobilized.

#### 3.1.2 Saturation profiles

To quantitatively analyze the evolution of oil saturation along the sand pack, the NMR imaging data was converted into saturation profiles in the manner described by Suekane et al (2009). The NMR signal intensity from any local position was proportional to the oil content in the porous media. This



Fig. 4 Distribution of NMR signal intensity in the sand pack at 5.4 MPa, 40  $^{\circ}$ C with gaseous CO<sub>2</sub> injection rate of 0.1 mL/min

means that the measured NMR signal intensity reflects the local oil saturation in the porous media. In the experiments, the distribution of the initial NMR signal intensity in the porous media saturated with oil was obtained. Then,  $CO_2$  was injected into the porous media with time-series acquisition of NMR images. The injected  $CO_2$  would displace some oil in the porous media, thus leading to a decrease in the NMR signal intensity. First, the NMR signals of all the 15 slices were added together, corresponding to the obtained two-dimensional projective distribution of three-dimensional oil saturation distribution in the sand pack, then the oil saturation

in each pixel was calculated as the ratio of the NMR signal with the CO<sub>2</sub> to that without the CO<sub>2</sub> (t = 0). Next, the oil saturation was added in a lateral direction within the sand pack, that is, for a given position z along the flow direction from the inlet of the sand pack (z = 0). One-dimensional saturation profiles along the sand pack at different injection volume of CO<sub>2</sub> was obtained and shown in Fig. 5(a). Finally, the oil saturation was also averaged in the total FOV. The saturation profile of total FOV versus injection volume of CO<sub>2</sub> was obtained and shown in Fig. 5(b).

The oil saturation profiles in the sand pack at 5.4 MPa,



Fig. 5 Evolution of oil saturation profiles in the sand pack at 5.4 MPa, 40 °C with gaseous CO<sub>2</sub> injection (a) one-dimensional saturation profile along the sand pack, profiles were obtained by piecewise spline fitting; (b) saturation profile of total FOV versus volume of CO<sub>2</sub> injected

40 °C with CO<sub>2</sub> injection were obtained and shown in Fig. 5. Fig. 5(a) shows two stages of the oil displacement by CO<sub>2</sub>. In the first stage, starting from the beginning of the injection up to the CO<sub>2</sub> breakthrough, the CO<sub>2</sub> tended to penetrate through the more permeable regions. The second stage started after CO<sub>2</sub> breakthrough and ended with the unchanged oil saturation distribution. Finally, the residual oil saturation in the inlet of the sand pack before the 20 mm was higher compared to the outlet in the migration direction.

Fig. 5(b) shows oil saturation in the total FOV as a function of volume of  $CO_2$  injection determined from NMR imaging. The profile of the  $CO_2$  motion can also be divided into three regions. In part ab, starting from the beginning of the injection up to the  $CO_2$  breakthrough, the oil saturation decreased linearly from 100% to 74.4% after the  $CO_2$  injection of 0.21 PV (0.52 mL). In part bc, the oil saturation decreased exponentially to 54.8% with the  $CO_2$  injection of 0.69 PV (1.72 mL). In part cd, continuous displacement of oil during  $CO_2$  flooding until residual oil saturation (53.1%) was reached (oil flow ceases), the additional injection of 0.69 PV (1.72 mL) of gas recovered only little oil because at this point the relative permeability to oil was near to zero since oil saturation was equal to residual oil saturation, and oil would not flow. The final oil recovery was 46.9%.

