Experimental investigation of hysteretic dynamic capillarity effect in unsaturated flow

Citation for published version (APA):

DOI:
10.1002/2017WR020895

Document status and date:
Published: 01/11/2017

Document Version:
Publisher’s PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:

• A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher’s website.
• The final author version and the galley proof are versions of the publication after peer review.
• The final published version features the final layout of the paper including the volume, issue and page numbers.

Link to publication

General rights
Copyright and moral rights for the publications made accessible in the public portal are retained by the authors and/or other copyright owners and it is a condition of accessing publications that users recognise and abide by the legal requirements associated with these rights.

• Users may download and print one copy of any publication from the public portal for the purpose of private study or research.
• You may not further distribute the material or use it for any profit-making activity or commercial gain
• You may freely distribute the URL identifying the publication in the public portal.

If the publication is distributed under the terms of Article 25fa of the Dutch Copyright Act, indicated by the “Taverne” license above, please follow below link for the End User Agreement:
www.tue.nl/taverne

Take down policy
If you believe that this document breaches copyright please contact us at:
openaccess@tue.nl
providing details and we will investigate your claim.

Download date: 05. Aug. 2019
Experimental Investigation of Hysteretic Dynamic Capillarity Effect in Unsaturated Flow

Luwen Zhuang1, S. Majid Hassanizadeh1,2, Chao-Zhong Qin3, and Arjen de Waal1

1Department of Earth Sciences, Utrecht University, Utrecht, The Netherlands, 2Soil and Groundwater Systems, Deltares, Utrecht, The Netherlands, 3Department of Mechanical Engineering, Eindhoven University of Technology, Eindhoven, The Netherlands

Abstract The difference between average pressures of two immiscible fluids is commonly assumed to be the same as macroscopic capillary pressure, which is considered to be a function of saturation only. However, under transient conditions, a dependence of this pressure difference on the time rate of saturation change has been observed by many researchers. This is commonly referred to as dynamic capillarity effect. As a first-order approximation, the dynamic term is assumed to be linearly dependent on the time rate of change of saturation, through a material coefficient denoted by γ. In this study, a series of laboratory experiments were carried out to quantify the dynamic capillarity effect in an unsaturated sandy soil. Primary, main, and scanning drainage experiments, under both static and dynamic conditions, were performed on a sandy soil in a small cell. The value of the dynamic capillarity coefficient γ was calculated from the air-water pressure differences and average saturation values during static and dynamic drainage experiments. We found a dependence of γ on saturation, which showed a similar trend for all drainage conditions. However, at any given saturation, the value of γ for primary drainage was larger than the value for main drainage and that was in turn larger than the value for scanning drainage. Each data set was fit a simple log-linear equation, with different values of fitting parameters. This nonuniqueness of the relationship between γ and saturation and possible causes is discussed.

1. Introduction

Capillary pressure is of importance in the quantification of multiphase flow in porous media. Capillary pressure is defined as the difference between the pressures of two fluids, and is always expressed as a function of saturation. The resulting relationship is commonly measured during quasi-static conditions. However, due to the dynamics of multiphase flow, particularly for fine porous media, measurements of the capillary pressure curves are time-consuming, often taking weeks or even months. Experimental methods, therefore, have been developed for rapid measurements of the relationship between capillary pressure and saturation (e.g., Elzeftawy & Mansell, 1975; Smiles et al., 1971; Stauffer, 1978; Topp & Peters, 1967; Vachaud et al., 1972; Wildenschild et al., 2001). However, many have observed a rate-dependence in this relationship when obtained using fast measurements. A number of concepts and formulations have been proposed to describe this phenomenon (Barenblatt & Gil’man, 1987; Cueto-Felgueroso & Juanes, 2009; Hassanizadeh & Gray, 1990; Hilpert, 2012; Kalaydjian, 1992; Stauffer, 1978).

