2013

Direct observation of the capillary mechanisms of liquid-liquid entrapment

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Direct observation of the capillary mechanisms of liquid-liquid entrapment and mobilization

by

William Gerard Gaul

A dissertation submitted to the graduate faculty in partial fulfillment of the requirements for the degree of

DOCTOR OF PHILOSOPHY

Major: Chemical Engineering

Program of Study Committee:
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Iowa State University
Ames, Iowa
2013

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DEDICATION

I dedicate this dissertation to my parents, Ted & Elaine Gaul. They have provided me with endless opportunities in my life. They have helped me to understand the importance of having a goal and working toward it through completion. They have been with me when things get bad and stood by me until I could stand alone. I want to show them they are the influence in my life that never fails.
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ABSTRACT

The dissertation develops further insight into the behavior of liquid-liquid flow in porous media. This topic is of great importance with applications in chemical reaction engineering, enhanced oil recovery, and environmental remediation. This work focuses on the experimental observation of invading immiscible drops, the long term behavior of their interface, and their entrapment via capillary pressure and mobilization through vibratory stimulation.

Using a simplified capillary physics mechanism several specific predictions can be made. So far these theories have generally agreed with CFD simulations, however a direct comparison to physical experiments is needed.

Experiments were conducted using single pore glass capillaries. Straight capillaries were used to study the film thickness left by an invading drop, which is critical for determining realistic initial conditions for the interface theory. A range of constricted capillaries were used in verifying the “breakup criterion” was influenced only by the geometric parameters of the capillary and not flow regimes. Additionally, the experiments and theory demonstrates qualitative agreement with respect to the evolution of the liquid-liquid interface. Finally, the theory of vibratory mobilization was verified by trapping
single drops with a pressure below their mobilization threshold and observing the drop’s mobilization over several oscillations.

The level of agreement between experiment, theory and CFD demonstrates the utility of the capillary physics mechanism in terms of reducing computational costs. The next goal is extending the theory to networks of pores so we can further our understanding of the oil mobilization and related processes, leading to useful technical applications.
CHAPTER 1. INTRODUCTION

1.1 Introduction

Understanding multi-phase flow and transport in porous media is of great importance with applications in chemical reaction engineering, enhanced oil recovery, and environmental remediation. In the field of reaction engineering a packed bed of solid catalyst particles, known as a trickle bed reactor, is used increase the surface area in order to more effectively dissolve an upward flowing gas in a downward flowing liquid. The dissolved gas subsequently makes contact with the solid catalyst where the desired reaction occurs. In petroleum engineering an enhanced oil recovery (EOR) method called waterflooding involves displacing the immiscible oil with water within an oil reservoir. A related example is encountered in groundwater where organic solvents leak into an aquifer from underground storage tanks, pipelines, surface spills and agricultural runoff. Many of these chemicals behave similar to oil, with many similar recovery methods. The solubility of these non-aqueous phase liquids (NAPLs) is very low, which preclude simple remediation methods like “pump and treat”, but the saturation concentrations far exceed safety limits which make the development of advanced groundwater remediation methods an environmental imperative.
A common phenomenon of multi-phase flow in porous media is the capillary entrapment of the non-aqueous phase (Fig. 1.1). A number of solutions for overcoming the capillary resistance and mobilizing the organic phase have been used, including water flooding (static pressure gradient), adding surfactants to lower the surface tension, increasing solubility with dispersants, electro-osmosis, biological remediation, and the use of seismic waves (Li et al., 2005). Beresnev and Johnson (1994) showed that the effect of vibration on entrapped fluids is well documented but poorly understood. So, understanding and predicting the motion of two-phase immiscible fluids in porous media, especially under the effect of vibration, is a practical, important, and challenging problem.

Therefore the basic physics behind this mobilization phenomenon needs to be better understood in terms of a pore-scale mechanism that describes the behavior of the entrapped ganglia in porous channels. Our project is devoted to furthering the development of the physical foundations of the vibratory EOR method. With this objective, the problems that we will approach are primarily single-pore, two-phase-flow problems, which become the focus of this research.

1.2 Background

The first step in understanding two-phase flow in porous media is recognizing that capillary forces are dominant over viscous forces. This is represented by the dimensionless quantity called the capillary number, $Ca \equiv \frac{\mu V}{\sigma}$, which is generally small ($Ca \ll 1$). Fairbrother and Stubbs (1935) found a relationship between the thickness of the film left
by a liquid displaced by a passing air bubble and the capillary number. This $\frac{1}{2}$ power law was further investigated by Taylor (1961) and found to be valid for small capillary numbers, while the film thickness approaches an asymptotic value for large capillary numbers ($Ca > 1$). Bretherton (1961) developed a separate theory resulting in a $\frac{2}{3}$ power law. Though derived for inviscid gas bubbles, this relationship was thought to hold for liquid-liquid flow as well because of the assumption of small capillary numbers. However, there is a great deal of disagreement between Bretherton’s theory and experimental observation of liquid drops. Since the long term behavior of the annular film is highly nonlinear and dependent on its initial condition (Hammond, 1983), a reliable theory to describe the thickness of a film left by an invading liquid drop, incorporating the viscosities of both fluids, is needed.

Since porous media can be fairly approximated as a capillary with a radius that varies in a quasi-periodic manner (Dullien, 1975), much research has been done to try to understand the physics of NAPL flow in sinusoidally varying constricted channels. Gardescu (1930) used the Young-Laplace equation to bubbles trapped in a capillary with a reduced opening to determine the force required to push the bubble through the constriction. Roof (1970) established the fact that the pore geometry was the controlling factor breakup of suspended drops. Despite this knowledge, several studies (Olbracht and Leal, 1983; Gaulglitz and Radke, 1990; Tsai and Miksis, 1994; Hemmat and Borhan, 1996) attempt to characterize the flow conditions under which breakup may occur. While they certainly observed the effects of the fluid property’s effect on mobilization and breakup, they could not reconcile the exact system parameters that led to or prevented breakup.
Beresnev et al. (2009) proposed a capillary mechanism to describe the conditions of a non-wetting drop in a sinusoidal tube. Under the assumption that the drop is large enough, it will take the shape of the channel with a thin aqueous layer \( t \ll r_{\text{min}} \). Under these conditions, the curvature and capillary pressures can be determined. The extrema of the capillary pressure naturally occur at the crest and trough of the channel. The condition that leads to drop breakup is that the capillary pressure must be greater in the trough than at the crest (Roof, 1970). The equation for a sinusoidally varying channel with minimum and maximum radii \( r_{\text{min}} \) and \( r_{\text{max}} \) and wavelength \( \lambda = 2L \) is

\[
r(z) = r_{\text{max}} \left\{ 1 + \frac{1}{2} \left( \frac{r_{\text{min}}}{r_{\text{max}}} - 1 \right) \left[ 1 + \cos \left( \frac{\pi z}{L} \right) \right] \right\}
\]

Applying the Roof condition leads to a 'static criterion' (Beresnev et al., 2009) for breakup.

\[
\lambda > 2\pi \sqrt{r_{\text{min}}r_{\text{max}}}
\]

If this condition is met, the drop will eventually breakup. It is important to note that the criterion is controlled only by the channel geometry. The criterion is well supported by published observations for small capillary numbers \( Ca \equiv \mu v / \sigma < 1 \), see Table 1.1.

Recalling that the NAPL is trapped due to capillary pressure and that water pressure is used to overcome it (Gardescu, 1930), how can more force be applied to the drop when increased water flow rates no longer increase the pressure due to geological bypass? Vibration allows the addition of a periodic body force \( F_{\text{osc}} = \rho_{\text{fluid}} a_{\text{wall}} \). As long as the negative phase of the vibratory forcing does not enter the flow region in the opposite direction, the drop will be nudged through the constriction 1.2, usually over a few cycles.
The effects of elastic waves were first noticed when it was found that well water level and oil production rose after seismic events. This and many other studies were compiled by Beresnev and Johnson (1994), which covered a wide range of frequencies, amplitudes, and results. The general agreement was that elastic waves increased the permeability of the oil, but there was no agreement on its physical cause. Early studies focused on ultrasound frequencies, which proved affective in mobilizing trapped NAPL, but required high intensity and were effective only over a short range (20cm, Roberts et al., 2001). Rather it was shown that trichloroethylene (TCE), an industrial solvent and common pollutant, effluent concentrations could be increased by as much as 20 times with frequencies lower than 100Hz. This led to further research using low frequency vibrations to enhance NAPL flow. Fontenot and Vigil (2002) studied the effects of low frequency pulsations in a three-dimensional micro-model. It was found that NAPL removal effectiveness was inversely proportional to the frequency. Li et al. (2005) confirmed the frequency relationship and determined that the NAPL liberation rate increased with increasing vibrational amplitude up to a point where the rate is limited by the aqueous phase flow. However, these studies observed networks of capillary constrictions. Single pore studies are needed to investigate the fundamentals behind vibratory forcing.

1.3 Objectives

To better understand the mechanisms of vibration on two-phase flow, we need to combine the knowledge and resolve the fundamental problems from the previous research mentioned above. Our work will focus on the behavior of an invading oil ganglia including
break-up, entrapment, and mobilization by vibratory stimulation. We will investigate the behavior of two-phase flow in straight and sinusoidally constricted tubes. Theoretical and numerical approaches were employed together to study the complex effect of vibration on oil-water flow in parallel with the experiments (Beresnev and Deng, 2010a,b).

The experiments are classified into three phases, each building upon the previous. The first is a repeat of studies by Fairbrother and Stubbs (1935); Taylor (1961); Bretherenton (1961), where a bubble was allowed to pass through a liquid filled capillary and the resulting film was measured. The experiment was modified to employ liquid drops rather than gas bubbles. The importance of this experiment is that though there is extensive data on films deposited by gas bubbles, little has been studied on liquid drops. The results were necessary in verifying a film theory that is critical in the numerical simulations of more complex flows. The second phase is much like the first except the capillary now has a sinusoidal constriction. The behavior is much more complex than with the straight capillary (Olbricht and Leal, 1983; Gaulglitz and Radke, 1990; Hemmat and Borhan, 1996). The system parameters were controlled to investigate the effect of capillary geometry and capillary number on the breakup of the liquid drop. The final phase involves the capillary entrapment and mobilization of an immiscible drop using vibratory stimulation.

1.4 Experimental Design

In designing the experiment several choices need to be made regarding which two fluids to use. The two fluids must have identical densities to minimize the effect of
buoyancy on the core fluid’s shape (axial symmetry), be immiscible and have a reasonably high interfacial surface tension as this increases the entrapment pressure to measurable levels. Additionally, the annular fluid must preferentially (with respect to the core fluid) wet the wall of the capillary, ideally perfect wetting. The most natural choice is to use an organic compound (a NAPL) for the core fluid and an aqueous solution for the annular fluid as it broadly satisfies the criteria and is analogous to real world scenarios such as EOR or groundwater remediation. It then comes down to which fluid is modified to match the density of the other.

