A New Unsteady-State Method of Determining Two-Phase Relative Permeability
Illustrated by CO₂-Brine Primary Drainage in Berea Sandstone

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Abstract: This study presents a new unsteady-state method for measuring two-phase relative permeability by obtaining local values of the three key parameters (saturation, pressure drop, and phase flux) versus time during a displacement. These three parameters can be substituted to two-phase Darcy Buckingham Equation to directly determine relative permeability. To obtain the first two, we use a medical X-ray Computed Tomography (CT) scanner to monitor saturation in time and space, and six differential pressure transducers to measure the overall pressure drop and the pressure drops of five individual sections (divided by four pressure taps on the core) continuously. At each scanning time, the local phase flux is obtained by spatially integrating the saturation profile and converting this to the flux using a fractional flow framework. One advantage of this local method over most previous methods is that the capillary end effect is experimentally avoided; this improvement is crucial for experiments using low viscosity fluids such as supercritical and gas phases. To illustrate the new method, we conduct five CO₂-brine primary drainage experiments in a 60.8 cm long and 116 mD Berea sandstone core at 20 °C and 1500 psi. In return, we obtain hundreds of unsteady-state CO₂ and brine relative permeability data points that are consistent with steady-state relative permeability data from the same experiments. Due to the large amount of relative permeability data

obtained by the new unsteady-state method, the uncertainties of the exponents in the Corey-type fits decrease by up to 90% compared with the steady-state method.

Keywords: relative permeability, unsteady-state method, local phase flux, fractional flow theory, CO₂-brine system, primary drainage

1. Introduction

Relative permeability in porous media is simply a *measure* of the reduction of permeability to a certain phase when that phase is not at complete saturation. It is used in the Darcy Buckingham equation as:

$$Q_i = \frac{k_{ri}KA}{\mu_i} \cdot \frac{\Delta P^i}{L} \quad (1)$$

where i is the subscript for the phase, Q_i is the phase i volumetric flow rate, k_{ri} is the phase i relative permeability, K is the permeability, A is the cross section area, ΔP^i is the phase i pressure drop, μ_i is the phase i viscosity and L is the length of porous media.

Typically, the relative permeability is assumed to be a function of the phase saturation (S_i). To measure relative permeability, one needs to have *local* measurements of saturation, flow rate and pressure drop of phase i . In terms of corefloods, obtaining *global* (core integrated) measurements of S_i , Q_i , and ΔP^i are typically straightforward; unfortunately the global values can vary significantly from the local values, which is primarily due to saturation variations in the core, caused by end effects and temporal changes in the core. The key to any relative permeability measurement method is

1 designing the method to obtain local values of the three quantities. Here, we first review
2 how these quantities are obtained in both steady and unsteady methods, which are the two
3 categories of relative permeability determination methods. Second, we give an overview
4 of the new method.

5   During steady-state methods, equilibrated two phase fluids are injected into a core
6 until the measured overall pressure drop and the overall saturation do not change with
7 time [1–23]. The phase flow rates, the saturation, and the pressure drop are constants
8 along the core when steady state is reached. Thus, for steady-state methods, the global
9 measurements are equivalent to the local measurements and the relative permeability
10 equation can be used directly. Potentially there are some complicating capillary effects
11 that occur at the inlet and outlet of the core; these effects can be remedied by measuring
12 pressure drops and saturations in the center of the core [16,24]. This simple equivalence
13 between global and local values makes steady-state methods the gold-standard of relative
14 permeability measurements. Since at each steady state only two data points are obtained
15 (the relative permeability of each phase), the process must be repeated for each flux ratio
16 to obtain a full relative permeability curve. This makes the steady-state methods very
17 time consuming (in addition, steady state is achieved very slowly at the end points) and
18 expensive.

19   To speed up the measurement process, unsteady-state methods have been
20 developed, particularly in determining oil-brine relative permeabilities [25–31]. Many of
21 these unsteady-state methods obtain the local saturations and phase flow rates from their
22 global measurements by solving the continuity equation along with Darcy Buckingham
23 equation either analytically or numerically. In the well-known JBN method

[27,28,30,31], one phase is injected into a core saturated with another phase and the overall pressure drop and outlet flux (effluent) are measured versus time. Again, the global measurements of pressure drop, phase flow rate, and saturation (through mass conservation) are taken, but now there are large differences between the global and local values both in time and space. The local values are obtained by assuming a 1-dimensional Buckley Leverett [25,26] type displacement, and from mathematical inversions both the relative permeabilities and the saturation at the outlet are calculated. This procedure is time dependent, allowing many different saturations and relative permeabilities to be obtained from one displacement.

There are other unsteady-state methods; all use slightly different ways of obtaining local values of the three key quantities. Hagoort et al. [29] extended the JBN method to oil displacement by air in a centrifuge. The expression for oil relative permeability at the outlet is simplified from the original JBN expression to the oil production rate due to high mobility of air and low capillary pressure. DiCarlo et al. [32,33] and Kianinejad et al. [34–38] conducted gravity drainage experiments and measured local saturations and fluxes in-situ using CT scanning. The local pressure gradient was taken to be the gravitational gradient, which was shown to be a good assumption for the center portion of the column.

The history matching method of obtaining unsteady-state relative permeability data has become a more popular method to obtain local values from measurements of overall pressure drop, effluent versus time, and, in some cases, in-situ saturation. The method presumes a relative permeability model (such as Corey model [39] and the LET model [40]) with prior fitting parameters and a capillary pressure curve (Leverett J

function [41]). Using the experimental boundary and initial conditions, the coupled Darcy Buckingham equation and continuity equation are numerically solved. This is compared to the experimental results, and the fitting parameters of the relative permeability model are adjusted to minimize the differences. Various simulators have been used/developed for this process, including (1) the commercial reservoir simulator ECLIPSE [42], (2) specialized 1D coreflood simulators, such as open-source SCORES [43–45], commercial software CYDAR^{TM} and Sendra, and (3) Shell proprietary 2D reservoir simulator (designed by Regtien et al. [46] and used by Berg et al. [47] in two-phase displacements). Two examples of this are Schembre et al. [42] and Berg et al. [47]. Both did two-phase displacement experiments and measured saturation profiles, pressure drop, and effluent versus time and obtained a posterior relative permeability model that best matched simulated results with measurements. Other authors have reported similar history matched relative permeability models [48–51]. However, the history matching method only obtains the fitting parameters of a particular function instead of actual relative permeability data.

In this paper, we develop a new unsteady-state method that is based on steady-state experiments – two phases are simultaneously injected into the core at a certain water fractional flow (which is the water flux divided by the total flux), local saturation is measured non-destructively using CT scanning, and pressure drops are measured through pressure taps to avoid the capillary end effect. Unlike the steady-state method, here we obtain local measurements at all times at each new water fractional flow, and not just at steady state. Local saturations are found from repeated CT scans, local pressure gradients are found from multiple pressure taps with continuously recorded pressure drops, and

1 local fluxes are found from saturation profile along the core versus time and a fractional
2 flow analysis. From these local measurements, from each water fractional flow step, we
3 obtain many points on a relative permeability curve rather than a single point obtained
4 using the steady-state method. Compared with standard unsteady-state methods (e.g. JBN
5 and history matching), we are able to avoid the capillary end effect by only using the
6 upstream four sections of the core.

7 To demonstrate this new method, it is used to obtain CO₂-brine relative
8 permeability. CO₂ relative permeability has been a key parameter in modeling multiphase
9 flow scenarios such as enhanced oil recovery using CO₂ as an agent [52–54], and CO₂
10 geological storage in deep saline aquifers [55–58]. The method is detailed, and then the
11 results are compared to steady-state data and to those obtained using another unsteady-
12 state scheme.

13

14 **2. Obtaining Local Flux from Saturation Data**

15 Any unsteady-state method needs to obtain the local flux. This section gives two
16 different approaches that use the measured saturation profiles with time to find the local
17 flux during the unsteady portion of the flow, which is the part that occurs during each
18 fractional flow step before steady state is reached. Note, each drainage experiment
19 consists of a few fractional flow steps, during which two phase fluids are injected at a
20 fixed fractional flow.

21 Firstly, we introduce a new procedure that uses the fractional flow theory
22 (Buckley-Leverett solution) [25] and integrates the water fractional flow derivative on the

spatial saturation difference. Secondly, we use a previously published solution that integrates the temporal saturation difference on space [32–35].