In this study, we follow the thermodynamic approach proposed by Hassanizadeh and Gray (1990, 1993). In this approach, capillary pressure is shown to be equal to the difference between fluid pressures only during quasi-static conditions. However, they are not equal during dynamic conditions. Indeed, the difference between macroscale fluids pressure difference and capillary pressure is found to depend on the time rate of change of saturation. In fact, under dynamic conditions, the difference between average pressures of the two fluids is not only due to capillary pressure, but also because of pressure gradient needed for overcoming viscous forces. So, the fluids pressure difference could be far from the average capillary pressure, depending on how fast the flow is (see, e.g., Hassanizadeh, 2015). The following linear approximation has been proposed (see, e.g., Hassanizadeh & Gray, 1993; Stauffer, 1978):
\[ p^n - p^w - P^c = -\tau \frac{\partial S^w}{\partial t} \] (1)

where \( p^n \) and \( p^w \) are nonwetting and wetting phase pressures under dynamic conditions, respectively, \( P^c \) is the capillary pressure, \( S^w \) is the wetting phase saturation, and \( \tau \) is a dynamic (or nonequilibrium) capillarity coefficient (or dynamic capillary coefficient). The coefficient \( \tau \) is a damping coefficient depending on material properties as well as the state of the system. Clearly, the difference between the two fluid pressures is equivalent to the capillary pressure during quasi-static conditions (i.e., \( \partial S^w/\partial t = 0 \)). Some authors have referred to \( p^n - p^w \) as “dynamic capillary pressure.” But, as it is well known, capillary pressure is a property of the solid-fluids system and it relates to processes occurring at the menisci. Fluids pressure differences, however, are not material properties. They are dictated by boundary and initial conditions and are different from capillary effects; for example, they depend on flow rate, which is not a material property. So, the term capillary pressure should not be used for fluids pressure difference (see also the discussion in Joekar-Niasar and Hassanizadeh (2012b)).

The values reported for \( \tau \) in the literature vary over a wide range. Hassanizadeh et al. (2002) calculated approximate values of \( \tau \), ranging from \( 3 \times 10^4 \) to \( 5 \times 10^7 \) Pa s, based on published experimental studies of dynamic capillarity effect. The values of \( \tau \) in numerical (see Diamantopoulos & Durner, 2012; Joekar-Niasar & Hassanizadeh, 2012a, for a review) and experimental studies (e.g., Bottero et al., 2011; Camps-Roach et al., 2010; Das & Mirzaei, 2012; Goel & O’Carroll, 2011; Lo et al., 2017; O’Carroll et al., 2005; Sakaki et al., 2010) have been mostly within this range. Part of the variation in reported values can be explained by the fact that \( \tau \) depends on saturation. However, various experimental studies have reported different or even contradictory dependence of \( \tau \) on saturation. Some studies have reported that \( \tau \) varies only slightly with saturation (e.g., Bottero et al., 2011; Camps-Roach et al., 2010; Oung et al., 2005), while others found that the value of \( \tau \) increased or decreased as the saturation decreased (e.g., Das & Mirzaei, 2012; Sakaki et al., 2010).

The scale-dependence of \( \tau \) has also been reported in the literature. In addition to the numerical study by Manthey et al. (2008), experimental works have been performed in experimental setups at different scales (Bottero et al., 2011; Camps-Roach et al., 2010; Das & Mirzaei, 2012; Goel & O’Carroll, 2011; O’Carroll et al., 2005, 2010; Oung et al., 2005; Sakaki et al., 2010). A few experimental studies have been carried out in small-volume sand columns (Abidoye & Das, 2014; Hassanizadeh et al., 2004; Hou et al., 2012). Hassanizadeh et al. (2004) estimated the magnitude of \( \tau \), based on a series of drainage experiments in a column with diameter 6 cm and height 3 cm, where water was displaced by PCE (Perchloroethylene). They found values ranging from \( 5 \times 10^5 \) to \( 6 \times 10^5 \) Pa s. Abidoye and Das (2014) calculated \( \tau \) values from silicone oil-water drainage experiments in a 4 cm long sand column. They found that the value of \( \tau \) increased as saturation decreased, ranging from \( 2 \times 10^5 \) to \( 1 \times 10^7 \) Pa s for silicone oils with different viscosities. In experiments in an unsaturated small sand column (1.27 cm in height and 2.54 cm in diameter), Hou et al. (2012) found \( \tau \) values of \( 10^3 \) Pa s or even smaller during dynamic drainage.