# 3.2 Supercritical CO<sub>2</sub> miscible displacement test

## 3.2.1 NMR image analysis

In the CO<sub>2</sub> miscible displacement test, supercritical CO<sub>2</sub> was injected vertically upward into the oil-saturated sand pack at a rate of 0.1 mL/min (at 8.1 MPa and 40 °C). To obtain

detailed information about the dynamic CO<sub>2</sub> displacement in the sand pack, the NMR images of oil distribution in the slices in the longitudinal direction were examined. Fig. 6 shows a series of NMR images, which illustrate the oil saturation at different injection volumes of CO<sub>2</sub> of 0, 0.26, 0.32, 0.48, 0.61, 0.76, 1.25, and 3.20 PV (corresponding time series was 0, 408, 504, 744, 936, 1,152, 1,896, and 4,800 s, respectively). The bright regions (orange and yellow) indicate the high NMR signal intensity corresponding to high oil saturation, while the dark regions (blue) stand for the low oil saturation. For instance, the first image shows the sand pack was 100% saturated with oil. The porosity distribution shown in Fig. 7 was calibrated with a reference standard of known porosity that was imaged with the sand pack. Then, with the injection of supercritical CO<sub>2</sub>, the piston-like displacement occurred, the phenomenon of viscous fingering and gravity override caused by the low viscosity and density of the gas was effectively restrained, thereby diverting the injected CO<sub>2</sub> to lower-permeability zones and improving displacement efficiency.

#### 3.2.2 Saturation profiles

Fig. 8(a) shows two stages of the oil displacement by supercritical  $CO_2$ . In the first stage from the beginning of the injection up to the  $CO_2$  breakthrough, piston-like displacement occurred. As can be seen from the profile of 0.61 PV in Fig. 8(a), this stage consisted of three regions: (a) a region with low oil saturation, where the front of  $CO_2$  has passed; (b) a region with high oil saturation, where the front of  $CO_2$  has not yet arrived; and (c) a transition region, where the front of  $CO_2$  is located. With  $CO_2$  injection, the oil saturation



NMR signal intensity/arbitrary unit

Fig. 6 Distribution of NMR signal intensity in the sand pack displaced by supercritical CO<sub>2</sub>



Fig. 7 One-dimensional distribution profile of porosity along the sand pack

decreased gradually from the inlet of the sand pack, and the low oil saturation region increased gradually. The front of  $CO_2$  proceeding in the sand pack can be measured clearly. The slope of the front tended to be at a shallow angle to the migration direction. The second stage started after  $CO_2$ breakthrough, and ended with the unchanged oil saturation distribution. This stage was characterized by secondary oil desaturation which started in the inlet of the sand pack and propagated toward the outlet. Finally, the oil saturation decreased down to about 30%-40%.

Fig. 8(b) shows oil saturation in the total FOV as a function of the volume of supercritical CO<sub>2</sub> injection (or time) determined from NMR imaging. This figure shows the oil saturation decreased gradually with the injection of CO<sub>2</sub>, and continuous displacement of oil during CO<sub>2</sub> displacement until residual oil saturation (33.9%) was reached (oil flow ceases) after the injection of 3.2 PV of CO<sub>2</sub>. In the oil saturation profile, four characteristic stages can be found when CO<sub>2</sub> front moved through the FOV (the total height of the sand pack is 200 mm and the height of the FOV is only 40 mm, the FOV is located at the centre of the sand pack), which were showed in Fig. 6 (real schematic) and Fig. 9 (simplified schematic). These stages are when (a) the head of  $CO_2$ displacement front moved into the FOV; (b) the tail of  $CO_2$ displacement front moved into the FOV; (c) the head of CO<sub>2</sub> displacement front broke through the FOV; (d) the tail of CO<sub>2</sub> displacement front broke through the FOV. So the process of the supercritical CO<sub>2</sub> motion can be divided into four regions as follows: in part ab, the oil saturation decreased gradually from 100% to 95% with the injection of CO<sub>2</sub>, the decrease in the oil saturation was more rapid in the end of the part AB due to irregular characteristics of the CO<sub>2</sub> front; in part



Fig. 8 Evolution of oil saturation profiles in the sand pack at 8.1 MPa, 40 °C with supercritical CO<sub>2</sub> injection (a) one dimensional saturation profile along the sand pack, profiles were obtained by polynomial fitting; (b) saturation profile of total FOV versus volume of CO<sub>2</sub> injected



Fig. 9 Simplified schematics of the characteristic times during CO<sub>2</sub> front moved through the FOV

BC, the oil saturation decreased linearly to 58.5% due to the uniform velocity of CO<sub>2</sub> front; in part CD, the oil saturation decreased exponentially to 43.8% with the process of total CO<sub>2</sub> displacement front breakthrough; in part DE, the oil saturation steady decreased to 33.9% with the process of secondary oil desaturation. The final oil recovery was 66.1%. The sweep efficiency in this test with supercritical CO<sub>2</sub> miscible displacement test.