Both approaches have the same initial condition and boundary condition. The initial condition is that the core starts with the uniform saturation achieved at the earlier fractional flow step:

$$S_w(x, t = 0) = S_w(x, \text{steady state of previous fractional flow step}) \quad (2)$$

In Equation 2, S_w is the water saturation, t is the time from the onset of a new imposed fractional flow, and x is the distance from the inlet of core (unit: cm). The boundary condition is the application of a new fractional flow:

$$f_w(x = 0, t > 0) = f_w(\text{imposed at the pumps}, t > 0) \quad (3)$$

In Equation 3, f_w is water fractional flow. The new method that we develop herein uses fractional flow theory [25,26] to obtain the local water fractional flow as a function of space and time. By assuming incompressible fluids and no diffusive transport, the continuity equation is:

$$\phi \frac{\partial S_w}{\partial t} + u \frac{\partial f_w}{\partial x} = 0 \quad (4)$$

In Equation 4, ϕ is porosity and u is the total Darcy velocity of the two phases (unit: cm/min). If gravitational and capillary forces are neglected then the fractional flow

1 is only a function of the saturation. Thus, Equation 4 is hyperbolic, and can be solved by
 2 introducing a new variable using the similarity transformation: $\xi = \frac{x}{t}$. Then Equation 4
 3 becomes the following equation:

4

$$5 \quad -\frac{\phi}{u} \cdot \xi \cdot \frac{dS_w}{d\xi} + \frac{df_w}{dS_w} \cdot \frac{dS_w}{d\xi} = 0 \quad (5)$$

6

7 Equation 5 has two solutions: a) a trivial one is $dS_w/d\xi = 0$, this occurs ahead of
 8 the front where the change in boundary condition has not yet propagated, and b)

9

$$10 \quad \frac{df_w}{dS_w} = \frac{\phi x}{ut} \quad (6)$$

11

12 Since the variables on the right hand side are easily found, Equation 6 gives the
 13 derivative of water fractional flow with respect to water saturation at any position and
 14 time. To obtain the local fractional flow at a particular injection time, this derivative is
 15 numerically integrated with respect to the spatial changes in saturation at the same time;
 16 this integration is done by a finite difference approximation shown in Equation 7. Note
 17 that when applying the boundary condition Equation 3, the most upstream slice of the
 18 core measured by CT is set to have f_w imposed by the pumps.

19

$$f_w(x,t) = f_w(0,t) + \int_{S_w(0,t)}^{S_w(x,t)} \frac{df_w}{dS_w} dS_w$$

$$\Rightarrow f(x=i,t) = f_w(x=1,t) + \sum_{x_0=2}^{x_0=i} \left\{ \frac{df_w}{dS_w} \bigg|_{x_0-1,t} \cdot [S_w(x_0,t) - S_w(x_0-1,t)] \right\} \quad (7)$$

The second method of calculating local phase fluxes is based on mass conservation, and has been previously published and used by DiCarlo et al. [32,33] and Kianinejad et al. [34,35] (hereafter called the previous method). In their gravity drainage experiments, they calculated the phase fractional flow by integrating the temporal phase saturation difference on space (the saturation difference between two consecutive scanning times at any position), and the integration was approximated with finite difference shown by Equation 8.

$$f_w(x,t) = f_w(0,t) + \frac{\phi}{u\Delta t} \int_0^x (S_w(x_0,t-\Delta t) - S_w(x_0,t)) dx_0$$

$$\Rightarrow f_w(x=i,t) = f_w(x=0,t) + \frac{\phi \cdot \Delta x_0}{u\Delta t} \sum_{x_0=1}^{x_0=i} (S_w(x_0,t-\Delta t) - S_w(x_0,t)) \quad (8)$$

The local flux obtained above along with the measured pressure drop of each section of the core can be used to find the relative permeabilities with the simple inversion of Equation 1, and the associated water saturation was measured locally.

3. Materials and Methods

Details of the experimental set-up are published elsewhere (manuscript A [24]); here we give an overview with a concentration on the particular parts that are needed for the unsteady-state technique.

3.1 Rock and Fluids

The rock sample is a 60.8-cm long and 7.14-cm diameter cylindrical Berea sandstone core (the same core used in [24]). It has a uniform porosity of 17.6% along the axial direction. The experiments were conducted at 20 °C and 1500 psi with 2 wt% NaCl brine and CO₂. To avoid mass transfer during the experiment, brine and CO₂ were equilibrated with each other and stored separately in two vertical piston-accumulators; details on making sure phase equilibration are in manuscript A [24]. The viscosities of non-equilibrated brine, equilibrated brine and equilibrated CO₂ are 1.040 cp [59], 1.081 cp [60,61], and 0.087 cp [62,63].

3.2 Core Flood Experiment Setup

Figure 1 shows the experiment setup. The three-layer-wrapped (heat-shrink tubing, aluminum foil, heat-shrink tubing) core sample was placed in a vertical aluminum core holder with a confining pressure of 2000 psi. Four pressure taps on the core divided it, from upstream to downstream, into five individual sections: section 1, section 2, section 3, section 4 and section 5. The wrapping was drilled through at taps located at 15.24, 25.40, 35.56, and 45.72 cm away from the inlet. The overall pressure drop (ΔP_{io}) and the pressure drops of five sections (from upstream to downstream named as ΔP_{i1} , ΔP_{12} , ΔP_{23} , ΔP_{34} , and ΔP_{4o}) were continuously monitored using six differential pressure

transducers. During experiments, the piston Accumulator C (vertically placed, Figure 1) received the two-phase effluents, which then pushed the piston to discharge pure water on the bottom through a backpressure regulator (BPR) set at 1500 psi. The use of the outlet accumulator prevented CO₂ decompression through the BPR and minimized pressure fluctuations during the experiment.

Figure 1. Schematic setup of core flooding apparatus in this study.

3.3 Determination of Saturation with X-ray Computed Tomography

The new method is predicated on obtaining accurate spatial saturations versus time. To achieve that, the core holder was vertically mounted onto a vertical positioning system (VPS) that can move up and down through a medical X-ray CT scanner. During the two-phase flow, when the total flow rate was 2 ml/min, 60 slices of CT images were obtained (1-cm thickness) along the core axis; the scanning took 6 minutes and 9

seconds. The imaging frequency was every 0.05 PV and the index between consecutive slice centers was 1 cm. When the total flow rate was 4 ml/min, 30 slices of CT images were obtained (1-cm thickness) along the core axis; the scanning took 3 minutes and 5 seconds. The imaging frequency was still every 0.05 PV but the index between consecutive slice centers was 2 cm. When the total flow rate was 8 ml/min and 16 ml/min, 30 slices of CT images were obtained along the core axis, with a scanning frequency of 0.10 PV and 0.20 PV, respectively. Saturation data are missing at the pressure taps where X-rays were blocked by the stainless steel fitting. The accuracy/error of CT-measured slice-wise water saturation is 0.4% (absolute saturation).

Although the saturation measurement along the core axis took a certain amount of time, it is assumed that the saturation profile was taken as a snapshot when using it for calculating local water fractional flows. As a consequence, this approximation brings an uncertainty to the calculation of the water fractional flow, which is within 3% for the new method and within 7% for the previous method; and this uncertainty propagates into the calculation of the relative permeability. Details on the uncertainty estimate are in Appendix A.

3.4 CO₂-Brine Two-Phase Primary Drainage

To determine two-phase relative permeabilities, we conducted five primary drainage experiments by simultaneously injecting equilibrated-brine (hereafter referred as brine) and equilibrated-CO₂ (hereafter referred as CO₂) into a brine-saturated core. Each experimental step consisted of applying a water fractional flow (f_w) that was lower than the previous step, measuring the saturation versus space and time and the local pressure

drops during the unsteady-state flow that occurred. Once steady state was reached for this step, another lower f_w was applied, and the method was repeated. Five primary drainage experiments were performed and their names and specific flow rates are listed in Table 1.

Table 1. Injection rates during primary drainage experiments

	qbrine:qCO ₂	100:0	50:50	10:90	1:99	0.1:99.9	0:100
Exp5	qbrine(ml/min)	2.000	1.000	0.200	0.040	0.008	0
	qCO ₂ (ml/min)	0	1.000	1.800	3.960	7.992	8.000
Exp6	qbrine(ml/min)	4.000	2.000	0.400	0.080	0.016	0
	qCO ₂ (ml/min)	0	2.000	3.600	7.920	15.984	16.000
Exp7d	qbrine(ml/min)	2.000	1.000	0.200	0.020	0.002	0
	qCO ₂ (ml/min)	0	1.000	1.800	1.980	1.998	2.000
Exp8d	qbrine(ml/min)						0
	qCO ₂ (ml/min)			NA			2.000
Exp9d	qbrine(ml/min)						0
	qCO ₂ (ml/min)			NA			4.000

4. Results

4.1 Single-Phase Brine Relative Permeability

Five primary drainage experiments (Exp5, Exp6, Exp7d, Exp8d, and Exp9d) were conducted. Before the beginning of each experiment, the single-phase absolute permeability to non-equilibrated brine was measured and reported in [24]. Table 2 shows that the brine permeability of the entire core and each individual section changed within 10% throughout all the five primary drainage experiments.