Potential dynamic effects in relative permeability have been also considered and have received limited attention in the literature (see, e.g., Goel et al., 2016; Joekar-Niasar & Hassanizadeh, 2011; Stauffer, 1978). Stauffer (1978) determined relative permeability under steady state and dynamic conditions in a soil column and found that differences were not significant. The dynamic model of Barenblatt and Gil’man (1987) also accounts for dynamic effects in relative permeability. Numerical studies were done by Joekar-Niasar and Hassanizadeh (2011) on the dynamic effect in relative permeability. Goel et al. (2016) have found there existed the differences between quasi-static and dynamic relative permeability-saturation curves experimentally. In this study, we are not considering that effect.

However, almost all reported studies only involved primary drainage processes. In particular, no experimental studies have been conducted to provide any information about the dynamic capillarity coefficient for scanning drainage or imbibition processes. Cycles of imbibition and drainage (main or scanning) may occur in many field applications, such as irrigation, groundwater recharge, infiltration and redistribution in vadose zone, and root uptake processes. They are also encountered in industrial applications, such penetration of ink into paper and its subsequent drying (see, e.g., Aslannejad et al., 2017), redistribution of moisture in layers of fuel cell (see, e.g., Qin, 2015), and infiltration and drainage of liquids in hygienic products such as diapers. Also, investigation of dynamic scanning drainage or imbibition will give a full insight into the dynamic capillarity coefficient over the entire hysteretic loops in a porous medium system.
In order to address some of the inconsistencies and shortcomings mentioned above, we performed a series of dynamic drainage experiments in a small-volume unsaturated sandy soil. The relationship between capillary pressure and saturation were first measured during quasi-static conditions. Afterward, primary, main, and scanning drainage experiments were carried out to investigate capillarity effect during dynamic drainage. Because the response time of the measurement devices could influence the estimation of dynamic capillarity, we used pressure transducers with a very fast response to measure water pressure during experiments. Results of experiments were used to estimate the values of the dynamic capillarity coefficient. This is the first time that values of $\tau$ at different saturations are obtained not only for primary and main drainage processes, but also for scanning processes.

2. Materials and Methods

2.1. Materials
The sand used in all experiments was obtained from a sand mining site (Sibelco, Antwerp, Belgium). It had a relatively narrow pore size distribution with particle diameters ranging from 0.1 to 0.5 mm. Prior to use, the sand was thoroughly washed a few times with deionized water and then air dried, in order to remove fine clay particles. The sand properties are listed in Table 1. All experiments were conducted under unsaturated conditions, with air as the nonwetting phase. Deionized, distilled, and degassed water was used as the wetting phase.

2.2. Experimental Setup
All experiments were conducted using a custom-built Plexiglas sandbox. A schematic view of the experimental setup is shown in Figure 1. The dimensions of the sandbox were 3 cm (height) $\times$ 3 cm (length) $\times$ 2 cm (width). A valve at the top of sandbox (Valve 1) connected the headspace to a sealed air bag, filled with moist air, in order to keep the unsaturated sand under constant atmospheric pressure. At the bottom of sandbox, we used a hydrophilic nylon membrane (VWR International B.V., the Netherlands; mean pore size, 5 $\mu$m), supported by a stainless-steel porous plate, to serve as a capillary barrier to the air phase. The water reservoir at the bottom of setup (below the porous plate) was connected with a short tube to a small water column, which was used to control the pressure head at the column outlet. The small hanging column had an overflow to allow drained water to leave from the column, and thus keep the water head in

![Figure 1. Schematic view of the experimental setup.](image)

<table>
<thead>
<tr>
<th>Properties</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Mean particle diameter, $d_{50}$ (mm)</td>
<td>0.20</td>
</tr>
<tr>
<td>Particle density, $\rho_s$ (g cm$^{-3}$)</td>
<td>2.56</td>
</tr>
<tr>
<td>Intrinsic permeability (m$^2$)</td>
<td>$1.7 \times 10^{-11}$</td>
</tr>
<tr>
<td>Average porosity, $\phi$</td>
<td>0.39</td>
</tr>
</tbody>
</table>

Table 1
Properties of the Sand Used in Experiments
the column constant. Silicone tapes were used at all joints to avoid any leakage. The entire sandbox was fixed with the aid of a frame on a three-digit precision balance (Kern & Sohn GmbH, Germany). Readings of the balance were used to calculate changes in average saturation.