Initially it was decided to use fluids similar to a previous researcher (Fontenot and Vigil, 2002). The aim in their experiment was to match refractive indices of both fluids as well as the glass support structure (analogous to our glass capillary tubes) since optical distortion would prevent any meaningful data gathering in a 3D model. However buoyant, rather than pressure driven, flow was used, so the densities of the two fluids did not match. In our experiment it was decided that the refractive index of the core phase need not match as this was an axisymmetric model and that the core phase would be effectively opaque as it would be dyed for visualization. So the density of the core fluid was matched to the annular fluid using a similar but slightly more dense organic liquid than used by Fontenot and Vigil. This choice of fluids proved to be problematic, however. Experimentally, the fluids had a fairly low surface tension, but in a practical sense the use of thiocyanate was a bad choice. It slowly damaged the seals on the experimental apparatus and corroded any metal it touched. A better fluid was needed.
Glycerol made an excellent replacement. It not only matched the refractive index of the capillary tube but also had a much higher interfacial surface tension with the NAPL. Additionally, glycerol was used to surround the capillary tubes after being placed inside of a square viewing box to remove optical distortion caused by the curved nature of the capillary tubes (Fig. 2.2). Glycerol’s high viscosity can be modified by diluting it with water, allowing for a broad range of capillary numbers to be studied with just one pair of fluid mixtures. This dilution changes the refractive index of the annular (film) fluid, but analysis shows that effect to be negligible (Appendix A). The fluid properties were measured with a falling ball viscometer and a DuNuoy tensiometer and are reported in 2.3 and Table 2.2.

The final requirement was that the annular fluid be “perfect wetting” with respect to the capillary wall. With real fluids this ideal is unworkable. However, a cleaning treatment was developed that made the glass capillaries much more preferentially wetting toward the aqueous phase and much less wetting with respect to the core phase. Soaking the glass capillaries in a 1:1 ratio of saturated solution of sodium hydroxide and anhydrous ethanol for 1 minute prior to their first use, or 15 seconds after subsequent uses, serves to both thoroughly clean the capillaries and render them near perfect water wetting. Caution is needed when using this solution as it is highly caustic, even to glass over extended periods, so it should be made fresh (very little solution is needed, <50mL) and disposed of properly after being used.
The various glass capillaries themselves were manufactured by a skilled glassblower to fit in the 10 cm long viewing box. This limited the constricted capillaries to wavelengths of approximately that length. Measurements of the tube’s internal radius were made via photography and a sinusoidal function was be fitted as in Gauglitz and Radke (1988) to match the tube’s profile. In all but the shortest wavelength capillaries, the most difficult to manufacture, the constrictions very nearly match an ideal sinusoid.

Finally, all of the experiments are carried out with high speed video with up to $11 \times$ magnification. This allows for detailed phenomena to be captured and the results can be compared directly to the theoretical and numerical simulations. The high magnification and high speed video require significant lighting. The experimental rig was back-lighted using a fiber optic halogen light source so as to keep the significant heat from the lamp from altering the physical properties of the fluids used.

1.5 Dissertation Organization

This dissertation is organized into a series of chapters consisting of three published papers. Chapter 2 discusses liquid-liquid, core-annular flow and the effect that the capillary number ($Ca$) has on the thickness of wetting fluid left by an invading drop. The theory developed generalizes previous theories to include viscous forces in both fluids. Chapter 3 deals with constricted capillaries and the effect of capillary geometry on the “snap-off” phenomenon. Chapter 4 revisits the capillary entrapment behavior and demonstrates the pore level mobilization via vibration. Finally, Chapter 5 draws conclusions about the importance of the work performed and makes recommendations for future work.
Figure 1.1  Capillary entrapment under an external pressure gradient.

Table 1.1  Analysis of published data with static breakup criterion.

<table>
<thead>
<tr>
<th>Study</th>
<th>Type of Study</th>
<th>$\frac{\lambda}{2\pi\sqrt{r_{\text{min}}r_{\text{max}}}}$</th>
<th>Static Criterion Satisfied?</th>
<th>Snap-off Observed?</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tsai and Miksis (1994)</td>
<td>Numerical model</td>
<td>0.38, 0.45, 0.50, 1.0</td>
<td>No</td>
<td>No at small Ca</td>
</tr>
<tr>
<td>Hemmat and Borhan (1996)</td>
<td>Experiment</td>
<td>1.3</td>
<td>Yes</td>
<td>Yes</td>
</tr>
<tr>
<td>Olbricht and Leal (1983)</td>
<td>Experiment</td>
<td>0.71</td>
<td>No</td>
<td>No at small Ca</td>
</tr>
<tr>
<td>Martinez and Udell (1989)</td>
<td>Numerical model</td>
<td>0.71, 0.92</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>Gaulglitz and Radke (1990)</td>
<td>Numerical model and experiment</td>
<td>4.5, 5.7, 7.1</td>
<td>Yes</td>
<td>Yes</td>
</tr>
</tbody>
</table>
Figure 1.2 Mobilization under an external pressure gradient using elastic waves.
CHAPTER 2. THICKNESS OF RESIDUAL WETTING FILM IN LIQUID-LIQUID DISPLACEMENT

A paper published in Physical Review E

Igor Beresnev, William Gaul, and R. Dennis Vigil

Abstract

Core-annular flow is common in nature, representing, for example, how streams of oil, surrounded by water, move in petroleum reservoirs. Oil, typically a non-wetting fluid, tends to occupy the middle (core) part of a channel, while water forms a surrounding wall-wetting film. What is the thickness of the wetting film? A classic theory has been in existence for nearly 50 years offering a solution, although in a controversial manner, for moving gas bubbles. On the other hand, an acceptable, experimentally verified theory for a body of one liquid flowing in another has not been available. Here we develop a hydrodynamic, testable theory providing an explicit relationship between the thickness of the wetting film and fluid properties for a blob of one fluid moving in another, with neither phase being gas. In its relationship to the capillary number $Ca$, the thickness of the film is predicted to be proportional to $Ca^2$ at lower $Ca$ and to level off at a constant value of 20% the channel radius at higher $Ca$. The thickness of the film is deduced
to be approximately unaffected by the viscosity ratio of the fluids. We have conducted our own laboratory experiments and compiled experimental data from other studies, all of which are mutually consistent and confirm the salient features of the theory. At the same time, the classic law, originally deduced for films surrounding moving gas bubbles but often believed to hold for liquids as well, fails to explain the observations.

2.1 The Problem

Consider a straight cylindrical capillary channel of small radius filled with a wetting fluid. Subsequently another, non-wetting, fluid begins to flow into the same capillary. This is the scenario of an invasion “core-annular” flow. The non-wetting fluid will tend to occupy the middle part of the channel (the “core”), and the one being displaced will stay adjacent to the channel’s wall as a residual wetting film. How thick is this annular film?

The phenomenon is common in nature. For example, it has long been suggested as a scenario for two-phase immiscible fluid motion through natural porous channels in petroleum reservoirs; it is thus crucial for understanding the oil recovery. Despite the apparent simplicity, no adequate, experimentally tested theoretical solutions for the thickness of the wetting film in liquid-liquid displacement, as a function of fluid parameters, have been proposed. The difficulty stems from the fact that the core-annular flow is a “free-boundary” problem, in which the fluid/fluid interface is not fixed but moves and deforms with the flow, with its configuration having to be obtained as part of the solution. Free-boundary fluid-mechanics problems are notoriously difficult to solve (e.g.,
The phenomenon is more easily theoretically described, with some assumptions, if the invading phase is gas. A classic theory of the thickness of an annular film left behind by a retreating pore-filling fluid driven by gas (e.g., air) in the limit of low capillary numbers $Ca$, was developed by Bretherton (1961). The theory predicted that the thickness of the wetting film, normalized by the channel radius (this dimensionless thickness will be denoted $b$ in the following) has a power-law relationship with the capillary number $Ca$: $b = 1.34Ca^{2/3}$ (Bretherton, 1961, Eq. 17). Although the predicted functional form has not truly withstood experimental verification, plausible theoretical explanations for its observed departure from at least a portion of experiments have been offered (see a synopsis of relevant studies in Table 2.1). Bretherton himself found a limited agreement with his experimental data, while noticing the dependence of the observed behavior on a particular wetting fluid used and contradiction of his data with earlier experiments of others (Bretherton, 1961; Fig. 4). Similar experiments repeated a quarter-century later by Schwartz et al. (1986, Fig. 3) found a dependence of the residual film thickness and the exponent of the power law on bubble’s length, which signaled further departure from Bretherton’s predicted behavior. There is general belief that the deviations noted by Bretherton; Schwartz et al. in the capillary-number range of $10^{-5}$–$10^{-2}$ are caused by the presence of trace amounts of surfactants and the resulting Marangoni surface flows in the fluids used. For example, Ratulowski and Chang (1990) found that an ad hoc combination of parameters controlling the convective, diffusive, and adsorption effects in a low-concentration surfactant solution (their “convective-equilibrium” asymptotics)
is capable of explaining the scatter of experimental relationships reported by Bretherton and Schwartz et al. (see Ratulowski and Chang, 1990, Fig. 8). In effect, the free, not experimentally constrained parameters of the model were adjusted to produce a fit to the data. An ad-hoc, albeit plausible, character of this explanation is somewhat unsatisfactory, since there was no guarantee that the required parameter regime was indeed realized in the experiments. Also note that Ratulowski and Chang (1990) did not explain the smaller exponent of the power law of $\frac{1}{2}$ found by Fairbrother and Stubbs (1935), which was found consistent with the measurements of Taylor (1961). Furthermore, Quéré (1999) shows that Ratulowski and Chang’s predictions are not necessarily compatible with fluid-coating observations. It can be further argued that the departure of Bretherton’s theory from experiments might alternatively be explained by the oversimplifications made in the theory, as discussed in a later section in this article.

Despite the not completely satisfactory experimental verification, Bretherton’s theory is generally believed to be accurate in its appropriate capillary-number range of $10^{-5}$–$10^{-2}$, found by Ratulowski and Chang (1990), and has been used extensively in the calculation of thicknesses of wetting films left behind by invading gas bubbles in capillary channels.

When the invading phase is gas, with some reservations, this usage is therefore still acceptable. However, Bretherton’s theory has often been assumed to equally apply to the films left behind by one fluid displacing another where neither phase is gas (e.g., Peña et al., 2009, pp. 1998). Park and Homsy (1984, Eq. 4.27) followed Bretherton’s
theoretical argument and arrived at an identical solution, although the original theory was limited exclusively to the films surrounding moving gas bubbles. Does such a suggestion stand experimental test? Experimental evidence has been scarce, controversial, and not necessarily supportive; we will address limited available data in a later section. Alternative hydrodynamic theories, which could be better compatible with the data, have never been explored. The questions, therefore, are still outstanding: are film thicknesses, deposited on capillary walls, the same for gas bubbles moving in liquids and for bodies of liquids moving in liquids? Is there sufficient theoretical and experimental evidence suggesting that Bretherton’s law necessarily applies to the liquid/liquid invasion? The answers have remained largely unclear. In this paper, we develop a hydrodynamic theory applying to the case of liquid/liquid displacement and test it in a direct laboratory experiment. We also examine existing experimental data in the context of present results. The conclusion, supported by both the theory and experiment, is that Bretherton’s power law does not apply to liquid-liquid invasion.

2.2 Theory

Our theoretical problem deals with core-annular, incompressible, axisymmetric Poiseuillean flow, which allows analytical solution of the Navier-Stokes equation. Hence, it is assumed that the Reynolds number is small. When the interface between the core and the annulus deforms, pressure gradients in each phase will generally be different because of the curvature of the interface according to Laplace’s law of capillary pressure. We thus need to begin with a general solution for two-phase Poiseuillean flow in axisymmetric
geometry with non-equal pressure gradients in both phases (Beresnev and Deng, 2010b).

The boundary conditions to satisfy are no-slip at the channel wall, continuity of velocity at the fluid-fluid interface, and continuity of shear stress at this interface.

In a typical capillary geometry, Bond numbers, expressing the ratio of gravitational to capillary forces, tend to be small \((10^{-2}–10^{-3})\) (Hammond, 1983; Beresnev and Deng, 2010b). It is known that core-annular flow with gravity effects can maintain steady state but acquire axial eccentricity (Ooms et al., 2007). We assume the smallness of Bond number, which allows us to neglect gravity and use an axisymmetric geometry.