Table 2. Single phase brine permeability (K)

		whole section	1section	2section	3section	4section	5section
	length(cm)	60.80	15.24	10.16	10.16	10.16	15.08
k _w (md)	Exp5	115.98	95.17	128.27	138.78	143.12	110.87
	Exp6	104.76	80.45	119.98	126.66	126.91	104.50
	Exp7d	114.36	91.82	128.50	135.03	141.15	110.02

Exp8d	117.63	101.55	134.22	138.24	135.15	116.04
Exp9d	120.56	101.67	135.86	147.46	132.13	113.66
avg(mD)	114.66	94.13	129.37	137.24	135.69	111.01
std/avg (%)	5.22	9.28	4.82	5.46	4.87	3.92

4.2 Saturation and Pressure Drop Data versus Space and Time

We take the fractional flow step of 0.1 during Exp5 as an example to show how the relative permeability is calculated with the new unsteady state method. Figure 2a shows the saturation profile development versus time at the fractional flow step of $f_w=0.1$ during Exp5. The initial condition of the core was after a steady-state flood with $f_w=0.5$. With the application of the lower water fractional flow of $f_w = 0.1$, the water saturation decreased from 0.90 to about 0.75 as the front moved through the core from left to right.

Figure 2b shows the measured pressure drops of the upstream four sections of the core versus total injected volume at the fractional flow step of $f_w=0.1$ in Exp5. Note that the pressure drop in section 1 was higher than the other sections, as it is longer (see Table 2). Section 5 is also longer (see Table 2) but not shown, as it is not used in calculating relative permeability. Soon after the early transient time (0-0.05 PV), the pressure drops of the core's upstream four sections monotonically decreased with time until reaching stable values. By comparing Figure 2a and Figure 2b, we find the evolutions of saturation and pressure drop of every section of the core were synchronized. For instance, for the entrance section of the core (section 1), between 0.05 PV and 0.20 PV, both the water saturation and the pressure drop decreased with time; since 0.25 PV, they both approximately reached steady-state values.

Figure 2. During the fractional flow step of $f_w=0.1$ in Exp5: (a) water saturation profiles recorded at continuous injection times; (b) the pressure drops of the upstream four sections of the core measured versus time.

4.3 Local Phase Flux and CO₂-Brine Relative Permeability

Figure 3a and Figure 3b show the calculated local water fractional flow versus space and time using the new method and the previous method for the fractional flow step of $f_w=0.1$ in Exp5. Again, the new method integrates the water fractional flow derivative on the spatial saturation difference based on the fractional flow theory; while the previous method integrates the temporal saturation difference on space based on mass conservation (details are given in Equations 2-8). In general, both figures show the calculated water fractional flow started at 0.1 at the inlet where the flow was controlled; progressing into the column the water fractional flow rose quickly in space to the fractional flow of the previous step (0.5). At later times, 0.55-0.60 PV, the imposed fractional flow of 0.1 was seen throughout the column.

1

2

3 **Figure 3. At the fractional flow step of $f_w=0.1$ during Exp5: the local water**4 **fractional flows versus space and time calculated with (a) the new method and (b)**5 **the previous method; the unsteady-state (c) brine and (d) CO₂ relative permeability**6 **data obtained with the new method and the previous method compared with the**7 **steady-state data at this step and the previous step ($f_w=0.5$).**

8

1 Comparing the two methods, the differences between the slice-wise water
2 fractional flows calculated with the new method and the previous method are: (1) the
3 water fractional flow profile at every injection time calculated with the new method is
4 less smooth than that calculated with the previous one; (2) the water fractional flow
5 profile calculated with the new method monotonically decreases with increasing time,
6 while the previous method has some water fractional flows that increases with increasing
7 time (see 0.20 PV and 0.25 PV).

8 The first difference is because the new method integrates on the spatial saturation
9 difference that has more scatter due to saturation heterogeneity. A special case is that at
10 the early transient times (0.05 PV & 0.10 PV), water saturation slightly decreases towards
11 the outlet, which makes water fractional flow decreases towards the outlet. However, this
12 case is not seen at later times. Another special case at the early transient times (0.05 PV
13 & 0.10 PV) is that both Figures 3a and 3b show that the water fractional flow exceeds the
14 steady-state value of the previous step (0.5) near the outlet. Again, this is because at early
15 times, the stronger saturation heterogeneity makes the error in the f_w calculation larger,
16 which is not seen at later times. The second difference is because: (1) in the new method
17 the integrand, $df_w/dS_w = \phi x/ut$, monotonically decreases with time and hence the integral at
18 the same position in space also monotonically decreases with time; (2) in the previous
19 method the integrand, $\phi \Delta S_w/u \Delta t$, does not monotonically decrease with time, but
20 depends on the measured temporal saturation difference. For instance, in the downstream
21 portion of the core, the temporal water saturation difference between 0.20 and 0.25 PV is
22 greater than that between 0.15 and 0.20 PV; hence the water fractional flow at 0.25 PV is
23 higher than that at 0.20 PV when using the previous method.

1 Since the local pressure drop is measured over each section, the f_w is averaged
2 over each section. This causes some of the scatter in f_w obtained by the new method to be
3 smoothed out. Comparing the section-wise f_w of these two methods, it is found that they
4 are consistent with an average relative difference (defined as the absolute difference in
5 water fractional flow between the two methods divided by the water fractional flow
6 obtained from the new method) of 20%.

7 Using the section-wise fractional flow of both methods and the pressure drop of
8 each of the upstream four sections, the unsteady-state CO₂ and brine relative permeability
9 can be directly calculated with the simple inversion of Equation 1. In Equation 1, Q_{CO_2}
10 $=Q_t*(1-f_w)$ and $Q_{brine}=Q_t*f_w$ where Q_t are the total flow rates of CO₂ and brine (Table 1);
11 μ_{CO_2} and μ_{brine} are the viscosity of equilibrated CO₂ and brine, which are 0.087 cp and
12 1.081 cp; L and K are the length and single phase brine permeability of the entire core or
13 a certain section, (Table 2); A is cross section area of the core (40.08 cm²). The pressure
14 drop ΔP was measured across each of the upstream four sections at each injection time; it
15 has been previously shown that this is the pressure drop of both phases ΔP^i in the center
16 of the core and that of the CO₂ phase ΔP^{CO_2} in the entrance section (section 1) [24]. We
17 exclude the exit section for the relative permeability calculation because the exit section
18 is dominated by the capillary end effect. Hence, this new method experimentally avoids
19 the capillary end effect that plagues relative permeability determinations, particularly for
20 low viscosity fluids such as supercritical and gas phases.

21 Figures 3c and 3d show the unsteady-state brine and CO₂ relative permeability
22 data obtained during the unsteady-state flow portion of $f_w=0.1$ step with both methods
23 along with the steady state data. For both methods, there are dozens of unsteady-state

relative permeability data points (30 for brine and 40 for CO₂) that fill the saturation gaps between the steady state data at consecutive steps.

By comparing the relative permeabilities obtained with the two methods, we find: (1) in terms of brine relative permeability, the average relative difference between these two methods is 20%, which reflects the 20% average relative difference between the section-wise water fractional flows calculated with these two methods; (2) in terms of CO₂ relative permeability, the average relative difference between these two methods is 6%. This is because at this step, local unsteady-state f_w is around 0.1; hence, the average relative difference in CO₂ fractional flow between these two methods is one order of magnitude less than the average relative difference in water fractional flow.

Using both the new and the previous methods, we calculate the local water fractional flows, and then obtain the unsteady-state CO₂ and brine relative permeability data from Exp5, Exp6 and Exp7d (5-step primary drainage experiment) at water fractional flow steps of $f_w = 0.5, 0.1$ and 0.01 , and from Exp8d and Exp9d (1-step primary drainage experiment at $f_w = 0$). Note that at fractional flow steps with low imposed f_w (0.001 and 0) during the 5-step primary drainage experiments, data are not obtained since the measured saturation changes at these steps were too small to apply the new method of calculating unsteady-state local water fractional flows. There are other instances of pressure tap failures, and hence some sections do not produce data. Data are not obtained before breakthrough because of saturation shock; details of which are discussed later in section 5.3. Still, through 5 experiments we have determined 388 CO₂ relative permeability data points and 266 brine relative permeability data points with both the new method and the previous method.