Two pressure transducers (HMUM100, First Sensor, Germany) were installed at depths of 1 and 2 cm alongside the sandbox (see Figure 1). The transducers did not protrude into the soil. The dead volume of the transducer was saturated with water before installation. The transducer opening had a diameter of 5 mm. It was covered by a hydrophilic membrane with a mean pore size of 0.45 μm (PVDF, Mdimembrane, India), which was in contact with the soil. The membrane was saturated with degassed water in all experiments. The two pressure transducers were connected to a CR1000 data logger (Campbell Scientific, Shepshed, UK). The pressure transducers were calibrated prior to use in the experiments. The response time of the transducers was determined to be 0.4 s, based on a test explained in Appendix A. The inner surface of the cell was scrubbed using a sandpaper. The hydrophilic membrane between the transducers and the sand was soft so that the sand particles would be in fully contact with the membrane.

2.3. Quasi-Static and Dynamic Drainage Experiments
The experimental setup was first mounted with the bottom reservoir and the cell being full of water and with the top lid removed. Deionized and degassed water was used to minimize air entrapment. The cell was packed by continuously pouring dry sand into the water through a funnel, and it was regularly tapped. A small comb was used to mix the sand as it was being poured into water to avoid layering. This resulted in a fairly uniform packing. The top lid was then installed. Thus, the sand sample was initially fully saturated. Each time a new series of primary drainage experiments was needed to be done, the column was repacked following above procedure. The packing porosity was the same (less than 2% difference) in all cases; otherwise, the cell was repacked to achieve that.

At the start of all quasi-static experiments, the level of water in the hanging water column was kept at the same level as the bottom of the sand sample. Valve 2 and Valve 3 (see Figure 1) were always open. For the primary, main, and scanning drainage processes, the elevation of the hanging column was decreased in small increments (1.5–2 cm H$_2$O). For the main imbibition process, the column was rewetted by raising the hanging water column at incremental steps, while adding water in order to keep water level at the outflow level. The readings from pressure transducers were monitored to ensure that the equilibrium was established before each change of elevation. The duration was almost 2 weeks for each $p^-$-$S^+$ loop (from primary drainage to main imbibition to main drainage, or to a scanning drainage).

All dynamic drainage experiments (primary, main, and scanning) were started under initially unsaturated conditions. In Appendix B, we explain why the dynamic primary drainage experiment was not performed starting at full initial saturation but at a saturation of about 0.83. To obtain the desired initial water saturation (pressure), the saturated sand sample was first drained (for primary drainage) or drained and rewetted (for scanning and main drainage) under quasi-static conditions. After reaching the desired water saturation (as the initial conditions for dynamic experiments), Valve 3 was closed and the hanging water column was lowered to 70 cm below the bottom of the sandbox. Dynamic drainage experiments started by opening Valve 3. This resulted in a rapid drainage of the sand sample. Data on local water pressures and average water saturation were collected every 0.5 s for a maximum duration of 4,000 s. We ensured that the initial water saturation and pressure for each loop of dynamic drainage experiments (primary, scanning, or main) were essentially identical (less than 3% difference).

In principle, dynamic imbibition experiments could be performed also using the above experimental setup. However, due to some shortcomings, we were not able to use the data from dynamic imbibition experiments. The problem was that the initial saturation of the main imbibition curve had to be approximately equal to the irreducible saturation. This resulted in a very low permeability of the sand sample. So, when dynamic imbibition started, water entering the sandbox from below started to accumulate at the bottom of the sand sample. Then, it moved slowly upward and reached the pressure transducers. This caused some mismatch between the saturation and the pressure readings during the dynamic imbibition processes; the average saturation started to change, while no change in water pressure was being registered. For this reason, in this study, we focused only on the dynamic drainage experiments. For dynamic imbibition experiments, we need to improve the experimental setup by adding a device (e.g., time-domain reflectometer) to measure local saturation in experiments.
Since we measured average saturations during the experiments, we calculated the corresponding arithmetic mean pressure values based on readings of the two pressure transducers for both quasi-static and dynamic experiments. All experiments were conducted in a constant-temperature room at $21 \pm 0.5 \, ^\circ C$. At least two replicates were conducted for each set of experiments.