It is convenient to carry out the analysis in non-dimensional variables. If we designate by asterisks the variables that have dimensions and drop asterisks for their non-dimensional counterparts, the latter are introduced as follows,

\[
\begin{align*}
  r^* &= \frac{r}{R^*}, \\
  x^* &= \frac{x}{R^*}, \\
  \tau^* &= \frac{\tau^*}{\mu_1^* R^*/\sigma^*}, \\
  p^* &= \frac{p^*}{\sigma^* R^*/\mu_1^*}, \\
  Q^* &= \frac{Q^*}{\sigma^* R^2/\mu_1^*}.
\end{align*}
\]

Here \(r^*\) and \(x^*\) are the radial and axial coordinates, respectively; \(t^*\) is the time, \(p^*\) is the pressure, \(Q^*\) is the volumetric flow rate, \(R^*\) is the radius of the tube, \(\sigma^*\) is the interfacial tension, and \(\mu_1^*\) is the core dynamic viscosity. The subscripts “1” and “2” denote the variables in the core and the suspending (wetting) fluids, respectively.

The boundary conditions resolve the axial-velocity profiles \(u_1^*(r^*, p_{1x}^*, p_{2x}^*)\) and \(u_2^*(r^*, p_{1x}^*, p_{2x}^*)\) in the core and the film, respectively, through the pressure gradients \(p_{1x}^*\) and \(p_{2x}^*\), where the subscript \(x^*\) indicates the derivative with respect to \(x^*\). These solutions are given by Beresnev and Deng (2010b, Eq. 6), and, for the sake of brevity, we do not reproduce them here. Integration of these profiles over the cross-sections of the core and the annu-
lus give volume fluxes $Q_1(p_{1x}^*, p_{2x}^*)$ and $Q_2(p_{1x}^*, p_{2x}^*)$ (Beresnev and Deng, 2010b, Eq. 7).

After nondimensionalization, for the straight tube they become

$$Q_1(p_{1x}, p_{2x}) = -\frac{\pi}{2} \kappa^4 \left[ \frac{p_{1x}}{4} + \frac{\mu_1^*}{2\mu_2^*} \left( \frac{1}{\kappa^2} - 1 \right) p_{2x} - \frac{\mu_1^*}{\mu_1^*} (p_{1x} - p_{2x}) \ln \kappa \right],$$

$$Q_2(p_{1x}, p_{2x}) = -\frac{\pi}{2} \frac{\mu_1^*}{\mu_1^*} \kappa^4 \left[ \frac{p_{2x}}{4} \left( \frac{1}{\kappa^2} - 1 \right) + \frac{1}{\kappa^2} (p_{1x} - p_{2x}) \left( \frac{1}{2} - \kappa^2 \ln \frac{\sqrt{\kappa}}{\kappa} \right) \right],$$

(2.1)

where $\kappa$ is the radial position of the fluid-fluid interface normalized by $R^*$. The volume fluxes (Equation 2.1) still contain the unknown pressure gradients; however, Laplace’s law supplies another (non-dimensionalized) equation relating $p_1$ and $p_2$: $p_1 - p_2 = \frac{1}{\kappa} - \kappa_{xx}$, where the right-hand side is the dimensionless mean curvature of the interface (Gauglitz and Radke, 1988, Eq. 7); (Beresnev et al., 2009, Eq. 2, A5-A7).

Here we used the assumption of a “small slope” of the interface, which allowed us to neglect the terms $\kappa_x^2$ with respect to unity in the full expression for the curvature. The same approach was taken by Bretherton (1961, Eq. 4), allowing the use of the solutions for Poiseuillean flow. Note, also, that the use of Laplace’s law has implied the smallness of the capillary number, which characterizes the ratio of viscous to capillary forces. We therefore neglect the viscous normal stresses in the moving fluid.

The last equation then leads to $p_{1x} - p_{2x} = -\kappa_{xx} \kappa_x^2$. If we combine it with the conservation-of-mass condition $Q_1(p_{1x}, p_{2x}) + Q_2(p_{1x}, p_{2x}) = Q$, where $Q$ is the total volume flux through the tube, the two equations form the system from which both $p_{1x}$ and $p_{2x}$ can be resolved. When substituted them back into $Q_1$ in Equation 2.1, this leads to an explicit formula
\[ Q_1 = \frac{\mu_1^*}{\mu_2^*} \frac{Q}{\kappa_2} \left[ 1 + 2 \frac{\mu_2^*}{\mu_2^*} \left( \frac{1}{\kappa^2} - 1 \right) \right] - \frac{\pi}{2} \kappa^2 \left( \kappa_x + \kappa_x \kappa_{xxx} \right) \left\{ \frac{\left( \frac{1}{\kappa^2} - 1 \right) \left[ \left( \frac{\mu_2^*}{\mu_2^*} - \frac{1}{4} \right) \frac{1}{\kappa^2} + \frac{3}{4} - \frac{\mu_1^*}{\mu_2^*} \right]}{\frac{1}{\kappa^4} + \frac{\mu_2^*}{\mu_1^*} - 1} + \frac{\mu_1^*}{\mu_2^*} \ln \kappa \right\} \right. \\
\left(2.2\right)
\]

\( Q_1 \) described by Equation 2.2 is the volume flux in the core fluid when it moves into the suspending phase that initially fills the capillary tube. Since we are looking for stationary solutions, the front meniscus, separating the two fluids, moves without changing its shape with constant speed \( U^* \). The geometry is illustrated in Figure 2.1. The core displaces part of the suspending fluid that is ahead of the moving meniscus, leaving part of it behind as a wetting film on the wall. The film thickness, normalized by \( R^* \), is \( b \).

In this geometry, conservation of mass requires that \( Q_1 \) be also equal to \( Q_1 = \pi U (1 - b)^2 \), which is the flow rate of the displaced fluid ahead of the invading core body (Goldsmith and Mason, 1963, Eq. 3), where the speed has been non-dimensionalized using \( U = \frac{U^*}{\sigma^*/\mu_1^*} \).

Note that the respective conservation-of-mass condition, used by Bretherton (1961, Eqs. 7 and 8) and reproduced by Middleman (1995, Eqs. 2-4.6 and 2-4.7), inconsistently neglects the flow in the film, whereas Goldsmith and Mason’s condition is more general and correct. The last formula, rewritten in terms of the capillary number, \( Ca = U^* \frac{\mu_1^*}{\sigma^*} = \frac{U^* \mu_2^*}{\mu_1^*} \), becomes \( Q_1 = \pi \frac{\mu_1^*}{\mu_2^*} Ca (1 - b)^2 \). Equating this expression for \( Q_1 \) with that from Equation 2.2 leads to the differential equation for the wetting-film profile \( \kappa(x) \).

The resulting equation is extremely complex and is by itself not very useful. We notice, however, that somewhere near the nose the moving front in Figure 2.1 has a round (but not necessarily spherical) shape. Following Bretherton (1961, discussion leading
from his Eq. 11 to Eq. 12) (also see Middleman, 1995, Eqs. 2-4.10 and 2-4.11), we seek a simplified form of Equation 2.2 somewhere in the transition zone between the nose of the invading core phase and the residual film, at small values of $\kappa$, by retaining only the important terms. At sufficiently small $\kappa$, constant terms can be neglected compared to $1/\kappa^2$, $1/\kappa^4$, and $\ln \kappa$. In the same approximation, we can neglect $2Q\kappa^2$ with respect to $Q_1$ and, assuming a bounded third derivative, $\kappa^2\kappa_{xxx}$ with respect to $\kappa_x$. This leads to a simpler, asymptotic form of the film-profile equation,

$$-\frac{1}{2}\kappa^2\kappa_x \ln \kappa = Ca(1 - b)^2.$$  

The transition to this form and the approximations made are equivalent to Bretherton’s numerical solution of his main film-profile equation (Eq. 11), assuming the asymptotic interface shape near the nose (Eq. 12), which led to the final equation (Eq. 17) (this transition is also discussed by Middleman, 1995, Eqs 2-5.41 — 2-5.45). Note that the viscosity ratio has dropped out. Rearranging, we write $\kappa_x = \frac{2Ca(1 - b)^2}{\kappa^2 \ln \kappa}$ and, differentiating one more time,

$$\kappa_{xx} = -\frac{8Ca^2(1 - b)^4}{\kappa^5 \ln^2 \kappa}$$  

Formula 2.3 is valid somewhere in the transition zone. In this region, the meniscus has a round shape, but, because the meniscus is moving, is not exactly a sphere. The curvature of the sphere would be $\frac{2}{1-b}$, and therefore a matching condition $\frac{1}{1-b} - \kappa_{xx} \approx \frac{2}{1-b}$, or $|\kappa_{xx}| \approx \frac{1}{1-b}$, will approximately hold (Bretherton, 1961, discussion leading from Eq. 12 to Eq. 13); (Middleman, 1995, Eqs. 2-4.12 — 2-4.13). To approximate the departure from the spherical shape, we add a correction to make the right-hand side equal $\frac{1}{1-b} + C_1$; the value of $C_1$ is to be determined. Using Equation 2.3 for $\kappa_{xx}$ in the result leads to the
equation for \( b \),

\[
b = \frac{C_2(1 + C_1) - 8Ca^2}{C_1C_2 - 40Ca^2}
\]

(2.4)

where we approximated \((1 - b)^5 \approx 1 - 5b\) and introduced a notation \( C_2 \equiv \kappa^5 \ln^2 \kappa \).

We are now able to constrain the value of \( C_1 \). At small capillary numbers, Equation 2.4 becomes \( b = \frac{1+C_1}{C_1} \). We immediately see that \( C_1 \) cannot be positive or zero, since \( b \), by definition, cannot be greater than one. Also, \( b \) must be non-negative, which further constraints \( C_1 \) to be \( C_1 \leq -1 \). Furthermore, the curvature in the right-hand side of the equation \( |\kappa_{xx}| = \frac{1}{1-b} + C_1 \) must be non-negative, \( \frac{1}{1-b} + C_1 \geq 0 \). For small values of \( b \), this leads to \( C_1 \geq -1 \). The only value of \( C_1 \) compatible with the last two conditions is \( C_1 = -1 \). Using this in Equation 2.4, we arrive at the final expression for the dimensionless thickness of the surrounding annulus, as a function of capillary number,

\[
b = \frac{8Ca^2}{C_2 + 40Ca^2}.
\]

(2.5)

Equation 2.5 still contains an unknown quantity \( C_2 \), which is the value of the function \( \kappa^5 \ln^2 \kappa \) at an unknown position in the transition region between the nose of the moving meniscus and the film. It cannot be too close to the nose, though, where this function has a mathematical limit of zero, because this would violate our assumption of the small slope of the fluid-fluid interface. All we can say at this point is that \( C_2 \) is small, but its exact value cannot be deduced from the theoretical argument alone.

We have two immediate, testable physical predictions from equation 2.5. At relatively large capillary numbers, the film thickness \( b \) levels off at a constant value of \( b = \).
0.2. At small capillary numbers, it becomes proportional to $Ca^2$, that is, has a constant slope of two if plotted on a log-log scale. The thickness dependence on the capillary number is therefore predicted to have a corner separating a sloping, lower-$Ca$, region from a constant, higher-$Ca$, region. The film thickness does not appear to depend on the viscosity ratio of the core and film fluids. These predictions can be tested against experiment; by fitting experimental data, the constant $C_2$ can be determined as well.

Theories of residual film thickness for liquid-liquid invasion scenarios in core-annular flow, developed earlier Schwartz et al. (1986); Teletzke et al. (1988) failed to propose a similar, closed-form and testable relationship.