Through analyzing part BC in the oil saturation profile, we can evaluate the approximate whole average velocity of  $CO_2$  front in the following manner. The variation of oil volume  $(\Delta V_o)$  in the FOV can be expressed as follows:

$$\Delta V_{\rm o} = A\phi\Delta h \tag{1}$$

where A is the cross-sectional area of the sand pack, cm<sup>2</sup>;  $\phi$  is the porosity, fraction;  $\Delta h$  is the displacement of CO<sub>2</sub> front, cm.

The variation of  $CO_2$  volume ( $\Delta Q$ ) in the FOV can be expressed as follows:

$$\Delta Q = \frac{q\Delta t}{V_{\rm p}} \tag{2}$$

The oil saturation  $(S_0)$  can be expressed as follows:

$$S_{\rm o} = \frac{V_{\rm o}}{V_{\rm p}} \tag{3}$$

where  $V_0$  is the oil volume in the sand pack, cm<sup>3</sup>.

From Eqs. (1)-(3), we can evaluate the approximate whole average velocity of CO<sub>2</sub> front ( $\overline{v}$ ):

$$\overline{v} = \frac{\Delta h}{\Delta t} = -\frac{\Delta S_{o}}{\Delta Q} \frac{q}{A\phi}$$
(4)

where  $\Delta S_{o}$  is the variation of oil saturation in the FOV, fraction.

 $\frac{\Delta S_o}{\Delta Q}$  in Eq. (4) can be obtained by assuming oil saturation

is linear with the volume of  $CO_2$  injection. Then we can obtain the approximate whole average velocity of  $CO_2$  front, which is 0.12 cm/min.

Table 2 summarizes oil recovery results with the four parts of supercritical  $CO_2$  injection for the total experimental process. In part BC, the oil recovery values obtained from the NMR imaging technique and material balance are 36.5% and 35.4%, respectively. The deviation is only 3%.

#### Table 2 Oil recovery with supercritical CO<sub>2</sub> injection

Characteristic regions	Volume of CO <sub>2</sub> injected, mL	Volum of CO <sub>2</sub> injected, PV	Oil recovery based on NMR imaging, %	Oil recovery based on material balance, %
AB	0.65	0.26	5	_
BC	1.24	0.5	36.5	35.4
CD	1.22	0.49	14.7	—
DE	4.85	1.95	9.9	—
Total	7.96	3.2	66.1	—

## 3.2.3 Core analysis methods

The coreflood interpretation method proposed by Goodfield et al (2001) was applied to the data generated from core displacement tests to determine the local Darcy phase velocities in the sand pack. The phase volumes per unit cross-sectional area between the core inlet ( $\zeta$ =0) and the current position ( $\zeta$ =z) are given by:

$$V_{\rm g}(z,t) = \int_0^z \phi(\zeta) S_{\rm g}(\zeta,t) \mathrm{d}\zeta$$
<sup>(5)</sup>

where  $\phi(z)$  is the porosity at position *z* shown in Fig. 7 and  $S_g(\zeta, t)$  is the CO<sub>2</sub> saturation at position *z* and time *t* shown in Fig. 8(a). Using a material balance approach, the local Darcy phase (CO<sub>2</sub> and oil) velocities can then be expressed in the following form:

$$U_{g}(z,t) = U(t)F_{g}^{inj}(t) - \frac{\partial V_{g}(z,t)}{\partial t}$$
(6)

$$U_{o}(z,t) = -\frac{\partial V_{o}(z,t)}{\partial t}$$
(7)

where  $U_g(z, t)$  and  $U_o(z, t)$  are the local Darcy phase velocities of CO<sub>2</sub> and oil, respectively; U(t) is the total Darcy velocity. and  $F_g^{inj}(t)$  is the fractional flow of CO<sub>2</sub> injection at time t. Then, the local Darcy phase velocities of CO<sub>2</sub> and oil were obtained with this method and shown in Fig. 10.