Due to the measurement error, the uncertainty in water saturation is 0.4 % (absolute saturation). For both the new and the previous methods, the uncertainty in the CO₂ relative permeability obtained in the center three sections of the core is within 3%, and the uncertainty in the CO₂ relative permeability obtained in the entrance section of the core is within 6%. The uncertainty in the brine relative permeability obtained in the center three sections of the core is within 4% for the new method, and 8% for the previous method. Details on the uncertainties of the unsteady-state relative permeability data are in Appendix A. All the unsteady-state CO₂-brine relative permeability data are tabulated in Appendix B.

5. Discussion

5.1 Comparison between Steady-State and Unsteady-State Relative Permeability

Figure 4 compares (a) the steady state and (b) the unsteady-state (using the new method) CO₂ and brine relative permeabilities of all five drainage experiments, of which Figure 4a is reproduced from Figure 5a of [24] with permission. The dashed line (Equation 9) and the solid line (Equation 10) in Figure 4a and Figure 4b are the Corey-type models that fit the steady-state (Figure 4a) and the unsteady-state (Figure 4b) CO₂ and brine relative permeabilities, respectively.

$$k_{rg} = \left(\frac{1 - S_w}{1 - S_{wr}} \right)^{n_g} \quad (9)$$

$$k_{rw} = \left(\frac{S_w - S_{wr}}{1 - S_{wr}} \right)^{n_w} \quad (10)$$

1

2

3 **Figure 4. (a) Steady-state and (b) unsteady-state (using the new method) CO₂ and**
 4 **brine relative permeability data determined in all five primary drainage**
 5 **experiments. The dashed line and the solid line are the Corey-type models that fit to**
 6 **the steady-state and the unsteady-state CO₂ and brine relative permeabilities,**
 7 **respectively.**

8

9 In Equation 9 and Equation 10, only the exponents, n_g and n_w , are the fitting
 10 parameters. The irreducible water saturation (S_{wr}) is the average measured S_{wr} of the five
 11 primary drainage experiments, given by $S_{wr} = (26 \pm 2) \%$. Using the least squares method,
 12 the exponents that fit to the steady-state relative permeability data are: $n_g = 1.8 \pm 0.1$ and
 13 $n_w = 5.2 \pm 0.3$ (given in manuscript A [24]); while the exponents that fit to the unsteady-
 14 state data are: $n_g = 1.78 \pm 0.01$ and $n_w = 4.64 \pm 0.04$. Because of the larger amount of data
 15 points in the new unsteady state method compared with the steady state method, the

1 uncertainties in the fitting exponents, n_g and n_w , are decreased by up to 90%, leading to
2 better constrained relative permeability models.

3 In general, the steady state and the unsteady state CO₂ and brine relative
4 permeabilities are consistent. In particular, the Corey-type exponents, n_g and n_w , in both
5 methods differ by 1% and 11%, respectively. The relative large difference in n_w reflects
6 the uncertainty in the calculated local water fractional flow with the new method. The
7 smaller relative difference in n_g compared to n_w is due to the local CO₂ fractional flow (f_g)
8 being close to 1 in the primary drainage. Hence, the uncertainty in f_g is one order (or two
9 orders) of magnitude less than the uncertainty in f_w .

11 5.2 Comparison between the New Method and the Previous Method

12 The new and the previous methods use slightly different ways to obtain local
13 water fractional flows: one integrates the water fractional flow derivative on the spatial
14 saturation difference, while the latter integrates the temporal saturation difference on
15 space [32–35]. If the actual flow exactly follows the fractional flow theory, then these
16 two methods should give identical results. However, this is not the case as Figure 3c
17 clearly shows that the brine relative permeabilities calculated by these two methods are
18 different. It can be seen that, the new method produces more consistent results as the
19 brine relative permeabilities of the new method are less scattered.

20 Figure 5 compares the brine and CO₂ relative permeability data calculated with
21 the new method and the previous method for all five experiments. Figure 5a shows brine
22 relative permeability data obtained from the previous method are roughly between 30%
23 more and 20% less of those obtained from the new method (average relative difference is

25%). Figure 5b shows CO₂ relative permeability data obtained from the previous method are within 2% of those obtained from the new method (average relative difference is 2%). As we have mentioned in the results section, the relative difference in the brine relative permeability data between the two methods only results from the same amount of relative difference in the section-wise water fractional flows between the two methods. And since CO₂ fractional flow is order/orders of magnitude higher than water fractional flow in the five primary drainage experiments, this 25% relative difference in water fractional flow becomes order/orders of magnitude less for CO₂ fractional flow.

Figure 5. Comparison of (a) brine relative permeability and (b) CO₂ relative permeability determined with this new method and the previous method. The black, green and blue solid lines are $y=x$, $y=1.3x$ and $y=0.8x$, respectively.

Again, the difference in water fractional flow calculated by the two methods is because the actual flow did not exactly follow the fractional flow theory. This could be

because the large viscosity ratio of the CO₂-brine system makes capillary forces not negligible. By choosing a different fluid pair with a smaller viscosity ratio, the difference between f_w obtained by the two methods may turn out smaller.

5.3 Limitations of the method

There are two limitations in this new unsteady-state method. First, in the Buckley-Leverett solution (Equation 6), a shock has to be introduced to avoid two values of the invading phase (using the subscript i) saturation at each position x (x is linear with df_i/dS_i according to Equation 6). Hence, relative permeability cannot be obtained below this shock saturation of invading phase. This is a general limitation for all 1D two-phase unsteady-state methods that ignore or avoid (in our case) the capillary term.

In our case, the invading phase is CO₂. Figure 6a plots both the CO₂ fractional flow (f_g) and the CO₂ fractional flow derivative (df_g/dS_g) versus CO₂ saturation (S_g).

$$f_g = \frac{1}{1 + \frac{k_{rw}\mu_g}{k_{rg}\mu_w}} \quad (11)$$

Equation 11 (obtained from Equation 1) is used to calculate f_g , in which k_{rg} and k_{rw} use Equations 9 and 10 and the fitting parameters for all unsteady-state data ($S_{wr}=0.26$, $n_g=1.78$, and $n_w=4.64$). As mentioned above, a shock has to be added in the df_g/dS_g - S_g curve (df_g/dS_g is linear with x) to avoid double values of S_g at each position x . And the invading phase saturation at the peak of the df_g/dS_g - S_g curve ($S_g=10\%$) is a lower bound of the shock saturation. This is consistent with our experimental observations of

1 water saturation in Figure 6b during the first primary drainage step of $f_w=0.5$. Hence,
 2 unsteady-state relative permeability cannot be obtained when water saturation is higher
 3 than roughly 90%. This is seen in Figures 4b where no relative permeability data is
 4 reported in this saturation range, because the new method does not calculate the relative
 5 permeability before breakthrough.
 6

7
 8 **Figure 6. (a) CO₂ fractional flow and CO₂ fractional flow derivative versus CO₂**
 9 **saturation curves (based on Equations 9, 10 and 11 and the fitting parameters in**

1 this manuscript), (b) water saturation profile evolution during the first drainage
2 step of $f_w=0.5$ in Exp5, (c) CO₂ fractional flow derivative versus CO₂ saturation
3 curves at different viscosity ratios of the invading phase to the defending phase.

4
5 Second, another limitation of the new method is that when conducting the 5-step
6 primary drainage experiments, the unsteady-state relative permeability cannot be
7 obtained at very low f_w steps (or very low water saturations). The reason is at the final
8 steps when the imposed f_w are very low, the measured water saturation changes within the
9 experimental duration were too small to calculate local unsteady-state water fractional
10 flows.

11 Although the first limitation happens for all 1D two-phase Buckley-Leverett
12 displacements that ignore/avoid the capillary term, the extent of the limitation is
13 determined by the viscosity ratio of the invading phase to the defending phase. In Figure
14 6c, by using Equations 9, 10 (the same fitting parameters) and 11, we plot the invading
15 phase fractional flow derivative (df_i/dS_i) versus the invading phase saturation (S_i) at five
16 different viscosity ratios of the invading phase to the defending phase (μ_i/μ_d), namely,
17 0.01, 0.1, 1, 10 and 100. It can be seen that, as the invading phase becomes more viscous,
18 the invading phase saturation at the peak (a lower bound of shock saturation) increases
19 from 3% to 52%; in turn, this limits the calculation of the relative permeability to
20 narrower saturation ranges. In our case, the CO₂-brine primary drainage experiments
21 have an invading phase to defending phase viscosity ratio of 0.08. Hence, this limitation
22 is small for the data shown here. Thus this method is most appropriate for displacements

of high viscosity fluids by low viscosity fluids (such as gases and supercritical phases), as the limitations are minor, but with the advantage of avoiding the capillary end effect.