We would like to emphasize here in what respects our theory deviates from Bretherton (1961). First, our theory is strictly applicable to the case of liquid-liquid displacement only. It assumes incompressibility of both phases and therefore is not expected to correctly describe the invasion of a gas bubble. A bubble is compressed in response to capillary pressure, but this effect is absent from the formulation of Laplace’s law that we have used. An augmented form of Laplace’s law, which would include the compressibility effect, would incorporate the volume of the bubble, resulting in the bubble’s size being controlled by the pressures in both phases (e.g., Gauglitz and Radke, 1989, pp. 233–234). This would make the problem even more complex. This is why our theory will only be compared to experimental results involving liquid-liquid displacement. Note that Bretherton (1961) also founded his theory on the same incompressible-fluid assumption, although he compared it with experiments involving gas bubbles.
Second, Bretherton’s derivation of his film-profile (Eq. 11), performed in the assumption of a “planar” interface (Bretherton, 1961, pp. 169), neglected the transverse-curvature term $\frac{1}{\kappa}$ (in our notation) in the expression for the mean curvature in Laplace’s law. This can be directly checked by re-deriving Bretherton’s Eq. 9 from his Eq. 7. We have retained this term, important for the rendition of the cylindrical geometry. Third, Bretherton’s analysis assumed a lubrication flow in the film but neglected it in the formulation of the mass balance underlying the ultimate film-thickness equation. We have chosen to use a different mass-balance formulation instead. Fourth, Bretherton made an assumption of constant, zero pressure inside the gas phase. This is also inaccurate. Bretherton (1961, pp. 169) assumes an inviscid bubble with zero tangential stress at the interface; therefore, there therefore no drag on the bubble in this model. In the absence of both the drag and an internal pressure gradient, a bubble would have no driving force to move. On the other hand, our approach uses the actual variable pressure in the core.

It is noteworthy in this regard that the validity of Bretherton’s relationship, for sufficiently small capillary numbers ($5 \times 10^{-5} – 10^{-2}$), was confirmed by direct computational-fluid-dynamics simulations (Giavedoni and Saita, 1997, Fig. 3). However, the authors’ model, similarly to Bretherton’s, postulated a constant zero pressure inside the moving inviscid gas bubble. It was therefore founded on the same approximation of the reality and cannot be considered a truly independent verification. Oversimplifications made in Bretherton’s theory may be an alternative reason for its departures from various experimental validations.
2.3 Experiment

An experimental apparatus was built to directly observe the displacement of the pore-filling suspending fluid by an invading liquid core phase (Figure 2.2). It consists of a glass capillary tube with the length of 100 mm and inner diameter of $1.14 \pm 0.05$ mm inside a transparent Lucite viewing box with square cross-section. The region between the viewing cell and the capillary tube is filled with glycerol to reduce optical distortion due to the curvature of the tube. Small ports at either end of the viewing cell provide access for feeding and removing the working fluids into or out of the capillary. At the inlet side (the right side of the picture) there are two ports: one for the annular fluid and one, covered with a rubber septum, for the injection of the core fluid. The viewing cell is illuminated from below using a fiber-optic light source and a diffuser. Images of the capillary tube are captured at a magnification of $(11.6 \pm 0.5) \times$ approximately 75 mm downstream from the entrance of the straight test section by using a reverse-mounted 28-mm Nikon lens and bellows connected to a Photron FASTCAM APX-RS high-speed digital camera mounted above the flow cell.

Aqueous solutions of glycerol were used as suspending fluids, as listed in Table 2.2. The core fluid was heptane dyed with Oil Blue N, with the viscosity of $3.9 \times 10^{-3} Pa \cdot s$. The fluid-fluid interfacial tension measured with a DuNouy tensiometer was $1.5 \times 10^{-2}$ N/m for all three glycerol/heptane systems studied.

To remove any organic residue and increase the hydrophilicity of the glass, the capillary was first cleaned in a NaOH-ethanol-deionized-water solution and then flushed
with deionized water and dried before being sealed in the viewing cell that coupled the tube with the flow system. In a given run, the capillary was first filled with the annular fluid, and the core phase was injected upstream of the test section via a syringe. The system was allowed to equilibrate, in such a way that there was no residual flow induced by the injection of the core fluid. A syringe pump then pumped the annular fluid through the capillary tube, and recording of images took place. The recording ended when the trailing meniscus exited the test section. The drop velocity $U$ could be controlled by changing the speed of the syringe pump.

The measurement of the film thickness in pixels was performed in a digital-image editor after the completion of the experiment. This thickness was converted to absolute units through the known optical magnification and the known camera-sensor’s absolute pixel size. The film thicknesses were obtained one-to-two tube diameters from the front meniscus, where the film reached a constant thickness (Figure 2.3 A and B). The thickness remained constant between this point and the trailing meniscus (Figure 2.3 C and D); the total drop length was approximately 16 tube diameters. Notice from Figure 2.3 that both the leading and the trailing menisci maintain constant shape as they pass through the tube.

2.3.1 Discussion

Our experimental data are tabulated in Table 2.4 in the rows without superscripts. The lower limit on the resolved film thickness is prescribed by the value of the magnification of the optical system. After magnification, the absolute value of one pixel is
1.46 $\mu$m, which, divided by the mean radius of the tube, gives $b = 2.55 \times 10^{-3}$ as the smallest value that we could resolve, corresponding to the thickness of one pixel. This is the smallest $b$ listed in Table 2.4 for our data. The experiments run at respectively lower capillary numbers led to the film “disappearance”, meaning that its thickness fell below the limit of resolution of one pixel.

A note is in order regarding the methods that various experimenters have used to report film thicknesses in either gas-liquid or liquid-liquid displacements. Bretherton (1961); Schwartz et al. (1986), who observed gas-bubble motion, employed a similar indirect approach, in which they observed a reduction in length of a moving liquid slug pushed by the gas phase. The film thickness was inferred from the length reduction over the distance traveled by the slug. This method can be argued to have a significant disadvantage of not seeing the film. For example, the authors have extended their experiments down to $Ca = 10^{-6}$ (Bretherton, 1961) and $10^{-5}$ (Schwartz et al., 1986). Using Bretherton’s relationship and the tube radii reported, the absolute film thicknesses at these capillary numbers are expected to be on the order of 0.1 and 1 $\mu$m, respectively. On the other hand, the roughness of the glass wall in the tubes, examined directly under an electron microscope in a related study by Chen (1986, pp. 344), was on the order of 1 $\mu$m. What is noteworthy is that both Bretherton (1961, Fig. 4) and Schwartz et al. (1986, Fig. 3) reported deviation of a constant-slope behavior (on the log-log scale) in the inferred film thickness toward leveling-off to zero slope around the same $Ca = 10^{-5}$. In either case, the theoretical film thickness at this capillary number is close to 1 $\mu$m, or the anticipated size of microscopic roughness of the surface. In such a situation, the amount
of wetting fluid left behind can be expected to become independent of the motion of the slug, as it should be entirely controlled by the fluid collecting in the hollows of the surface. A trend toward capillary-number-independent constant value of film thickness at very small $Ca$ was qualitatively predicted by Teletzke et al. (1988, Fig. 5) and attributed to molecular effects. However, a simpler explanation, based on the disappearance of a continuous film because of rough surface, seems to be more natural. On the other hand, if the film could be visually observed, this difficulty of resolving a “film” if none actually existed would not arise.

Chen (1986) reported film thicknesses for both gas- and liquid-liquid displacement using a different but also indirect method. The technique, borrowed from Marchessault and Mason (1960), consisted in measuring electrical resistance of a capillary containing the core and film fluids. The resistance was converted into film thickness using an idealized model of an electrical conductor composed of coaxial inner and outer cylinders. A disadvantage of this technique is its reliance on an idealized cylindrical geometry. An (unknown) increase in measured conductance is always contributed by the fluid collected in the hollows of the tube surface. This contribution becomes dominant when the size of the surface roughness is comparable to the expected thickness of the film. The inferred thickness is therefore always inaccurate and becomes virtually meaningless in the latter case. Calibration studies were not reported that could quantify the error.

Because of the increase in conductance contributed by the fluid in the roughness, the method is expected to always over-predict the true film thickness and measure a
“constant” thickness at capillary numbers at which the expected thickness is comparable to the roughness size. Because the amount of over-prediction grows with the decreasing capillary number, the method will also tend to diminish the true slope of the $b$-$Ca$ relationship. Chen (1986, Fig. 3) indeed over-predicts Bretherton’s theoretical film thickness at all capillary numbers and also reports a slope reduced compared with Bretherton’s relation. The “leveling-off” to a constant value around $Ca = 10^{-5}$ for gas invasion (which virtually coincides with the respective leveling-off $Ca$ values reported by Bretherton and Schwartz et al.) and $10^{-4}$ for liquid invasion is also reported. For the tube radius used, Bretherton’s relationship gives anticipated film thicknesses of approximately 0.2 and 1 $\mu$m, respectively. The depth of the hollows, estimated from electron microscopy, is 1 $\mu$m, too. The film over-prediction is evident from the fact that, for stationary air bubbles, Chen (1986, pp. 346) measured the film “thickness” of 0.7 $\mu$m, while it should theoretically be zero. An anomalous character of Chen’s data is also indicated by Ratulowski and Chang (1990, Fig. 8). While the authors were able to give a consistent theoretical fit, based on a single model, to the scatter of experimental results of Bretherton and Schwartz et al., they could not fit Chen’s constant-slope data within the same approach. Over-prediction with the electrical-resistance method is also demonstrated by the fact that Marchessault and Mason (1960) results (as compared by Bretherton, 1961, Fig. 4) consistently exceed Bretherton’s predicted film thickness at all capillary numbers as well.

For the reasons explained, we prefer to use only the data on film thicknesses in liquid-liquid invasion measured by direct film observation by optical means. Table 2.4 contains a compilation of such measurements in addition to ours; (Goldsmith and Mason,
1963, Fig. 3), (Aul and Olbricht, 1990, Table 2), (Soares et al., 2005, Figures 7, 8, 12, 13); identical rows mean availability of experiments with exactly same results. All data are graphically presented in Figure 2.4, where the values from various authors are indicated by different symbols. For our experiments, the average uncertainty of a film-thickness measurement is 0.5 pixel, which leads to the relative error of 50% when the thickness approaches the limit of resolution. This determines the value of the error bars for our results shown in Figure 2.4. Clearly, the errors grow as \( b \) decreases but are only visible, on the scale of the graph, at the three lowest values of the capillary number; in all other cases, the errors are smaller than the circle size.

The solid curve in Figure 2.4 is Equation 2.5 visually fit to all the data, with \( C_2 = 3 \times 10^{-4} \). Bretherton’s equation \( b = 1.34 \times Ca^{2/3} \) is plotted as the solid straight line. The combined experimental data unambiguously indicate the presence of a “corner” between \( Ca = 10^{-3} \) and \( 10^{-2} \), separating two general slopes in the observed relationship, which is missed by Bretherton’s single-slope theory. The low-\( Ca \) slope is correctly captured by the present theory, as well as the leveling off at \( b \approx 0.3 \) at the higher end of capillary number. The intermediate range is captured reasonably well, with deviations of no more than a factor of 2 to 3. Data compiled from different investigations are mutually consistent, in that no systematic deviation of points of one study from those of another is seen.

One corollary from the present theory is an approximate independence of the film thickness of the viscosity ratio. Figure 2.5 presents the same data points as in Figure 2.4, which are coded in groups of close viscosity ratios. No systematic separation of
one group from another is found, despite the fact that viscosity ratios vary by more
than three orders of magnitude. Points, different in viscosity ratio by this amount, in
certain instances overlap. This supports the theory’s conclusion, in the range of capillary
numbers tested.