Darcy's law in the new interpretation methods for the phase velocities is expressed according to:

$$U_{\alpha} = -k\lambda_{\alpha}(\frac{\partial p_{\alpha}}{\partial z} + \rho_{\alpha}g)$$
(8)

where  $\alpha$  denotes each phase of CO<sub>2</sub> (g) and oil (o); *U* is the local Darcy velocity;  $\lambda_a$  is the mobility,  $\lambda_a = k_{ra}/\mu_a$ ; *k* is the absolute permeability; *p* is the pressure;  $\rho$  is the density; *g* is the gravitational acceleration;  $k_r$  is the relative permeability;  $\mu$  is the viscosity.



Fig. 10 Local phase velocities of (a) CO<sub>2</sub> and (b) oil

The capillary pressure  $P_{co}(S_o)$  is defined in the standard convention by:

$$p_{\rm co}(S_{\rm o}) = p_{\rm g}(S_{\rm o}) - p_{\rm o}(S_{\rm o})$$
<sup>(9)</sup>

From Eqs. (8) and (9), the local Darcy velocity of  $CO_2$  can be expressed as follows:

$$U_{g} = U(t) \frac{\lambda_{g}}{\lambda_{g} + \lambda_{o}} \left( 1 + \frac{kg(\rho_{o} - \rho_{g})}{U(t)} \lambda_{o} \right)$$
  
$$-k \frac{\lambda_{g} \lambda_{o}}{\lambda_{g} + \lambda_{o}} \frac{d p_{co}}{d S_{o}} \frac{\partial S_{o}}{\partial z}$$
(10)

where U is the total flow rate.

The viscous-dominant fractional flow function, gravity countercurrent flow function, and the capillary dispersion rate can be defined as follows, respectively:

$$f_{g}(S_{o}) = \frac{\lambda_{g}}{\lambda_{g} + \lambda_{o}}$$
(11)

$$G_{g}(S_{o}) = g(\rho_{o} - \rho_{g}) \frac{\lambda_{g} \lambda_{o}}{\lambda_{g} + \lambda_{o}}$$
(12)

$$d_{\rm cpo}(S_{\rm w}) = -\frac{\lambda_{\rm g}\lambda_{\rm o}}{\lambda_{\rm g} + \lambda_{\rm o}} \frac{\mathrm{d}\,p_{\rm co}}{\mathrm{d}\,S_{\rm o}} \tag{13}$$

Then Eq. (10) can be expressed as follows:

$$U_{g} = U(t)f_{g}(S_{o}) + kG_{g}(S_{o}) + kd_{cpo}(S_{o})\frac{\partial S_{o}}{\partial z}$$
(14)

And Eq. (14) can also be written as follows:

$$U_{\rm g} = U(t) f_{\rm g}^{\rm (g)}(S_{\rm o}, U) + kd_{\rm cpo}(S_{\rm o}) \frac{\partial S_{\rm o}}{\partial z}$$
(15)

with

$$f_{g}^{(g)}(S_{o},U) = f_{g}(S_{o}) + \frac{k}{U}G_{g}(S_{o})$$
(16)

For any saturation  $S_o^*$ , the corresponding position in the sand pack can be expressed as a function of time:

$$S_{o}(z^{*}(t), t) = S_{o}^{*}$$
<sup>(17)</sup>

Using this function, Eq. (15) can be written as follows:

$$U_{g}(z^{*}, t) = U(t)f_{g}^{(g)}(S_{o}^{*}, U) + kd_{cpo}(S_{o}^{*})\frac{\partial S_{o}}{\partial z}\Big|_{z^{*}, t}$$
(18)