3. Results

3.1. Quasi-Static Experiments

The $p^e-S^w$ data obtained during quasi-static conditions are shown in Figure 2. Different symbols represent measured data during the primary, main, and scanning drainage and main imbibition processes. The measured data were fitted by van Genuchten equation (1980) written as

$$p^e(S^w) = \frac{1}{a} \left[ S_e^{-1/m} - 1 \right]^{1/n}$$

In this formula, the effective saturation $S_e$ is expressed as

$$S_e = \frac{S^w - S^w_{ir}}{1 - S^w_{ir} - S^a_r}$$

where $S^w_{ir}$, $S^a_r$, and $S_e$ are the irreducible water saturation, the residual air saturation, and the effective water saturation, respectively, $a$ and $n$ are the fitting parameters, and $m = 1 - 1/n$. The fitted curves are shown as solid lines in Figure 2. The irreducible water saturation $S^w_{ir}$ was set to the same value for all $p^e-S^w$ curves. The residual air saturation $S^a_r$, $a$, and $n$ were fitted for each curve separately. The resulting parameters are listed in Table 2. These fitted curves along with the dynamic experimental data were used later to calculate the values of the dynamic capillarity coefficient $\tau$.

3.2. Dynamic Drainage Experiments

Measured water pressures at two elevations and saturation as a function of time during dynamic drainage experiments are shown in Figure 3. Note that only water pressures at early times are shown here, when large changes occurred in water pressure and saturation. As can be seen, water pressure transducers reacted immediately as soon as Valve 3 (Figure 1) was opened ($t = 0$); pressure decreased drastically.

One interesting phenomenon we observed was that the water pressures measured by the lower pressure transducer, which was closer to the outlet, showed a nonmonotonic behavior during initial stage of dynamic main drainage (see the inset in Figure 3c). A similar nonmonotonic behavior in capillary pressure was observed earlier by Bottero et al. (2011), who performed a series of dynamic primary drainage experiments in a PCE-water system. Their experimental results showed that at high injection pressures (when dynamic capillarity became important), the measured capillary pressure showed a nonmonotonic variation at all measurement locations.

The pressure overshoot could be observed only when water flow rate and the initial water saturation were high enough. The initial water pressure difference went from $-10$ to $-70$ cm during main drainage, which was highest among all dynamic drainage processes. Also, the initial water saturation was relatively high (around 0.86) during main drainage. So, it is reasonable that the pressure overshoot was observed during main drainage. Now, there is always a gradient in flow velocity; it was zero at the top surface and largest at the bottom part of the cell. The bottom velocity was highest at the start of the experiment, and that was where the nonmonotonic pressure change was observed. Conditions were not right at upper part of the cell and at later time for this effect to occur. Bottero et al. (2011) also reported that the pressure overshoot was more distinct at the location closer to the injection surface. Furthermore, this nonmonotonic behavior in capillary pressure could be modeled only by including the dynamic capillarity term (Berentsen et al., 2006; Bottero, 2009).

![Figure 2](image_url). Capillary pressure-saturation data measured under quasi-static conditions (symbols) and fitted curves (lines) based on van Genuchten formula. Measurement data from two replicates are shown.

**Table 2**

Values of the Fitted Van Genuchten Parameters

<table>
<thead>
<tr>
<th>Experiment</th>
<th>$a$ (cm$^{-1}$)</th>
<th>$n$</th>
<th>$S^w_{ir}$</th>
<th>$S^a_r$</th>
</tr>
</thead>
<tbody>
<tr>
<td>Primary drainage</td>
<td>0.028</td>
<td>12</td>
<td>0.21</td>
<td>0</td>
</tr>
<tr>
<td>Main imbibition</td>
<td>0.048</td>
<td>10</td>
<td>0.21</td>
<td>0.14</td>
</tr>
<tr>
<td>Main drainage</td>
<td>0.028</td>
<td>12</td>
<td>0.21</td>
<td>0.14</td>
</tr>
<tr>
<td>Scanning drainage</td>
<td>0.026</td>
<td>10</td>
<td>0.21</td>
<td>0.39</td>
</tr>
</tbody>
</table>
As mentioned earlier, readings of the two pressure transducers were averaged to obtain average water pressure at different times. The difference between air and water pressure are plotted as a function of average saturation in Figure 4. The van Genuchten fitted quasi-static curves are shown as solid lines (same as those shown in Figure 2). The two replicates are shown as dashed and solid lines. They almost overlap with each other. Because of the close overlap of the replicates, we did calculations for one set of data only. It is clear that at a given saturation the values of air-water pressure difference are much larger than the corresponding value of capillary pressure for dynamic primary and main drainage. However, for dynamic scanning drainage, the air-water pressure difference initially coincided with the capillary pressure, and then it became larger and deviated from the capillary pressure. It is evident that the deviation in pressure differences between dynamic and quasi-static processes depends on both the imposed boundary and initial conditions (see also Bottero et al., 2011; Camps-Roach et al., 2010; Sakaki et al., 2010).