6. Conclusions

This study presents a new unsteady-state method of determining the two-phase relative permeability by using the local pressure drop, local saturation, and local phase fluxes during the unsteady-state flow portion of conventional steady-state primary drainage experiments. The local pressure drop and saturation are experimentally measured, while the local phase flux is obtained by integrating the water fractional flow derivative on the spatial saturation difference based on the fractional flow theory [25,26]. In particular, we use the new method to determine the CO₂-brine relative permeability from five primary drainage experiments in a Berea sandstone core. We find that the unsteady-state and steady-state CO₂-brine relative permeability data are consistent. Compared with the steady state method, the large amount of relative permeability data obtained by the new unsteady-state method decrease the uncertainties of the exponents in the Corey-type fits by up to 90%. We also use a previous approach by DiCarlo et al. [32,33] to calculate local phase fluxes based on mass conservation to integrate the temporal saturation difference on space. The comparison between the new method and the previous method in terms of section-wise phase fluxes and relative permeabilities proves that the new method is rigorous in finding the local phase fluxes and unsteady-state relative permeabilities. Also, this new method is especially useful but not limited to displacements of a high viscosity fluid by a low viscosity fluid (such as gas or supercritical phase).

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8. Appendix

A. The Uncertainty of Water Fractional Flow and Relative Permeability Calculated with the New Method and the Previous Method

To estimate the uncertainty in water fractional flow due to neglecting the time it takes to measure local saturation, we take Exp5 as an example and correct the time offsets of the saturation profiles at the water fractional steps of $f_w=0.5$, 0.1 and 0.01. When the total flow rate was 2 ml/min, 60 X-ray CT scans were taken along the core axial direction to measure slice-wise saturations, which took 6 minute and 9 seconds. Except for the saturations at $x=30$ cm, saturation scanned at any other place of the core had a time offset with the planned time. The scanning frequency at total flow rate of 2 ml/min was every 0.05 PV. The first order correction of saturation profile is to assume that saturations at each position of the core were changing linearly with time between consecutive scanning times. Based on this assumption, all saturation profiles were

corrected to be exactly at the planned scanning times. Using the corrected saturation profiles, the local water fractional flows (f_w) were calculated again using both the new method and the previous method.

We find the average relative difference in f_w between using the actual saturation profiles and using the corrected saturation profiles is 3% for the new method and 7% for the previous method based on 160 f_w data points. Hence, due to neglecting the saturation measurement time, the water fractional flow has a 3% relative error of its own value for the new method, and a 7% relative error of its own value for the previous method.

In manuscript A [24], we have shown that due to the measured pressure drop fluctuations and the non-uniform saturation in the entrance section of the core, steady-state CO₂ and brine relative permeability determined in the center three sections of the core had a 3% relative error of its own value and steady-state CO₂ relative permeability determined in the entrance section of the core had a 6% relative error of its own value. For the unsteady-state relative permeability we determined herein, the extra error source is from the local fractional flows of water and CO₂. As for the uncertainty in the local fractional flow of CO₂ (f_g), since f_g was close to 1 in the primary drainage, its uncertainty is negligible compared with the uncertainty due to pressure fluctuations.

Apply the uncertainty propagation to the two-phase Darcy Buckingham equation, the uncertainties in unsteady-state relative permeability data were estimated using Equation A1. First, for both the new method and the previous method, the uncertainty of CO₂ permeability determined in the core's center sections is 3% of its own value, and the uncertainty of CO₂ relative permeability determined in the core's entrance section is 6% of its own value. Second, the uncertainty of brine relative permeability determined in the

1 core's center sections is 4% of its own value for the new method, and 8% of its own
2 value for the previous method.

3

$$4 \quad \frac{\sigma(k_{ri})}{k_{ri}} = \frac{\sqrt{\left(\frac{\partial k_{ri}}{\partial f_i}\right)^2 \sigma(f_i)^2 + \left(\frac{\partial k_{ri}}{\partial \Delta P}\right)^2 \sigma(\Delta P)^2}}{k_{ri}} = \sqrt{\left(\frac{\sigma(f_i)}{f_i}\right)^2 + \left(\frac{\sigma(\Delta P)}{\Delta P}\right)^2} \quad (A1)$$

5

6 B. Unsteady-State Relative Permeability Data Obtained with the New Method and the
7 Previous Method

8

9 **Table B1. CO₂-Brine Relative Permeability Calculated with the New Method in Exp5**

fw	Time	sec1			sec2			sec3			sec4		
	(PV)	Sw	krw	Sw	krw	krw	Sw	krw	krw	Sw	krw	krw	
0.5	0.30	0.887	0.0507	0.898	0.5014	0.0390	0.903	0.5127	0.0385	0.910	0.4739	0.0341	
	0.35	0.890	0.0509	0.904	0.4887	0.0379	0.907	0.5561	0.0421	0.913	0.4761	0.0351	
	0.40	0.892	0.0509	0.900	0.4852	0.0383	0.904	0.5479	0.0424	0.907	0.4674	0.0360	
0.1	0.05	0.810	0.1176	0.875	0.4850	0.0548	0.882	0.5191	0.0448	0.884	0.4849	0.0398	
	0.10	0.764	0.1545	0.845	0.3982	0.0751	0.862	0.4462	0.0612	0.874	0.4214	0.0455	
	0.15	0.753	0.1793	0.823	0.3122	0.0923	0.851	0.3650	0.0716	0.869	0.3781	0.0565	
	0.20	0.748	0.1903	0.803	0.2662	0.1054	0.826	0.2933	0.0861	0.855	0.3550	0.0694	
	0.25	0.739	0.2027	0.783	0.2772	0.1332	0.799	0.2655	0.1031	0.831	0.3365	0.0859	
	0.30	0.739	0.2039	0.779	0.2456	0.1291	0.792	0.2375	0.1073	0.820	0.3076	0.0974	
	0.40	0.738	0.1874	0.766	0.2534	0.1546	0.768	0.2083	0.1236	0.788	0.2398	0.1123	
	0.45	0.733	0.1962	0.765	0.2301	0.1405	0.766	0.2068	0.1233	0.788	0.2369	0.1130	
	0.55	0.738	0.1875	0.764	0.2188	0.1417	0.757	0.1930	0.1300	0.772	0.2102	0.1228	
0.60	0.730	0.1859	0.759	0.2451	0.1577	0.756	0.2002	0.1309	0.772	0.2276	0.1291		
0.01	0.30	0.644	0.2989	0.726	0.1755	0.1872	0.736	0.1802	0.1502				
	0.40	0.632	0.3026	0.711	0.1584	0.2213	0.726	0.1498	0.1589				
	0.45	0.627	0.3153	0.706	0.1440	0.2230	0.721	0.1392	0.1626				
	0.55	0.622	0.3305	0.698	0.1216	0.2285	0.717	0.1266	0.1721				
	0.60	0.619	0.3593	0.695	0.1096	0.2255	0.711	0.1170	0.1812				
	0.65	0.618	0.3328	0.689	0.1151	0.2602	0.705	0.1092	0.1851				
	0.75	0.613	0.3431	0.682	0.1040	0.2698	0.698	0.0997	0.1936				
	0.90	0.600	0.3581	0.662	0.0988	0.3057	0.677	0.0839	0.1989				
	1.00	0.595	0.3890	0.657	0.0817	0.2703	0.672	0.0840	0.2146				
	1.10	0.594	0.3954	0.657	0.0806	0.2795	0.670	0.0800	0.2219				
	1.15	0.595	0.3598	0.652	0.0845	0.3205	0.666	0.0765	0.2263				
	1.25	0.592	0.3689	0.650	0.0816	0.3161	0.664	0.0740	0.2301				
	1.40	0.592	0.3678	0.646	0.0786	0.3326	0.658	0.0699	0.2419				
	1.50	0.590	0.3670	0.640	0.0797	0.3591	0.651	0.0650	0.2460				