Except in the special case of zero total Darcy velocity (considered below), Eq. (18) can be rearranged to give:

$$\frac{U_{g}(z^{*},t)}{U(t)} = f_{g}^{(g)}(S_{o}^{*},U) + d_{cpo}(S_{o}^{*})\frac{k}{U(t)}\frac{\partial S_{o}}{\partial z}\Big|_{z^{*},t}$$
(19)

where  $\frac{U_{g}(z^{*},t)}{U(t)}$  and  $\frac{k}{U(t)} \frac{\partial S_{o}}{\partial z} \Big|_{z^{*},t}$  are calculated from the observed saturation data. Provided gravity forces are negligible, or for periods of time when the total flow rate is constant, this equation gives  $\frac{U_{g}(z^{*},t)}{U(t)}$  as a linear function of  $\frac{k}{U(t)} \frac{\partial S_{o}}{\partial z} \Big|_{z^{*},t}$ , where the intercept is given by  $f_{g}^{(g)}(S_{o}^{*},U)$ and the gradient by  $d_{cpo}(S_{o}^{*})$ . Thus plotting the observed values of  $\frac{U_{g}(z^{*},t)}{U(t)}$  against  $\frac{k}{U(t)} \frac{\partial S_{o}}{\partial z} \Big|_{z^{*},t}$ ,  $f_{g}^{(g)}(S_{o}^{*},U)$  and  $d_{cpo}(S_{o}^{*})$  are estimated from the intercept and gradient of linear approximation of these data. The calculated capillary

dispersion rate  $d_{cpo}(S_o^*)$  is shown in Fig. 11.

It is worth noting that this capillary dispersion rate, which is used for reservoir simulation, can be estimated from only in-situ phase distribution data and the injection flow rate without additional measurements such as pressure drop across the sand pack. In other words, without the measurement of capillary pressure function  $p_{cw}$ , the capillary dispersion rate function can be evaluated. The capillary dispersion rate is a maximum at an oil saturation of 0.9.



Fig. 11 Capillary dispersion rate at 8.1 MPa, 40 °C

# **4** Conclusions

This article has provided experimental results on both gaseous  $CO_2$  immiscible displacement and supercritical  $CO_2$  miscible displacement of oil in high-permeable sand packs. The main conclusions are as follows:

1) A new high-pressure sand-pack cell was designed and constructed for the NMR imaging system with a RF probe of 40 mm inner diameter. The maximum working pressure and temperature of the sand-pack cell are 15 MPa and 80 °C, respectively. By the high-pressure cell the process of  $CO_2$  injected into the sand pack at high pressure and temperature can be visualized using NMR imaging.

2) The fundamental characteristics of the flooding process such as onset of  $CO_2$  channeling or fingering, the piston-like front of  $CO_2$ , and the distribution of oil in porous media can be accurately detected.

3) It was found that for gaseous  $CO_2$  immiscible displacement,  $CO_2$  channeling or fingering occurred due to the difference in the viscosity and density of fluids, so the sweep efficiency was low and the final residual oil saturation was 53.1%. For supercritical  $CO_2$  miscible displacement, pistonlike displacement occurred,  $CO_2$  channeling or fingering was restrained effectively, and the velocity of  $CO_2$  front was uniform. The sweep efficiency was high and the final residual oil saturation was 33.9%. So supercritical  $CO_2$  miscible displacement could enhance oil recovery more efficiency than gaseous  $CO_2$  immiscible displacement.

4) The oil saturation distributions were monitored in-situ as function of volume of  $CO_2$  injected, through analyzing the oil saturation profile the velocity of  $CO_2$  front could be evaluated. A special core analysis method has been applied to in-situ oil saturation data to evaluate directly the local Darcy phase velocities and capillary dispersion rate.

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