The rate of change in saturation with time, $\frac{\partial S_w}{\partial t}$, was calculated using a backward difference approximation and plotted as a function of saturation in Figure 5. As expected, the absolute values of $\frac{\partial S_w}{\partial t}$ in all dynamic drainage experiments increased dramatically to reach a maximum value, and then gradually approached zero. The largest maximum absolute value was found to be $0.05 \text{ s}^{-1}$ during main drainage, while it was smallest, around $0.02 \text{ s}^{-1}$, during primary drainage.

3.3. Calculation of the Dynamic Capillarity Coefficient $\tau$

Based on equation (1), calculating the values of $\tau$ requires the corresponding values of $\frac{\partial S_w}{\partial t}$, $p_a - p_w$, and $p_c$ at a given saturation value. Assuming the air pressure to be zero, equation (1) can be recast into
At any given time, the average values of saturation $S_w$ and water pressure $p_w$ were known as explained earlier. The corresponding quasi-static capillary pressure was calculated using the fitted van Genuchten equation given by equation (2). The resulting values of $\tau$ are plotted as a function of saturation in Figure 6. Only positive values are shown, and the data corresponding to small values of $\partial S_w/\partial t$ were discarded.

4. Discussion

4.1. Saturation Dependence of $\tau$

The values of $\tau$ for primary, main, and scanning drainage are shown in Figure 6 using different colored symbols. As shown in the figure, the magnitude of the dynamic capillarity coefficient $\tau$ mainly increased with decreasing water saturation for all dynamic drainage processes. For primary drainage, the value of $\tau$ first decreased to reach minimum values at a saturation of around 0.8, and then increased nearly log-linearly as saturation decreased. For main drainage, the trend is similar to that of primary drainage, but the minimum values of $\tau$ were reached in the saturation range $0.70 < S_w < 0.85$. A similar trend for the $\tau$-$S_w$ relationship for primary or main drainage processes has been reported in the literature (see, e.g., Abidoye & Das, 2014; Camps-Roach et al., 2010; Goel & O’Carroll, 2011; Sakaki et al., 2010). Bottero et al. (2011) reported that the value of $\tau$ varied little in the range $0.50 < S_w < 0.85$, which is more or less the same as in our experiments. By comparison, for scanning drainage, $\tau$ values increased almost log-linearly as saturation decreased within the measured saturation range. At any given saturation, the value of $\tau$ for primary drainage was larger than the value for main drainage and that was in turn larger than the value for scanning drainage. The $\tau$-$S_w$ data were fitted using a log-linear function given as:

$$\log_{10} \tau = A(S_0 - S_w) + \log_{10} \tau_0$$  \hspace{1cm} (5)

in which $S_0$ and $\tau_0$ are some threshold values and $A$ is the slope of the lines shown in Figure 6. The values of fitting parameters $S_0$, $\tau_0$, and $A$ could be various for different dynamic processes. We note that we only did preliminary optimizations here. The value of $\tau_0$ was set to $2 \times 10^4$ Pa s, the minimum value of $\tau$ for primary drainage. The value of $A$ was optimized based on the data for primary drainage, and fixed as the same value when fitting the data for scanning and main drainage. The values of $S_0$ were fitted separately for the three series of data. The fitted values are listed in Table 3.