10

1

Table B2. CO₂-Brine Relative Permeability Calculated with the Previous Method in Exp5

fw	Time (PV)	sec1		sec2			sec3			sec4		
		Sw	krw	Sw	krw	krw	Sw	krw	krw	Sw	krw	krw
0.5	0.30	0.887	0.0511	0.898	0.4926	0.0397	0.903	0.5098	0.0388	0.910	0.5219	0.0302
	0.35	0.890	0.0517	0.904	0.4568	0.0404	0.907	0.4942	0.0471	0.913	0.4050	0.0409
	0.40	0.892	0.0522	0.900	0.4779	0.0389	0.904	0.5485	0.0424	0.907	0.4815	0.0349
0.1	0.05	0.810	0.1002	0.875	0.6098	0.0447	0.882	0.6304	0.0359	0.884	0.6205	0.0289
	0.10	0.764	0.1467	0.845	0.5086	0.0663	0.862	0.5513	0.0527	0.874	0.4978	0.0394
	0.15	0.753	0.1839	0.823	0.2825	0.0947	0.851	0.3131	0.0757	0.869	0.2950	0.0632
	0.20	0.748	0.1945	0.803	0.2543	0.1064	0.826	0.3208	0.0839	0.855	0.3641	0.0687
	0.25	0.739	0.2021	0.783	0.3473	0.1276	0.799	0.4041	0.0920	0.831	0.4886	0.0737
	0.30	0.739	0.2075	0.779	0.1907	0.1336	0.792	0.1900	0.1111	0.820	0.2298	0.1037
	0.40	0.738	0.1895	0.766	0.2440	0.1554	0.768	0.2511	0.1202	0.788	0.3137	0.1064
	0.45	0.733	0.1951	0.765	0.2534	0.1387	0.766	0.2306	0.1214	0.788	0.2246	0.1140
	0.55	0.738	0.1901	0.764	0.1745	0.1453	0.757	0.1764	0.1313	0.772	0.2061	0.1232
	0.60	0.730	0.1830	0.759	0.3233	0.1514	0.756	0.2837	0.1242	0.772	0.2877	0.1243
0.01	0.30	0.644	0.2985	0.726	0.1698	0.1876	0.736	0.1590	0.1520			
	0.40	0.632	0.3042	0.711	0.1564	0.2214	0.726	0.1524	0.1587			
	0.45	0.627	0.3157	0.706	0.1305	0.2241	0.721	0.1359	0.1629			
	0.55	0.622	0.3328	0.698	0.0969	0.2305	0.717	0.0957	0.1746			
	0.60	0.619	0.3612	0.695	0.0746	0.2283	0.711	0.0957	0.1829			
	0.65	0.618	0.3357	0.689	0.1032	0.2612	0.705	0.1244	0.1839			
	0.75	0.613	0.3436	0.682	0.0982	0.2703	0.698	0.1033	0.1933			
	0.90	0.600	0.3567	0.662	0.1758	0.2996	0.677	0.1722	0.1918			
	1.00	0.595	0.3888	0.657	0.0986	0.2690	0.672	0.1015	0.2132			
	1.10	0.594	0.3960	0.657	0.0451	0.2824	0.670	0.0393	0.2252			
	1.15	0.595	0.3647	0.652	0.0485	0.3234	0.666	0.0838	0.2257			
	1.25	0.592	0.3678	0.650	0.0779	0.3164	0.664	0.0648	0.2308			
	1.40	0.592	0.3698	0.646	0.0593	0.3342	0.658	0.0606	0.2427			
	1.50	0.590	0.3667	0.640	0.0928	0.3581	0.651	0.0986	0.2434			

2

3

Table B3. CO₂-Brine Relative Permeability Calculated with the New Method in Exp6

fw	Time (PV)	sec1		sec2			sec3			sec4		
		Sw	krw	Sw	krw	krw	Sw	krw	krw	Sw	krw	krw
0.5	0.32	0.879	0.0497	0.892	0.5877	0.0456	0.901	0.3532	0.0260	0.911	0.5644	0.0389
0.1	0.10	0.754	0.1578	0.825	0.3635	0.0750	0.854	0.2486	0.0307	0.870	0.5727	0.0508
	0.20	0.730	0.1906	0.780	0.2882	0.1181	0.806	0.1745	0.0511	0.833	0.3489	0.0694
	0.32	0.728	0.2006	0.766	0.2528	0.1367	0.782	0.1559	0.0709	0.808	0.2342	0.0774
	0.41	0.729	0.2025	0.763	0.2456	0.1443	0.773	0.1465	0.0793	0.800	0.1982	0.0798
	0.50	0.732	0.2026	0.763	0.2372	0.1463	0.768	0.1429	0.0851	0.787	0.1676	0.0831
0.01	1.10	0.586	0.4277	0.665	0.0997	0.2792	0.703	0.1068	0.1891			
	1.47	0.560	0.4438	0.628	0.0832	0.3013	0.665	0.0684	0.1622			

* At fw=0.01, relative permeability data of section 3 are calculated from the total pressure drops of sec 3&4

4

5

Table B4. CO₂-Brine Relative Permeability Calculated with the Previous Method in Exp6

fw	Time (PV)	sec1		sec2			sec3			sec4		
		Sw	krw	Sw	krw	krw	Sw	krw	krw	Sw	krw	krw
0.5	0.32	0.879	0.0511	0.892	0.5584	0.0479	0.901	0.3458	0.0267	0.911	0.5907	0.0368
	0.10	0.754	0.1362	0.825	0.5893	0.0568	0.854	0.3353	0.0238	0.870	0.7003	0.0405
	0.20	0.730	0.1906	0.780	0.3490	0.1132	0.806	0.2243	0.0471	0.833	0.4187	0.0638

0.1	0.32	0.728	0.2034	0.766	0.2200	0.1394	0.782	0.1465	0.0717	0.808	0.2133	0.0791
	0.41	0.729	0.2050	0.763	0.2029	0.1477	0.773	0.1242	0.0811	0.800	0.1511	0.0836
	0.50	0.732	0.2050	0.763	0.1877	0.1503	0.768	0.1162	0.0872	0.787	0.1344	0.0858
0.01	1.10	0.586	0.4271	0.665	0.1231	0.2773	0.703	0.1240	0.1877			
	1.47	0.560	0.4419	0.628	0.1390	0.2968	0.665	0.1279	0.1574			

* At fw=0.01, relative permeability data of section 3 are calculated from the total pressure drops of sec 3&4

1

2

Table B5. CO₂-Brine Relative Permeability Calculated with the New Method in Exp7d

fw	Time	sec1		sec2			sec4		
	(PV)	Sw	krw	Sw	krw	krw	Sw	krw	krw
0.5	0.25	0.896	0.0483				0.924	0.4998	0.0353
	0.30	0.900	0.0485				0.921	0.4853	0.0365
	0.35	0.899	0.0483				0.919	0.5130	0.0389
0.1	0.20	0.745	0.1654	0.796	0.2763	0.1111			
	0.25	0.743	0.1773	0.787	0.2422	0.1156			
	0.30	0.741	0.1778	0.778	0.2310	0.1221			
	0.35	0.742	0.1786	0.776	0.2167	0.1233			
	0.40	0.742	0.1784	0.773	0.2179	0.1288			
	0.45	0.744	0.1792	0.775	0.2135	0.1301			
	0.50	0.745	0.1791	0.774	0.2087	0.1303			
	0.55	0.746	0.1800	0.772	0.2086	0.1332			
	0.60	0.743	0.1810	0.773	0.2122	0.1354			
	0.65	0.746	0.1806	0.775	0.2110	0.1362			
	0.70	0.745	0.1804	0.772	0.2076	0.1358			
	0.75	0.744	0.1801	0.774	0.2062	0.1346			
	0.80	0.743	0.1808	0.774	0.2083	0.1365			
	0.85	0.746	0.1815	0.774	0.2055	0.1364			
	0.90	0.745	0.1804	0.774	0.2052	0.1367			
0.01	0.95	0.745	0.1806	0.773	0.2044	0.1370			
	1.00	0.744	0.1818	0.774	0.2015	0.1347			
	1.10	0.740	0.1813	0.770	0.2038	0.1374			
	1.20	0.732	0.1811	0.764	0.2049	0.1383			
	1.30	0.730	0.1813	0.762	0.2031	0.1377			
	0.45	0.636	0.3223	0.731	0.1472	0.1768			
	0.50	0.631	0.3293	0.728	0.1384	0.1799			
	0.55	0.627	0.3334	0.721	0.1246	0.1802			
	0.60	0.625	0.3398	0.717	0.1164	0.1837			
	0.65	0.621	0.3378	0.710	0.1092	0.1889			
	0.70	0.618	0.3405	0.706	0.1034	0.1906			
	0.75	0.615	0.3465	0.704	0.1007	0.1962			
	0.80	0.612	0.3435	0.698	0.1004	0.2098			
	0.85	0.611	0.3444	0.695	0.0974	0.2154			
	0.90	0.608	0.3464	0.692	0.0947	0.2183			
	0.95	0.605	0.3535	0.690	0.0931	0.2236			
	1.00	0.605	0.3626	0.686	0.0867	0.2230			
	1.10	0.595	0.3657	0.675	0.0826	0.2280			
	1.20	0.582	0.3496	0.661	0.0786	0.2320			
	1.30	0.581	0.3590	0.659	0.0742	0.2330			
	1.40	0.579	0.3626	0.655	0.0721	0.2396			
	1.50	0.574	0.3621	0.648	0.0695	0.2438			
	1.60	0.567	0.3494	0.641	0.0670	0.2434			
	1.70	0.567	0.3671	0.642	0.0649	0.2449			