The present theoretical derivation, as well as previous attempts to build such a
theory, seem to indicate that, due to complexity of the phenomenon, it is not possible
to deduce a simple relationship between the annular film thickness and $Ca$ that would
be able to match experimental data with better precision in the entire capillary-number
range.

### 2.4 Conclusions

A hydrodynamic theory of the thickness of a residual wetting film left behind during
the invasion of a core fluid into a capillary channel leads, asymptotically, to Equation
2.5. Notwithstanding the value of the constant $C_2$, the theory predicts two distinct
slopes in the dependence of $b$ on capillary number, separated by a “bend”. The lower-$Ca$
asymptotic behavior is $Ca^2$, while a constant level of $b = 0.2$ is predicted to be reached
at the higher end.

We collected laboratory measurements in which the values of $b$ for liquid-liquid
invasion have been reported using direct optical observation. Direct observation is the
most unambiguous method that does not infer the presence of deposited films but rather
measures them only if they indeed exist. We have also conducted our own laboratory
experiment. The data, obtained by different investigators, are mutually consistent, and the theoretically predicted behavior is seen in all the data.

The only conclusion about the constant $C_2$ in Equation 2.5, available within the theory, is that it is a small number. Based on experimental data, $C_2$ turns out to be 0.0003. With this value, the film thickness in the entire range of capillary numbers available from the experiments is matched reasonably well.

It follows from the theory that the film thickness, described by Equation 2.5, is roughly independent of the viscosity ratio between the core and film fluids. Data, coded according to viscosity ratio in Figure 2.4, do not show any significant scatter, with the viscosity ratios changing over three orders of magnitude. This supports the theory's conclusion. Computational-fluid-dynamics simulations by Soares and Thompson (2009, Fig. 6) reached the same conclusion for $Ca$ of less than approximately 0.2, while the authors computationally predicted thickening of the films with decreasing film-to-core viscosity ratios at greater capillary numbers. The data in Figure 2.5 do not show a clear trend toward such behavior, although it cannot be ruled out that it may reveal itself at higher capillary numbers.

The classic theory of Bretherton (1961), developed for the displacement of liquids by gas bubbles but often believed to describe the wetting films during liquid-liquid invasion, fails to capture the salient features of the experimental relationship.
Hodges et al. (2004) proposed an asymptotic theory, seeking the dimensionless film thickness $b$ in the form $b = F(\mu_1^{*}/\mu_2^{*}, Ca) Ca^{2/3}$. The results are presented implicitly: the function $F$ is expressed in different asymptotic regimes defined by $b$ itself, offering no explicit, testable predictions for $b$. For all small core-to-film viscosity ratios, Bretherton’s relation is predicted to hold, which contradicts the experiments. Park and Homsy (1984, Eq. 4.27) also suggested that Bretherton’s result extended without change to liquid-liquid invasion. Their main evolution equation (4.15), on which this conclusion is based, has the same form as Bretherton’s equation (11), which allowed the authors to apply their theory to liquid-liquid invasion. However, Park and Homsy did not provide the derivation of this equation. If it is the same as Bretherton’s derivation, the latter was explicitly dependent on the assumption of constant zero pressure in the core (gas) phase. This is an oversimplification, which may explain why the suggested behavior is not seen in the experiments. To fit an experimentally observed $b-Ca$ relationship, we have used the actual variable pressure gradients in the core.

Overall, the experiments do not support the proposed extensions of Bretherton’s single-slope relation to liquid-liquid invasion. Note that the present theory is not expected to be valid for invading gas bubbles due to possible effects of compressibility (Gauglitz and Radke, 1989).

**Acknowledgments**

This study was supported through the National Science Foundation (Award No. EAR-0602556) and the Petroleum Research Fund (Award No. 45169-AC9). The authors
are grateful to R. Ewing for fruitful discussions. Comments and suggestions from two anonymous reviewers have helped improve the clarity of the manuscript.
Table 2.1 Synopsis of studies on $b$-$Ca$ behavior for moving gas bubbles.

<table>
<thead>
<tr>
<th>Authors</th>
<th>Range of Ca $b = 1.34Ca^{2/3}$ or $0.5Ca^{1/2}$?</th>
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<tr>
<td>Fairbrother and Stubbs</td>
<td>$10^{-4} \text{–} 10^{-2}$</td>
</tr>
<tr>
<td>Bretherton</td>
<td>$10^{-6} \text{–} 10^{-2}$</td>
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<td>Schwartz et al.</td>
<td>$10^{-5} \text{–} 10^{-3}$</td>
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<td>Ratulowski and Chang</td>
<td>$10^{-6} \text{–} 10^{-1}$</td>
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Table 2.2 Properties of the Wetting (Annulus) Fluid.

<table>
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<th>Annulus fluid</th>
<th>Viscosity (Pa-s)</th>
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<tr>
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<td>Glycerol 85% wt</td>
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<tr>
<td>Glycerol 75% wt</td>
<td>0.036</td>
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Figure 2.1  Geometry of the core-fluid invasion.

Figure 2.2  Schematic of the experimental apparatus showing the capillary tube inside the viewing box.
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<tr>
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<th>$b$</th>
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<th>$b$</th>
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*Data from (Soares et al., 2005, Figures 7, 8, 12, 13)
†Data from (Aul and Olbricht, 1990, Table 2)
‡Data from (Goldsmith and Mason, 1963, Figure 3)
Figure 2.3 Series of images showing a long heptane drop invading the capillary tube filled with glycerol solution. Flow is from left to right; for this experiment, $Ca = 0.04$. The capillary wall is nearly invisible due to index-of-refraction matching between the suspending fluid and the glass. These images were obtained at low magnification ($1\times$) to allow viewing of a large portion of the drop. Accurate measurements of film thicknesses were obtained from images at much higher magnification ($11.6\times$).
Figure 2.4 Summary of experimental data and theories on the wetting-film thickness versus capillary number.
Figure 2.5  Data from Figure 2.4 coded according to viscosity ratios.
CHAPTER 3. FORCED INSTABILITY OF
CORE-ANNULAR FLOW IN CAPILLARY
CONSTRUCTIONS

A paper published in Physics of Fluids
Igor Beresnev, William Gaul, and R. Dennis Vigil

Abstract

Instability of fluid cylinders and jets, a highly nonlinear hydrodynamic phenomenon, has fascinated researchers for nearly 150 years. A subset of the phenomenon is the core-annular flow, in which a non-wetting core fluid and a surrounding wall-wetting annulus flow through a solid channel. The model, for example, represents the flow of oil in petroleum reservoirs. The flow may be forced to break up when passing through a channel’s constriction. Although it has long been observed that the breakup occurs near the neck of the constriction, the exact conditions for the occurrence of the forced breakup and its dynamic theory have not been understood. Here, we test a simple geometric conjecture that the fluid will always break in the constrictions of all channels with sufficiently long wavelengths, regardless of the fluid properties. We also test a
theory of the phenomenon. Four constricted glass tubes were fabricated above and below the critical wavelength required for the fluid disintegration. In a direct laboratory experiment, the breakup occurred according to the conjecture: the fluids were continuous in the shorter tubes but disintegrated in the longer tubes. The evolution of the interface to its pinch-off was recorded using high-speed digital photography. The experimentally observed core-annulus interface profiles agreed well with the theory, although the total durations of the process agreed less satisfactorily. Nonetheless, as the theory predicts, the ratio between the experimental and theoretical times of the breakup process tends to one with decreasing capillary number. The breakup condition and the dynamic theory of fluid disintegration in constricted channels can serve as quantitative models of this important natural and technical phenomenon.

3.1 Introduction

Spontaneous breakup of liquid cylinders and jets into isolated drops is a fascinating natural phenomenon that has captured researchers' attention since the time of Lord Rayleigh (1945, Secs. 357–359). The effect is known as the Plateau-Rayleigh instability. A related phenomenon is instability of the core-annular flow, or the flow of two fluids in a solid capillary channel, in which one (the core) occupies the center of the channel and the other forms the surrounding wall-wetting film. Both phenomena are free-surface, capillarity-controlled flows, a class of strongly nonlinear problems for which no exact theories exist (Middleman, 1995, Secs. 2-3 and 4-3).
A typical approximate theoretical approach to understanding the onset of fluid instability is to subject an initially cylindrical free interface to a small sinusoidal disturbance and determine the wavelength that has the fastest growth rate, assuming the linearity in the early stages of evolution. This is the subject of the linear stability analysis. The size of the resulting drops for both the free jets and the core-annular flows in straight cylindrical capillaries is controlled by this critical wavelength, which depends on the viscosities and the radii of the core and the annulus (Rayleigh, 1945, Secs. 357–359; Tomotika (1935); Goren (1962); Hu and Joseph (1989); Preziosi et al. (1989). The linear-stability approach provides a useful practical means to determine under what conditions the flow is stable and under what it is not; however, this does not provide a description of the nonlinear dynamics of the interface during its evolution to a complete breakup nor does it reveal the characteristic time scale of the process.

The class of problems discussed so far is the spontaneous breakup in cylindrical channels. For the core-annular flow, the disintegration process can be forced by driving the fluids through a constriction. It is known from experience that, in this case, the core pinches off in the vicinity of the neck of the constriction. A wavy wall is essential for the observation of the forced-breakup phenomenon. The effect is widely observed in natural core-annular flows, for example, in the movement of oil surrounded by water in petroleum reservoirs. Constricted pore geometry is typical of natural core-annular flows, in which the potential occurrence of fluid breakup is no longer spontaneous but imposed by the geometry of the channel. It is believed that the phenomenon explains why oil predominantly exists in nature in the form of dispersed tiny droplets.
Although it is known that the forced breakup always occurs near the necks of the constrictions, it has been all but enigmatic as to why in some cases it does take place but not in the others. A unifying framework capable of explaining miscellaneous computational and experimental observations, for arbitrary fluid viscosities, has been lacking. Is it possible to explain the full gamut of observations from a single theoretical standpoint?

We addressed this question theoretically and computationally in two recent studies. First, starting from an earlier suggestion that the channel geometry plays an important role in the occurrence of fluid breakup Roof (1970), we derived an approximate pressure distribution in the core phase based on Laplace’s law of capillary pressure (Beresnev et al., 2009). The criterion for the development of instability in a wavy channel can then be formulated as the exceedance in the pressure in the neck of the profile relative to the pressure in the “crest”, causing the core fluid to leave the neck. In geometric terms, for a sinusoidal channel, this leads to an inequality

\[ L > 2\pi \sqrt{r_{\text{min}}r_{\text{max}}}, \]  

where \( L \) is the wavelength of the channel and \( r_{\text{min}} \) and \( r_{\text{max}} \) are the minimum and maximum radii of the core phase. It turned out that all published experimental and computational studies of the breakup in flows dominated by capillary forces (that is, having relatively small capillary numbers \( Ca \)) in sinusoidal channels, as reviewed by Beresnev et al. (2009), complied with this criterion. It is noteworthy that the criterion 3.1

*Rayleigh (1945); Middleman (1995); Tomotika (1935); Goren (1962); Hu and Joseph (1989); Preziosi et al. (1989); Olbricht and Leal (1983)*
is derived from a simple pressure argument, although it appears to be universally valid. It should, therefore, be more appropriately called a “breakup conjecture”. It declares that, regardless of the viscosities of the fluids, geometry is the only factor controlling whether the fluid disintegrates or not. The first goal of the present publication is to subject this conjecture to a direct experimental test.