The data shown in Figure 6 suggest that the $\tau$-$S_w$ relationship is non-unique. Nonuniqueness in the $\tau$-$S_w$ relationship has only been reported by Mirzaei and Das (2013) and Sakaki et al. (2010) for primary drainage and main imbibition. The nonuniqueness in the $\tau$-$S_w$ relationship can only be explained by examining microscale redistribution processes during fast drainage. Such information is currently not available. Nevertheless, we know that water configurations in the pores are definitely different during primary, main, and scanning drainage (under quasi-static or dynamic conditions), at a given macroscale saturation. This in turn leads to nonuniqueness in values of material properties. Well-known examples are nonuniqueness in capillary pressure and relative permeability, which has been attributed to differences in...
Figure 6. Calculated $\tau$ values for primary, main, and scanning drainage. Solid lines show fitting the data using equation (5). Note that the vertical axis is in logarithmic scale.

4.2. Inaccuracies in the Estimation of $\tau$

Scale-dependence of the dynamic capillarity coefficient $\tau$ has been reported in experimental studies by Bottero et al. (2011) and Abidoye and Das (2014), and in numerical studies by Manthey et al. (2005) and Dahle et al. (2005). We note that some measurement artifacts may exist in the estimation of $\tau$. In our study, average saturation was calculated based on the total volume change of the entire sample. In some cases, the average $\partial S^w/\partial t$ may be smaller than the local $\partial S^w/\partial t$ values, leading to some overestimation of $\tau$ values. In fact, Bottero et al. (2011) showed that the local $\partial S^w/\partial t$ may be one order of magnitude larger than the average $\partial S^w/\partial t$ over the length of 11 cm of their setup. However, we note that their experiments were done in a PCE (Perchloroethylene)-water system, with heavier PCE displacing water vertically up against the gravity. This may have led to a very sharp PCE-water front, which in turn may have caused a large difference between local and average values of $\partial S^w/\partial t$. In air-water systems, air is much less viscous and lighter than water. Also, in our setup, the water displacement was downward, and it could have caused smearing of the drainage front. This should lead to small differences between average and local $\partial S^w/\partial t$ values. Camps-Roach et al. (2010) have reported that there is almost no difference between the local and average values of $\partial S^w/\partial t$ for air-water systems in a domain of 9 cm in length. Moreover, based on numerical simulations in air-water system, Hou et al. (2014) found that the local values could be approximated by the average values, even for a 15 cm long sand column.

The determination of dynamic capillarity effect and the calculation of $\tau$ require high temporal resolution of the measurement of saturation and water pressure. When saturation is determined from volumetric measurements, this will not be a limitation. But when TDR sensors are used, the response time of sensors has to be properly taken into account. The same applies for the response time of pressure transducers (see, e.g., Hou et al., 2012). We determined the response time of our pressure transducers to be 0.4 s (see Appendix A), which is comparable to the ones used by Bottero et al. (2011). The measuring time was set to 0.5 s, which is quite smaller than the recording time in other experimental studies performed in longer sand columns (Bottero et al., 2011; Camps-Roach et al., 2010; Sakaki et al., 2010). Hou et al. (2012) reported dynamic drainage experiments performed in a small sand column with the length of 1.27 cm. However, the values of $\tau$ reported in their work are at least one order of magnitude smaller than those in our study. This can be explained by

<table>
<thead>
<tr>
<th>Processes</th>
<th>$A$</th>
<th>$S^0$</th>
<th>$\tau^0$ (Pa s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Primary drainage</td>
<td>9.4</td>
<td>0.85</td>
<td>$2 \times 10^4$</td>
</tr>
<tr>
<td>Main drainage</td>
<td>9.4</td>
<td>0.73</td>
<td>$2 \times 10^4$</td>
</tr>
<tr>
<td>Scanning drainage</td>
<td>9.4</td>
<td>0.53</td>
<td>$2 \times 10^4$</td>
</tr>
</tbody>
</table>
the differences in sand properties and setup scales in the two experiments.

5. Summary and Conclusions

A series of drainage experiments were carried out to quantify the dynamic capillarity effect in an unsaturated soil. Primary, main, and scanning drainage experiments were performed under both quasi-static and dynamic conditions. The value of the dynamic capillarity coefficient $\tau$ was calculated based on the values of average air-water pressure differences and average saturation during static and dynamic drainage experiments.