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Table B6. CO₂-Brine Relative Permeability Calculated with the Previous Method in Exp7d

fw	Time	sec1		sec2			sec4		
	(PV)	Sw	krw	Sw	krw	krw	Sw	krw	krw
0.5	0.25	0.896	0.0490				0.924	0.5296	0.0329
	0.30	0.900	0.0503				0.921	0.4599	0.0385
	0.35	0.899	0.0480				0.919	0.5135	0.0389
0.1	0.20	0.745	0.1676	0.796	0.2505	0.1132			
	0.25	0.743	0.1806	0.787	0.2032	0.1188			
	0.30	0.741	0.1783	0.778	0.2235	0.1228			
	0.35	0.742	0.1820	0.776	0.1647	0.1275			
	0.40	0.742	0.1796	0.773	0.1924	0.1309			
	0.45	0.744	0.1821	0.775	0.1519	0.1351			
	0.50	0.745	0.1805	0.774	0.1826	0.1324			
	0.55	0.746	0.1815	0.772	0.1823	0.1353			
	0.60	0.743	0.1802	0.773	0.2126	0.1354			
	0.65	0.746	0.1837	0.775	0.1553	0.1407			
	0.70	0.745	0.1803	0.772	0.2084	0.1357			
	0.75	0.744	0.1802	0.774	0.1931	0.1357			
	0.80	0.743	0.1804	0.774	0.2013	0.1370			
	0.85	0.746	0.1841	0.774	0.1604	0.1401			
	0.90	0.745	0.1805	0.774	0.1963	0.1374			
	0.95	0.745	0.1811	0.773	0.1969	0.1377			
	1.00	0.744	0.1821	0.774	0.1911	0.1356			
	1.10	0.740	0.1807	0.770	0.2196	0.1361			
	1.20	0.732	0.1792	0.764	0.2394	0.1355			
	1.30	0.730	0.1812	0.762	0.2067	0.1374			
0.01	0.45	0.636	0.3240	0.731	0.1140	0.1795			
	0.50	0.631	0.3306	0.728	0.0990	0.1831			
	0.55	0.627	0.3353	0.721	0.0924	0.1828			
	0.60	0.625	0.3416	0.717	0.0790	0.1867			
	0.65	0.621	0.3389	0.710	0.0962	0.1899			
	0.70	0.618	0.3410	0.706	0.0824	0.1923			
	0.75	0.615	0.3480	0.704	0.0693	0.1987			
	0.80	0.612	0.3442	0.698	0.0935	0.2104			
	0.85	0.611	0.3460	0.695	0.0707	0.2175			
	0.90	0.608	0.3461	0.692	0.0816	0.2194			
	0.95	0.605	0.3523	0.690	0.0724	0.2253			
	1.00	0.605	0.3660	0.686	0.0467	0.2262			
	1.10	0.595	0.3631	0.675	0.1331	0.2240			
	1.20	0.582	0.3463	0.661	0.1551	0.2259			
	1.30	0.581	0.3598	0.659	0.0443	0.2355			
	1.40	0.579	0.3631	0.655	0.0590	0.2407			
	1.50	0.574	0.3617	0.648	0.0894	0.2423			
	1.60	0.567	0.3468	0.641	0.1008	0.2407			
	1.70	0.567	0.3690	0.642	0.0283	0.2478			

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Table B7. CO₂-Brine Relative Permeability Calculated with the New Method in Exp8d

fw	Time	sec1		sec2			sec3			sec4		
	(PV)	Sw	krw	Sw	krw	krw	Sw	krw	krw	Sw	krw	krw
	0.25	0.673	0.2524	0.790	0.1919	0.1132	0.833	0.2237	0.0774	0.874	0.2905	0.0603
	0.30	0.660	0.2590	0.774	0.1796	0.1332	0.813	0.1925	0.0889	0.844	0.2405	0.0746
	0.35	0.648	0.2642	0.761	0.1549	0.1383	0.797	0.1662	0.0951	0.827	0.2155	0.0841
	0.40	0.638	0.2852	0.751	0.1429	0.1472	0.784	0.1542	0.1059	0.814	0.1986	0.0934
	0.45	0.630	0.2967	0.741	0.1398	0.1675	0.776	0.1381	0.1087	0.806	0.1803	0.0969
	0.50	0.622	0.3097	0.735	0.1279	0.1696	0.770	0.1317	0.1150	0.797	0.1728	0.1066
	0.55	0.618	0.3091	0.727	0.1129	0.1690	0.760	0.1272	0.1271	0.789	0.1616	0.1130

0	0.60	0.611	0.3139	0.722	0.1060	0.1732	0.755	0.1168	0.1282	0.784	0.1556	0.1183
	0.65	0.607	0.3166	0.715	0.0956	0.1737	0.749	0.1088	0.1311	0.777	0.1538	0.1296
	0.70	0.602	0.3168	0.710	0.0901	0.1780	0.741	0.0988	0.1317	0.770	0.1443	0.1330
	0.76	0.594	0.3377	0.700	0.0823	0.1802	0.735	0.0970	0.1371	0.763	0.1361	0.1362
	0.80	0.589	0.3360	0.694	0.0779	0.1813	0.723	0.0907	0.1447	0.753	0.1267	0.1377
	0.85	0.588	0.3434	0.694	0.0777	0.1911	0.727	0.0915	0.1487	0.756	0.1218	0.1388
	0.90	0.585	0.3513	0.690	0.0727	0.1893	0.723	0.0879	0.1525	0.750	0.1127	0.1396
	0.95	0.578	0.3535	0.687	0.0732	0.1969	0.720	0.0865	0.1557	0.748	0.1117	0.1434
	1.00	0.578	0.3673	0.683	0.0696	0.2035	0.715	0.0808	0.1597	0.744	0.1100	0.1518
	1.05	0.575	0.3651	0.679	0.0701	0.2167	0.711	0.0771	0.1597	0.739	0.1075	0.1576
	1.10	0.572	0.3629	0.676	0.0659	0.2143	0.708	0.0743	0.1616	0.737	0.1047	0.1595
	1.15	0.568	0.3757	0.674	0.0656	0.2206	0.706	0.0722	0.1618	0.735	0.1059	0.1681
	1.20	0.564	0.3677	0.669	0.0634	0.2213	0.701	0.0704	0.1660	0.731	0.1021	0.1680
	1.25	0.563	0.3751	0.667	0.0595	0.2222	0.698	0.0658	0.1657	0.728	0.0974	0.1695
	1.30	0.560	0.3735	0.664	0.0574	0.2218	0.696	0.0653	0.1700	0.724	0.0935	0.1706
	1.35	0.557	0.3785	0.661	0.0547	0.2203	0.692	0.0633	0.1735	0.722	0.0909	0.1711
	1.40	0.554	0.3794	0.660	0.0545	0.2250	0.690	0.0657	0.1851	0.720	0.0884	0.1721
	1.45	0.552	0.3786	0.656	0.0525	0.2258	0.689	0.0638	0.1827	0.716	0.0841	0.1713
	1.50	0.549	0.3803	0.654	0.0529	0.2332	0.687	0.0660	0.1951	0.715	0.0831	0.1732
	1.60	0.541	0.3831	0.643	0.0480	0.2317	0.675	0.0607	0.1970	0.704	0.0781	0.1764
	1.70	0.530	0.3831	0.633	0.0458	0.2345	0.666	0.0590	0.2001	0.694	0.0755	0.1810
	1.80	0.525	0.3832	0.627	0.0421	0.2285	0.660	0.0565	0.2044	0.689	0.0725	0.1841
	1.90	0.520	0.3816	0.624	0.0416	0.2367	0.657	0.0542	0.2051	0.685	0.0679	0.1832
	2.00	0.518	0.3817	0.619	0.0393	0.2409	0.650	0.0495	0.2029	0.680	0.0666	0.1893