Second, the geometric condition 3.1 provides a “yes”/“no” answer but does not establish a dynamic model of the phenomenon. We consequently developed a hydrodynamic theory of the forced instability, which, unlike the previous theoretical studies, is able to follow the interface evolution to a complete disintegration for arbitrary viscosities $\mu_c$ and $\mu_f$ of the core and film fluids, respectively (Beresnev and Deng, 2010b, this publication also summarizes the earlier theoretical studies of the phenomenon). An analytical treatment of this complex hydrodynamic problem is only possible if one assumes smallness of the slope of the interface. For example, suppose the non-dimensional shape of the sinusoidal channel in cylindrical coordinates is

$$\lambda(x) = 1 - a(1 + \cos(\pi \alpha x)), \quad (3.2)$$

where $\lambda(x)$ is the radial position of the channel’s wall, $x$ is the axial coordinate, $a$ is the geometric parameter quantifying the minimum radius of the pore, and $\alpha$ is the slope constant defining the tube’s wavelength $L$, $\alpha = 2/L$ (here all quantities having the dimension of distance have been normalized by the maximum radius of the sinusoidal tube $R_t$). The smallness of the slope is then equivalent to the condition $\alpha \ll 1$. The small-slope assumption, also supposing the smallness of capillary and Reynolds numbers,
allows the use of expressions for Poiseuillean flow for the annulus and the core. The smaller $\alpha$, the better the approximation. The analysis then leads to a fourth-order nonlinear partial-derivative equation for the temporal evolution of the non-dimensional interface profile $\kappa(x, \tau)$: (Beresnev and Deng, 2010b)

$$\frac{\partial \kappa}{\partial \tau} = -\frac{1}{2\pi \mu_c \kappa} \frac{\mu_f Q \partial}{\partial x} \left[ \frac{1 + \frac{2\mu_c}{\mu_f} \left( \frac{\lambda^2}{\kappa^2} - 1 \right)}{\frac{\lambda^2}{\kappa^2} + \frac{\mu_f}{\mu_c} - 1} \right] + \frac{1}{4\kappa} \frac{\partial}{\partial x} \left\{ \kappa^2 \left( \frac{\partial \kappa}{\partial x} + \kappa^2 \frac{\partial^2 \kappa}{\partial x^2} \right) \right\} \times \left\{ \left( \frac{\lambda^2}{\kappa^2} - 1 \right) \left[ \left( \frac{\mu_c}{\mu_f} - \frac{1}{4} \right) \frac{\lambda^2}{\kappa^2} + \frac{3}{4} - \frac{\mu_c}{\mu_f} \right] - \frac{\mu_c}{\mu_f} \ln \frac{\lambda}{\kappa} \right\},$$

(3.3)

where $Q$ is the total volume flux of both fluids through the channel, $\tau$ is the non-dimensional time (the time normalized by the characteristic time scale $\mu_c R_t/\sigma$), and $\sigma$ is the core-film interfacial tension. The equation reflects the complexity of the underlying nonlinear physics but allows inexpensive numerical analysis. An experimental test of the predicted evolution of the interface to its snap-off, governed by Eq. 3.3, is the second goal of the present study.

We are therefore set to perform two tests of the current theoretical understanding of the forced instability in constricted channels. We test the validity of the geometric conjecture 3.1 for the occurrence of core-fluid disintegration. We also validate the correctness of the dynamic theory expressed by differential Eq. 3.3. We next describe the laboratory experiments and their results.

### 3.2 Experiment

Four axisymmetric constricted tubes were manufactured from commercially available straight glass tubing by a glassblower. They will be referred to as the “Short1”, “Short2”,...
“Medium”, and “Long” models. The geometry was prescribed in such a way that the wavelengths of the Short1 and Short2 tubes did not satisfy the condition 3.1; the core fluid, therefore, was not expected to snap off. These were the shortest that the glassblower could produce. On the other hand, the wavelengths of the Medium and Long tubes satisfied the condition. These tubes differed in the value of their slope constant: $\alpha = 0.36$ and $0.061$, respectively.

The photographed tube-wall profiles with least-squares fitted sinusoidal lines are shown in 3.1 (the non-dimensionalization is by $R_t$). For technical reasons, it was much easier to manufacture longer-wavelength constricted tubes close to sinusoidal shapes than the shorter ones. Because of this, the Medium and Long tubes follow nearly perfect sinusoidal lines, while the short ones (especially Short1) exhibit significant departure. However, this does not affect our ability to test the breakup criterion, in which only the maximum and minimum radii matter.

Table 3.1 summarizes the geometric characteristics of all tubes. The radii are the optically measured values. In all cases except Short1, the wavelengths were obtained from the sinusoidal fitting. For the Short1 tube, the wavelength was measured as the distance from the center of the constriction to the point where the radius reached its maximum value.

An experimental apparatus was built to observe the displacement of the suspending fluid by an invading core phase (Figure 3.2). The capillary was placed inside a transparent viewing box. The region between the viewing cell and the tube was filled with glycerol
to reduce optical distortion due to the curvature of the tube. Small ports at either end of the viewing cell provided access for feeding and removing the working fluids into or out of the capillary. At the inlet side, there are two ports: one for the annular fluid and one, covered with a rubber septum, for the injection of the core fluid. Images of the tube were captured by a Photron FASTCAM APX-RS high-speed digital camera mounted above the flow cell.

Aqueous solutions of glycerol were used as suspending fluids (Table 3.2). The core fluids were trichloroethylene (TCE)-heptane mixtures dyed with Oil Blue N. The densities of the two phases were matched to eliminate buoyancy effects. The fluid-fluid interfacial tension measured with a DuNouy tensiometer was $8 \times 10^{-3}$ N/m for all three glycerol/TCE-heptane systems studied.

To remove any organic residue and increase the hydrophilicity of the glass, the capillary was first cleaned in a NaOH-ethanol-deionized-water solution and then flushed with deionized water and dried. In a given run, the capillary was first filled with the annular fluid and the core phase was injected upstream of the test section via a syringe. The volume of the injected core droplet was kept constant at 200 µL. A syringe pump then pumped the annular fluid through the capillary and recording of images took place. The recording ended when the core fluid broke up or the trailing meniscus exited the constriction. The drop velocity (used to calculate the capillary number) could be controlled by changing the speed of the syringe pump. The measured velocities are listed in Table 3.3.
The measurement of the annular-film thickness in pixels was performed in a digital-image editor after the completion of the experiment. This thickness was converted to absolute units through the known optical magnification and the known camera-sensor’s absolute pixel size. Optical resolution of the camera was about 16 µm for the Short and Medium tubes and 1.5 µm for the Long tube. The upstream film thickness was constant before the snap-off process (if any) occurred (e.g., see the movie attached as an electronic supplement to the article. The units on the axes in the movie are mm). This constant thickness was taken as the initial condition in the respective theoretical simulation. The time to breakup was measured by counting the image frames from the high speed camera for shorter breakup times or by a hand timer for longer (greater than 60 s) times.

In an experiment with set parameters (tube type, fluid system, and capillary number), several breakup events were typically analyzed. If the breakup times were relatively short and the core drop was long enough, it could undergo several separate breakup events (if any) as it passed through the constriction. All of them were recorded. For longer breakup times, only one breakup event usually occurred. In that case, several experimental runs were performed. The total number of events analyzed is listed in Table 3.3, in which the film thicknesses and experimental breakup times are the averages of all breakup events for given experiment.
3.3 Results

3.3.1 Test of the geometric conjecture

Table 3.1 lists the minimum wavelength of the channel, calculated from inequality [Eq. 3.1], above which the core fluid is conjectured to always pinch off in the neck. The wavelengths of the Medium and Long tubes are above the threshold value, and the breakup of the core was observed for all capillary numbers (typically between 0.001 and 0.8), except for one case for the Medium tube discussed in 3.3.2. The wavelength of the Short2 tube is below the threshold; breakup in this tube was not observed.

The manufactured wavelength of the Short1 tube turned out to be exactly at the threshold value of 9.2 mm. The core fluid snapped off at capillary numbers above approximately 0.03 and did not at their lower values. This behavior can be understood. The calculation of the threshold wavelengths in Table 3.1 assumed negligible thickness of the wall-wetting film, or the equality between the radii of the tube and the core phase. The Short1 tube therefore was not expected to produce breakup if that was exactly the case. In the experimental runs with larger capillary numbers, the film was visible, which led to the reduction in the values of $r_{\text{min}}$ and $r_{\text{max}}$ relative to the calculation. The breakup condition, therefore, turned out to be satisfied. At smaller capillary numbers, the film could no longer be seen, making the assumption of negligible thickness applicable. The breakup was not observed either.

For the Short2 tube, as calculation using Eq. 3.1 shows, assuming a constant film thickness, the film would have to be thicker than approximately 0.15 mm to reduce the
$r_{\text{min}}$ and $r_{\text{max}}$ for the core phase to make the wavelength of 9.0 mm exceed the threshold. Such thicknesses, even if the film was visible, were never observed (also see Table 3.3), explaining why the snap-off never took place in the Short2 tube.

The experiments thus confirmed the occurrence of fluid instability according to the geometric criterion.

### 3.3.2 Validation of the dynamic theory

In cases of the Medium and Long tubes, we compared the observed dynamic behavior with that predicted by the evolution Equation 3.3, including both the ability of the theory to predict the shape of the interface and the timing of the process.

There are two time scales in the process: a long one during most of the thickening of the suspending-fluid lens in the neck of the constriction and a much shorter one during the precipitous collapse of the interface at the final stage (Gaulglitz and Radke, 1990; Beresnev and Deng, 2010b). Our earlier numerical simulations showed that this latter stage is the one where, over the short time scale, most of the change in the interface profile takes place (Beresnev and Deng, 2010b). This final stage, therefore, provides the crucial testing of the correctness of the theory.

The summary of experiments is presented in Table 3.3. The viscosity ratios tested varied by a factor of 40. The values of $h$ are the initial film thicknesses formed by the invasion of the core through the neck of the constriction. The breakup times ($t_T$ -
theoretical, \( t_E \) - experimental) were counted from the moment when the film was formed until the completion of the pinch-off.

Figures 3.3 and 3.4 present the observed and theoretically predicted shapes of the core-annulus interface (the flow is to the right). There are differences in the recording of the process between the Medium and Long tubes that are important for understanding the way the results are shown. As seen from Table 3.3, the breakup times are much shorter for the Medium tube (on the order of seconds) compared with the Long tube (minutes). Because of the precipitous nature of the collapse of the interface at the final stage, and in order to capture the dynamics near the final point, the sampling interval of the digital camera had to be sufficiently small. The limited camera memory then imposed restrictions on the record duration. The entire evolution of the interface from invasion to pinch-off could still be recorded for the Medium tube; however, this was not possible for the Long tube because of the much longer duration. In the latter case, we could only film a fraction of the total time of the process near the pinch-off moment.

Figure 3.3 shows the position of the wall channel as well as the theoretical and experimental interface profiles for the Medium tube for two values of the capillary number. The “snapshots” were taken very near collapse. Numerical restrictions did not allow advancing the theoretical interface arbitrarily close to the channel axis, as a singularity was quickly approached: \( \kappa \) tends to zero in the denominator of \( 1/\kappa^4 \) in Eq. 3.3 and the solution “blows up”. The theoretical and experimental shapes agree very closely.
Figure 3.3(B) also shows the interface obtained by using the commercial computational-fluid-dynamics (CFD) code FLUENT. As in the simulations by Beresnev and Deng (2010b), FLUENT was run in the volume of fluid (VOF) axisymmetric model for the immiscible multi-phase flow. The mesh-generation software GAMBIT was used to construct the grid on which the axial and radial momentum, continuity, and volume-fraction equations were solved. The following solution schemes for the VOF model were applied: the PREssure STaggering Option (PRESTO) for pressure interpolation, the second-order discretization for the volume-fraction equation, the Pressure Implicit with Splitting of Operators (PISO) scheme for pressure-velocity coupling, and the second-order upwind discretization for the momentum equations. As seen from Figure 3.3(B), the deviation from the experiment around the neck of the constriction in the FLUENT simulation is greater than that for the theoretical curve obtained from the solution of Eq. 3.3.