A similar dependence between $\tau$ and saturation was found for primary, main, and scanning drainage. With the decrease in saturation, the value of $\tau$ varied slightly at moderate saturation levels, but then increased nearly log-linearly. At any given saturation, the value of $\tau$ for primary drainage was larger than the value for main drainage and that is in turn larger than the value for scanning drainage. This implies that the relationship between $\tau$ and saturation is nonunique. This non-unique behavior in the value of $\tau$ has rarely been reported before.

This may also suggest that $\tau$ is not only a function of saturation but a function of some other variables. The value of $\tau$ in our study was found to vary from $2 \times 10^4$ to $6 \times 10^6$ Pa s for primary and main drainage, consistent with reported values for air-water systems.

Appendix A: Determining the Response Time of Pressure Transducers

Preliminary tests were conducted to verify the response time of the pressure transducers. A schematic view of the test setup is shown in Figure A1. The idea was to monitor the readings of the pressure transducer when a sudden pressure difference was applied. The duration to reach stable readings of the pressure transducer was considered be the response time. To do so, a Plexiglas column (length = 50 cm and ID = 5 cm) was filled with degassed water. The top of the column was open to air. The pressure transducer was connected to the bottom of the column through a three-way valve. The details of the connection between the transducer and the column are shown as the insets of Figure A1. The pressure transducer was connected to the valve via a short tube filled with water. The same hydrophilic membrane that was used in our experimental setup placed at the opening of the tube. Readings of the pressure transducer were recorded every 0.1 s. At the start of the test, the valve was set to be connecting the transducer tube to the air. The reading of the transducer was taken as the reference pressure. Then, the valve was set to connect the transducer to the column. The transducer reading reached a table and constant value within 0.4 s. It corresponded to the height of the water column exactly.

Appendix B: Dynamic Primary Drainage Starting at Full Initial Saturation

As mentioned in the main text, we did not start the dynamic primary drainage experiments at full saturation. This was due to a mismatch between saturation and pressure measurements inherent to our experimental setup as explained below.

In fact, we did perform a dynamic primary drainage experiment, starting with fully saturated soil. The hanging water column was lowered...
to 40 cm below the bottom of the sandbox. Data on local water pressures and average water saturation were collected every 0.5 s. The local values of difference between air and water pressure, as measured by the two transducers, are plotted as a function of average saturation in Figure B1. Upon opening of Valve 3, the pressure transducers, which were showing hydrostatic pressure distribution, reacted immediately. First, water pressure decreased quickly and reached a value close to —40 cm; see data point 1 in Figure B1. Note that in this figure we are showing air-water pressure difference and not capillary pressure. In fact, at this point, while average saturation is less 100% (as water had started to leave the soil sample from below), but the soil in front of the pressure transducers remained saturated, as it took time for the desaturation front to arrive there. Then, water pressure at transducers started to increase (thus air-water pressure difference decreased) until the desaturation front reached the transducers (point 2 in Figure B1). As soon as the soil became unsaturated, water pressure started to decrease again (i.e., air-water pressure difference increased due to the dynamic capillarity effect).

If we use these measured values of air-water pressure difference under dynamic conditions and the quasi-static capillary pressure curve, we find negative values for $\tau$. That is of course not meaningful. Therefore, we have chosen for first creating unsaturated conditions in the soil quasi-statically, i.e., following the primary drainage capillary pressure curve until point 3 (that was an average saturation of about 0.83 and then start the dynamic drainage. This is actually pseudoprymary dynamic drainage; nevertheless it is primary dynamic drainage.

**Acknowledgments**

We gratefully acknowledge Thom Claessen and Sander Deelen of Utrecht University for their technical support. The first author is supported by China Scholarship Council (201206380076). The second author has received funding from the European Research Council under the European Union’s Seventh Framework Program (FP/2007–2013)/ERC grant agreement 341225. We gratefully acknowledge funding from the European Research Council. The second author has received a Marie Curie fellowship from the European Union’s Seventh Framework Program (FP/2007–2013)/ERC grant agreement 227671. The first author is supported by China Scholarship Council (201206380076). We gratefully acknowledge constructive comments by Associate Editor Markus Hilpert and three anonymous reviewers, which helped to improve the manuscript. The data from this work are available at https://doi.org/10.6084/m9.figshare.5414137.

**References**