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Table B8. CO₂-Brine Relative Permeability Calculated with the Previous Method in Exp8d

fw	Time	sec1		sec2			sec3			sec4		
	(PV)	Sw	kr _g	Sw	kr _w	kr _g	Sw	kr _w	kr _g	Sw	kr _w	kr _g
0	0.25	0.673	0.2533	0.790	0.1683	0.1151	0.833	0.2387	0.0762	0.874	0.3376	0.0565
	0.30	0.660	0.2572	0.774	0.1709	0.1340	0.813	0.1927	0.0889	0.844	0.2748	0.0718
	0.35	0.648	0.2630	0.761	0.1564	0.1382	0.797	0.1779	0.0942	0.827	0.2356	0.0825
	0.40	0.638	0.2845	0.751	0.1318	0.1481	0.784	0.1492	0.1063	0.814	0.1963	0.0936
	0.45	0.630	0.2960	0.741	0.1197	0.1691	0.776	0.1265	0.1097	0.806	0.1566	0.0988
	0.50	0.622	0.3086	0.735	0.1168	0.1705	0.770	0.1146	0.1164	0.797	0.1508	0.1084
	0.55	0.618	0.3106	0.727	0.0762	0.1719	0.760	0.1080	0.1286	0.789	0.1413	0.1147
	0.60	0.611	0.3123	0.722	0.0922	0.1743	0.755	0.0993	0.1296	0.784	0.1208	0.1212
	0.65	0.607	0.3169	0.715	0.0837	0.1747	0.749	0.1018	0.1317	0.777	0.1400	0.1307
	0.70	0.602	0.3163	0.710	0.0727	0.1794	0.741	0.0918	0.1322	0.770	0.1400	0.1334
	0.76	0.594	0.3356	0.700	0.1108	0.1779	0.735	0.1279	0.1347	0.763	0.1602	0.1342
	0.80	0.589	0.3347	0.694	0.1058	0.1791	0.723	0.1526	0.1398	0.753	0.2346	0.1290
	0.85	0.588	0.3436	0.694	0.0260	0.1953	0.727	0.0206	0.1544	0.756	0.0196	0.1471
	0.90	0.585	0.3511	0.690	0.0688	0.1896	0.723	0.0777	0.1534	0.750	0.1071	0.1400
	0.95	0.578	0.3502	0.687	0.0902	0.1956	0.720	0.0960	0.1550	0.748	0.1063	0.1438
	1.00	0.578	0.3681	0.683	0.0412	0.2058	0.715	0.0579	0.1616	0.744	0.0873	0.1536
	1.05	0.575	0.3649	0.679	0.0685	0.2169	0.711	0.0802	0.1594	0.739	0.1078	0.1576
	1.10	0.572	0.3617	0.676	0.0592	0.2148	0.708	0.0685	0.1621	0.737	0.0864	0.1610
	1.15	0.568	0.3753	0.674	0.0596	0.2211	0.706	0.0637	0.1624	0.735	0.0774	0.1704
	1.20	0.564	0.3643	0.669	0.0951	0.2188	0.701	0.0980	0.1638	0.731	0.1373	0.1652
	1.25	0.563	0.3750	0.667	0.0404	0.2238	0.698	0.0487	0.1671	0.728	0.0717	0.1716
	1.30	0.560	0.3726	0.664	0.0541	0.2221	0.696	0.0627	0.1702	0.724	0.0908	0.1708
	1.35	0.557	0.3777	0.661	0.0607	0.2198	0.692	0.0731	0.1728	0.722	0.0914	0.1711
	1.40	0.554	0.3783	0.660	0.0494	0.2254	0.690	0.0542	0.1861	0.720	0.0707	0.1735
	1.45	0.552	0.3772	0.656	0.0538	0.2257	0.689	0.0658	0.1826	0.716	0.0763	0.1720
	1.50	0.549	0.3788	0.654	0.0554	0.2330	0.687	0.0612	0.1955	0.715	0.0636	0.1748
	1.60	0.541	0.3806	0.643	0.0962	0.2279	0.675	0.1298	0.1915	0.704	0.1629	0.1695
	1.70	0.530	0.3791	0.633	0.1050	0.2297	0.666	0.1296	0.1945	0.694	0.1527	0.1748

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Table B9. CO₂-Brine Relative Permeability Calculated with the New Method in Exp9d

fw	Time	sec1			sec2			sec3			sec4		
	(PV)	Sw	krw	Sw	krw	krw	Sw	krw	krw	Sw	krw	krw	
0	0.25	0.677	0.2356	0.752	0.1395	0.1302	0.791	0.1942	0.0933	0.842	0.2847	0.0656	
	0.30	0.663	0.2572	0.737	0.1296	0.1484	0.768	0.1581	0.1043	0.809	0.2286	0.0813	
	0.35	0.651	0.2721	0.726	0.1164	0.1535	0.756	0.1429	0.1126	0.795	0.2137	0.0940	
	0.41	0.641	0.2956	0.715	0.1069	0.1674	0.746	0.1280	0.1204	0.783	0.1986	0.1076	
	0.45	0.637	0.3120	0.707	0.0955	0.1758	0.737	0.1171	0.1295	0.772	0.1752	0.1127	
	0.50	0.630	0.3257	0.702	0.0912	0.1864	0.729	0.1071	0.1346	0.767	0.1643	0.1172	
	0.55	0.624	0.3259	0.698	0.0874	0.1896	0.724	0.1019	0.1394	0.761	0.1576	0.1235	
	0.60	0.615	0.3274	0.690	0.0858	0.2008	0.719	0.1013	0.1473	0.756	0.1520	0.1287	
	0.65	0.612	0.3366	0.685	0.0799	0.2069	0.713	0.0932	0.1508	0.749	0.1426	0.1349	
	0.70	0.608	0.3483	0.679	0.0736	0.2142	0.707	0.0891	0.1583	0.744	0.1342	0.1380	
	0.75	0.603	0.3552	0.676	0.0713	0.2161	0.703	0.0867	0.1652	0.741	0.1307	0.1428	
	0.80	0.603	0.3608	0.672	0.0646	0.2192	0.698	0.0858	0.1839	0.737	0.1174	0.1396	
	0.95	0.588	0.3734	0.659	0.0612	0.2405	0.687	0.0779	0.1893	0.725	0.1096	0.1544	
	1.00	0.583	0.3845	0.650	0.0558	0.2459	0.678	0.0691	0.1848	0.716	0.1115	0.1690	

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Table B10. CO₂-Brine Relative Permeability Calculated with the Previous Method in Exp9d

fw	Time (PV)	sec1		sec2			sec3			sec4		
		Sw	krq	Sw	krw	krq	Sw	krw	krq	Sw	krw	krq
0	0.25	0.677	0.2332	0.752	0.2033	0.1251	0.791	0.2798	0.0865	0.842	0.3914	0.0570
	0.30	0.663	0.2539	0.737	0.1960	0.1431	0.768	0.2335	0.0982	0.809	0.3112	0.0747
	0.35	0.651	0.2668	0.726	0.1697	0.1492	0.756	0.1868	0.1091	0.795	0.2283	0.0928
	0.41	0.641	0.2941	0.715	0.1310	0.1654	0.746	0.1370	0.1197	0.783	0.1831	0.1089
	0.45	0.637	0.3161	0.707	0.0867	0.1765	0.737	0.1307	0.1284	0.772	0.1811	0.1123
	0.50	0.630	0.3233	0.702	0.1020	0.1855	0.729	0.1196	0.1336	0.767	0.1408	0.1191
	0.55	0.624	0.3231	0.698	0.1043	0.1882	0.724	0.1084	0.1389	0.761	0.1281	0.1259
	0.60	0.615	0.3232	0.690	0.1491	0.1957	0.719	0.1505	0.1434	0.756	0.1703	0.1273
	0.65	0.612	0.3362	0.685	0.0761	0.2072	0.713	0.0897	0.1511	0.749	0.1243	0.1364
	0.70	0.608	0.3498	0.679	0.0746	0.2141	0.707	0.0998	0.1574	0.744	0.1179	0.1393
	0.75	0.603	0.3522	0.676	0.0847	0.2150	0.703	0.0894	0.1650	0.741	0.1040	0.1450
	0.80	0.603	0.3651	0.672	0.0224	0.2226	0.698	0.0517	0.1867	0.737	0.0670	0.1436
	0.95	0.588	0.3712	0.659	0.1001	0.2374	0.687	0.1114	0.1866	0.725	0.1180	0.1537
	1.00	0.583	0.3830	0.650	0.1330	0.2397	0.678	0.1797	0.1759	0.716	0.2332	0.1592

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