The electronic-supplement movie shows the full recorded process of the breakup in the experiment corresponding to Figure 3.3(B), where the complete details of the evolution can be seen.

In one instance, indicated in Table 3.3, the breakup in the Medium tube did not take place ($Ca = 0.02$). This is the only experiment with the Medium and Long tubes at which the film was not visible either. Our interpretation of the fact is that, although the breakup was allowed by the geometry, the influx of the suspending fluid into the neck was insufficient, due to the thinness of the film, to complete the process before the drop exited the constriction.
Figure 3.4 presents the results of the comparison for the Long tube. The thin lines show the wall and the theoretical interface near the breakup. The camera’s field of view in the Long-tube experiment covered a smaller portion of the total wavelength of the tube: the respective sections of the wall and the observed interface are shown by the thick lines. For the reason explained, we filmed only the final stage of the evolution near the interface collapse, while numerical instability did not allow advancing the theoretical solution arbitrarily close to the collapse. The lower thick line shows the experimentally observed initial position of the interface when the filming started. There is certain mismatch in the timing at which the theoretical and experimental snapshots are taken; given this circumstance, the agreement between the experimental and theoretical shapes can be considered good. The locations of the pinch-off, slightly shifted downstream from the neck, match better between the theory and the experiment as the capillary number decreases.

The theory, therefore, is capable of adequately capturing the details of the shape of the evolving interface for both the Medium and Long tubes. We also tested the ability of the theory to explain the correct timing of the process. The ratios of the theoretical-to-observed breakup times $t_T/t_E$ are listed in Table 3.3. For the Medium tube, there is under-prediction by the factors of two and nine. For the Long tube, there is, conversely, over-prediction; however, it tends to decrease from approximately a factor of four to close to one from large to small capillary numbers. The greater mismatch between the theory and experiment for the Medium tube and for larger capillary numbers is to be expected, recalling the assumptions of the small slope and small capillary number built into the
theory. Curiously, the ratio of $t_F/t_E$, where $t_F$ is the breakup time obtained by FLUENT, corresponding to the CFD simulation shown in Figure 3.3(B), is 1.1. FLUENT, thus, better than the theory reproduced the timing of the process but not the shape of the interface.

### 3.4 Conclusions

Instability and disintegration of the core-annular flow can be forced by passing the fluids through a capillary constriction. Such a forced breakup has been observed in some instances but not in the others, with the deciding factors remaining unclear.

We formulated a “geometric breakup conjecture” stating that, regardless of the fluid properties, the channel geometry is the factor determining whether the fluid snap-off will take place. The geometric condition 3.1 is derived from simple pressure argument and is consistent with the literature on the phenomenon. The criterion 3.1 is counterintuitive at first glance: it indicates that the fluid in longer channels with gently sloping walls is susceptible to instability, whereas that in “sharper” channels with short wavelengths will flow continuously. The explanation of this phenomenon will bear on our ability to understand the state of oil in tortuous pore spaces in petroleum reservoirs.

Four constricted glass tubes were fabricated below and above the critical wavelength defined by inequality 3.1. In all tubes, the breakup occurred as expected from the conjecture.
A theory of the evolution of the interface to its disintegration, for arbitrary fluid viscosities, has not been known either. We have proposed the nonlinear evolution Equation 3.3 as a quantitative model of the process. The smaller the slope parameter $\alpha$ and the capillary number, the more accurate the theory is expected to be. The interface profiles and breakup times, following from this equation, were compared to the experimentally observed ones in the tubes with two slope parameters, $\alpha = 0.36$ and 0.061. For both tubes, the experimental shapes of the interface near the breakup agreed with the theoretical prediction, the match improving with decreasing capillary number. Note that one consequence of the imposed flow is the occurrence of the breakup downstream of the constriction, as seen in Figures 3.3 and 3.4; the exact location of the pinch-off is also theoretically captured better as $Ca$ decreases. The agreement in the timing of the process is less satisfactory. Nonetheless, the ratio between the theoretical and experimental breakup times for the smallest-slope tube tends to one with decreasing capillary number, which also is an expectation of the theory.

Note that we have not attempted to extend the applicability of both the geometric conjecture and the dynamic evolution equation into the range of capillary numbers exceeding one. The derivation of the equation and the formulation of the criterion explicitly assume the preponderance of capillary forces over the viscous ones, expressed in the smallness of $Ca$. Our experiments thus stayed approximately within the limits of applicability of the approach, to which the conclusions apply. However, in the capillary-number range tested, from 0.006 to 0.8, there is still a variety of observed behaviors (Table 3.3). In the practical applications to oil recovery that we have already mentioned
and in the flow of fluids through natural porous media, the smallness of the capillary number is a very good approximation (Melrose and Brandner, 1974).

The theory and the yes/no criterion for the occurrence of forced breakup in core-annular flow in capillary constrictions compare favorably against experiment. They can serve as quantitative models of this widespread natural and technical phenomenon within their limits of applicability.

Acknowledgements

This study was supported through National Science Foundation (Award No. EAR-0602556) and Petroleum Research Fund (Award No. 45169-AC9). The CFD simulations for Figure 3 were performed by W. Deng. The authors are indebted to Robert Ewing for helpful discussions and to the two anonymous referees whose comments improved the clarity of the presentation.
Table 3.1  Geometric parameters of the constricted tubes.

<table>
<thead>
<tr>
<th>Tube type</th>
<th>Minimum radius (mm)</th>
<th>Maximum radius (mm)</th>
<th>Wavelength (mm)</th>
<th>Predicted minimum wavelength for the breakup (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Short1</td>
<td>0.5</td>
<td>43</td>
<td>9.2</td>
<td>9.2</td>
</tr>
<tr>
<td>Short2</td>
<td>0.6</td>
<td>4.6</td>
<td>9.0</td>
<td>10.4</td>
</tr>
<tr>
<td>Medium</td>
<td>0.4</td>
<td>4.3</td>
<td>24.0</td>
<td>8.2</td>
</tr>
<tr>
<td>Long</td>
<td>0.3</td>
<td>4.2</td>
<td>138.5</td>
<td>7.0</td>
</tr>
</tbody>
</table>

Table 3.2  Properties of the wetting (annulus) fluid.

<table>
<thead>
<tr>
<th>Annulus fluid</th>
<th>Viscosity (Pa-s)</th>
<th>Density (g/cm³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Glycerol 100% wt</td>
<td>1.4</td>
<td>1.3</td>
</tr>
<tr>
<td>Glycerol 85% wt</td>
<td>0.11</td>
<td>1.2</td>
</tr>
<tr>
<td>Glycerol 75% wt</td>
<td>0.036</td>
<td>1.2</td>
</tr>
</tbody>
</table>
Figure 3.1 Profiles of the experimental tubes fitted to sinusoidal shapes.

Figure 3.2 Schematic of the experimental apparatus.
Table 3.3 Experimental Values of Frequencies, Pressure Drops, and Ganglion Lengths

<table>
<thead>
<tr>
<th></th>
<th>Medium tube ((\alpha=0.36))</th>
<th>Long tube ((\alpha=0.061))</th>
</tr>
</thead>
<tbody>
<tr>
<td>(\mu_f/\mu_c)</td>
<td>2820 103 71</td>
<td>2820 2820 218 218 71</td>
</tr>
<tr>
<td>Number of events(^a)</td>
<td>5 1 0</td>
<td>4 5 4 4 3</td>
</tr>
<tr>
<td>(h) (mm)</td>
<td>0.11 0.096 Unresolved</td>
<td>0.099 0.064 0.044 0.023 0.017</td>
</tr>
<tr>
<td>(Ca)</td>
<td>0.2 0.06 0.02</td>
<td>0.8 0.2 0.06 0.02 0.006</td>
</tr>
<tr>
<td>(t_T) (s)</td>
<td>5.1 0.7 -</td>
<td>62 332 84 497 316</td>
</tr>
<tr>
<td>(t_E) (s)</td>
<td>10.7 6.6 No breakup</td>
<td>83 74 144 230</td>
</tr>
<tr>
<td>(t_T/t_E)</td>
<td>0.48 0.11 -</td>
<td>4.1 4.0 1.1 3.5 1.4</td>
</tr>
</tbody>
</table>

\(^a\)Total number of observed breakup events.
Figure 3.3  Experimental and theoretical interface profiles near breakup for the Medium tube
Figure 3.4  Experimental and theoretical interface profiles near breakup for the Long tube
CHAPTER 4. DIRECT PORE-LEVEL OBSERVATION OF PERMEABILITY INCREASE IN TWO-PHASE FLOW BY SHAKING

A paper published in Geophysical Research Letters
Igor Beresnev, William Gaul, and R. Dennis Vigil

Abstract

Increases in permeability of natural reservoirs and aquifers by passing seismic waves have been well documented. If the physical causes of this phenomenon can be understood, technological applications would be possible for controlling the flow in hydrologic systems or enhancing production from oil reservoirs. The explanation of the dynamically increased mobility of underground fluids must lie at the pore level. The natural fluids can be viewed as two-phase systems, composed of water as the wetting phase and of dispersed non-wetting globules of gas or organic fluids, flowing through tortuous constricted channels. Capillary forces prevent free motion of the suspended non-wetting droplets, which tend to become immobilized in capillary constrictions. The capillary entrapment significantly reduces macroscopic permeability. In a controlled experiment with a constricted
capillary channel, we immobilize the suspended ganglia and test the model of capillary entrapment: it agrees precisely with the experiment. We then demonstrate by direct optical pore-level observation that the vibrations applied to the wall of the channel liberate the trapped ganglia if a predictable critical acceleration is reached. When the droplet begins to progressively advance, the permeability is restored. The mobilizing acceleration in the elastic wave, needed to “unplug” an immobile flow, is theoretically calculated within a factor of 15 of the experimental value. Overcoming the capillary entrapment in porous channels is hypothesized to be one of the principal pore-scale mechanisms by which natural permeabilities are enhanced by the passage of elastic waves.

4.1 Introduction

Permeability increases in porous geologic formations induced by the passage of seismic waves, e.g., at the times of earthquakes, have been well documented. In nature, they may lead to a greater inflow of groundwater into aquifers or enhanced productivity of oil wells (Beresnev and Johnson, 1994; Roberts, 2005; Elkhoury et al., 2006). Despite sufficient evidence of the widespread nature of the phenomenon, its underlying physical mechanism has remained unclear. Understanding this mechanism could help harness the phenomenon and turn it into a technology of controlled permeability, with far-reaching practical implications in hydrology and reservoir engineering (Roberts et al., 2003; Elkhoury et al., 2006). Direct observation of the enhanced fluid flow under the effect of elastic waves at the pore level could be a way of revealing such an underlying pore-scale mechanism.
A feature that most aquifers and petroleum reservoirs share is their being two-phase hydrological systems, in which one phase is wetting and one is non-wetting. The non-wetting phase forms isolated droplets, surrounded by the suspending wetting fluid, usually water. In petroleum reservoirs, the non-wetting phase is oil, existing in the form of isolated droplets (ganglia), and in aquifers it is composed of air bubbles. Realistic pore channels also are tortuous, formed by a succession of open spaces connected by narrow constrictions (pore throats). The flow of dispersed non-wetting globules of oil and air through the constrictions is resisted by capillary forces: if the pressure gradient driving the flow is sufficiently low, entrapment of the globules in pore constrictions takes place. This capillary entrapment is known as the Jamin effect (Taber, 1969). The Jamin effect, restricting the two-phase flow through channels of variable cross-section, explains, for example, the danger of introducing air bubbles into the blood stream. A natural porous system contains many channels with widely varying tortuous geometries. The entrapped non-wetting droplets “plug” the flow in some of them, while in others the flow may still be occurring. When the effect is summed up over a multitude of channels, the macroscopic permeability of the porous volume is significantly reduced.

One of the mechanisms of reduced permeability in two-phase hydrological systems thus has capillary nature. If the local vibrations, induced by the passage of seismic waves, can mobilize the entrapped globules that plug the flow, the flow rate through the volume will increase, which will be seen as increased macroscopic permeability. This is a possible explanation of the enhancing effect of seismic waves on the permeability in reservoirs and aquifers.
We built an experimental apparatus that allows us to entrap a ganglion of a non-wetting fluid, suspended in a wetting phase, in a transparent constricted capillary. The capillary is subjected to longitudinal vibrations, which correspond to the oscillations induced on the wall of a natural pore by a passing wave, resolved onto the pore axis. We present the demonstration of the mobilization of an initially immobile ganglion with the beginning of vibrations and address the quantitative explanation of the phenomenon.

4.2 Experiment Description

An axisymmetric sinusoidally-constricted capillary was manufactured from commercial glass tubing by a glassblower. The tube minimum \( r_{\text{min}} \) and maximum \( r_{\text{max}} \) radii are 0.12 and 4.23 mm, and the wavelength is 24.2 mm. The capillary was placed inside a transparent Lucite viewing box. The region between the viewing cell and the tube was filled with a glycerol solution matched to the refractive index of the glass tube (Figure 4.1). Small ports at either end of the viewing cell provided access for feeding and removing the working fluids into or out of the capillary. At the inlet side there are two ports: one for the suspending fluid and one, covered with a rubber septum, for the injection of the core (non-wetting) fluid. Images of the tube were captured by a high-speed digital camera with a magnifying lens mounted above the flow cell.

Deionized water was used as the suspending fluid (viscosity of \( 10^{-3} \) Pa\( \cdot \)s). The core organic phase was a trichloroethylene-heptane mixture dyed with Oil Blue N (viscosity of \( 0.44 \times 10^{-3} \) Pa\( \cdot \)s). The densities \( \rho \) of the two phases were matched to 998 kg/m\(^3\) to eliminate buoyancy effects. The fluid-fluid interfacial tension \( \sigma \) was \( 16 \times 10^{-3} \) N/m.
To remove any residue and increase the hydrophilicity of the glass, the capillary was first cleaned in a NaOH-ethanol-deionized-water solution and then flushed with deionized water and dried. In a given run, the capillary was first filled with the suspending fluid, and a known volume of the core phase sufficient to fill the constriction was injected upstream of the constriction via a syringe. Flow was controlled by a gravity feed system. A syringe pump constantly fills an overflow feed tank whose height can be adjusted precisely with a micrometer stage while the outflow tube is kept at a fixed height: this creates a constant external pressure drop $\Delta P_e$ along the capillary, driving the flow. The pressure drop was measured with an inclined manometer and a differential pressure transducer.

As a suspended droplet of the core fluid moves toward the channel’s constriction, and if $\Delta P_e$ is below a certain critical value (the “static” threshold $\Delta P_{th}$), the droplet is unable to overcome the capillary barrier and is trapped. The flow is “plugged” and the permeability is effectively zero. The mobilization threshold can be experimentally determined by first keeping $\Delta P_e < \Delta P_{th}$, entrapping the drop, and then slowly raising the feed tank until the droplet movement through the constriction takes place. The flow is “unplugged” and the permeability is restored. The mobilizing pressure drop is then $\Delta P_{th}$.

For the experiment with vibrations, the background pressure drop $\Delta P_e$ is established below $\Delta P_{th}$, to first entrap the ganglion. The shaker then starts to sinusoidally vibrate the capillary-tube assembly in the axial direction at a given frequency. The vi-
ibration starts at a low amplitude and is increased in a stepwise manner, lasting several tens of vibration periods at each step to wait for the mobilization. The amplitude $A$ of the longitudinal acceleration of the tube is measured by an accelerometer. If an amplitude can be reached at which the ganglion starts to progressively advance and exits the constriction, it is referred to as the threshold acceleration amplitude $A_{th}$. The flow is unplugged and permeability is restored solely by the application of vibrations.

4.3 Demonstration of the Mobilization of Plugged Flow by Vibrations

As discussed by Beresnev (2006); Beresnev and Deng (2010a), the value of the mobilizing acceleration can significantly increase with the frequency. Higher-frequency waves also attenuate in rock much faster, which makes them less practically relevant. For these reasons, the experiments were run at the vibration frequencies of 5, 7.4, 10, and 14.2 Hz in the “seismic” range. The frequencies are increased by $\sqrt{2}$, with minor deviations for purely technical reasons. At each frequency, runs were made with one to three different values of the background pressure drop $\Delta P_e$, as summarized in Table 4.1. The experimentally determined static thresholds $\Delta P_{th}$ and the lengths $l$ of the entrapped ganglia, measured between the front and rear three-phase contact lines, are also indicated.

If the model of capillary entrapment holds, the static threshold can be determined via

$$\Delta P_{th} = 2\sigma (1/r_{min} - 1/r_r), \quad (4.1)$$
where $r_r$ is the radius of the rear meniscus, assuming the spherical menisci shapes (Taber, 1969; Beresnev, 2006). The experimentally determined values of $\Delta P_{th}$ from Table 4.1 are compared with those calculated from equation 4.1, for the different lengths of the entrapped ganglion $l$, in Figure 4.2. The agreement is excellent.

In all cases but one (5 Hz, $\Delta P_e = 95$ Pa) (Table 4.1), an entrapped ganglion, resting under the external pressure drop $\Delta P_e$ with the front meniscus inside the constriction and unable to move, became mobilized (and the flow unplugged) when a vibration of sufficiently large amplitude was applied. The threshold acceleration amplitudes $A_{th}$ are also shown in Table 4.1. Figure 4.3 presents, as an example, a magnified camera view (from above) of the movement of the initially entrapped droplet (dark color at right) through the constriction, displacing the water ahead (light color at left), for the $\Delta P_e = 171$ Pa, 5 Hz case. The position of the front meniscus is indicated by the arrows. Small air bubbles are also seen in the water. The time stamps are from the start of the vibrations. The images are separated by exactly two periods of vibration. It should be noted, though, that Figure 4.3 is provided for a rough illustration of the process only. Since the mobilization takes many periods to be complete, it is impossible to render its rich dynamic process in a few reduced-size still images. Therefore, a complete animation is provided as Animation S1 in the auxiliary material*, which the reader needs to watch before continuing.

The movie starts with an entrapped ganglion at rest. The time when the vibrations began is marked as zero. The starting amplitude of the vibrations is $0.06 \, g$ (0.6 m/s$^2$).

*http://dx.doi.org/10.1029/2011GL048840
(\(g\) is the acceleration of gravity). It increases to 0.08 \(g\) (0.8 m/s\(^2\)) at approximately 5.3 s, and to 0.09 \(g\) (0.9 m/s\(^2\)) at 14.9 s. As one can see, the two initial amplitudes are insufficiently strong to mobilize the droplet, which simply continued back-and-forth movements within the constriction. However, when the threshold value of 0.9 m/s\(^2\) was attained, the droplet became fully mobile in approximately eleven periods, resuming its unrestricted flow through the capillary. The capillary barrier was overcome.

4.4 Explanation of the Observations

Is it possible to quantitatively explain the existence and variability of the threshold acceleration needed to "unplug" an immobile two-phase flow system? A "static" mobilization criterion and a dynamic theory of the mobilization of entrapped blobs of non-wetting fluids have been proposed by Beresnev (2006); Beresnev and Deng (2010a). In the immobile state, the background pressure drop along the blob is resisted by capillary force. The application of vibrations creates an extra "inertial" forcing that adds to the background pressure gradient. When the sum of the background and the vibrational inertial forcing exceeds the resisting capillary force, the mobilization takes place. The relationship between these three body forces at the liberation moment leads to the mobilization criterion for the threshold acceleration amplitude (Beresnev, 2006),

\[
A_{th} = (\Delta P_{th} - \Delta P_e)/\rho l. \tag{4.2}
\]

On the other hand, the balance of all body forces acting on the ganglion, including the external gradient, the capillary force, the oscillatory forcing created by vibrations, and
the viscous drag, can be combined in a single equation of motion, leading to a dynamic theory of the phenomenon (Beresnev, 2006; Beresnev and Deng, 2010a).

Equation 4.2 shows that the inertial body force $\rho A_{th}$ has to be added by vibrations to compensate for the “resisting” body force $(\Delta P_{th} - \Delta P_e)/l$. This can be thought of as an additional external pressure gradient instantly added to the system.

Figure 4.4 shows the comparison of the mobilizing accelerations predicted by both the mobilization criterion 4.2 and the dynamic theory, relative to the experimental values. To obtain the theoretical values, Beresnev (2006, Eq. 8), with the viscous term introduced by Beresnev and Deng (2010a, Eq. 6) added, was numerically solved. Comparisons are presented for all combinations of experimental parameters in Table 4.1. The dynamic theory is generally closer, expectedly, to the experimental values than a simpler static criterion, predicting the mobilizing acceleration almost exactly for the case of 14.2 Hz. Overall, the theory consistently over-predicts the observed acceleration by a factor of 1.2 to 5.3. The deviations from the experiment can be due to a number of factors, including the dynamic contact angle, the non-ideal spherical shape of the menisci, and the non-ideal sinusoidal shape of the constriction. Since there is not a satisfactory analytical model for a dynamic contact angle and the shape of a moving meniscus, a theory providing a more accurate match to the observations seems unfeasible at present.

The value of $A_{th}$ is generally dependent on both the magnitude of the “capillary barrier” $(\Delta P_{th} - \Delta P_e)$ and the frequency of vibrations (Beresnev, 2006; Beresnev and Deng, 2010a). The mobilization criterion 4.2 only captures the dependence on $(\Delta P_{th} -$
ΔP_e) (from which its name “static” is derived), while the full dynamic theory is needed to capture both effects. This is one of the explanations why the values calculated from the theory in Figure 4.4 are generally in better agreement with the experiment.

Beresnev (2006, Eq. 9–10) also proposed that the frequency of the vibratory action be low enough for the stimulation to be effective, to allow the fluids sufficient time to respond. This threshold frequency is estimated as the inverse of the viscous response time τ, \( \tau = \frac{\rho r^2}{\mu} \), where \( r \) is the channel radius and \( \mu \) is the fluid viscosity. For the meniscus in the constriction, with the values of the order \( \rho = 10^3 \text{ kg/m}^3 \), \( r = 0.1 \text{ mm} \), \( \mu = 10^{-3} \text{ Pa·s} \), the threshold frequency is \( 1/\tau = 100 \text{ Hz} \). The frequencies used in the experiments are thus below the estimated threshold.

The theoretical framework available also offers an explanation of the only case (5 Hz, ΔP_e = 95 Pa) when the mobilization of the entrapped droplet was not attained. The technical limit on the maximum acceleration amplitude of the shaker is 0.45 g (4.4 m/s²); the application of this acceleration did not lead to the mobilization. The threshold amplitudes \( A_{th} \) calculated from equation 4.2 and the dynamic theory are 12.6 and 4.7 m/s², respectively. These values are consistent with the observation: the experimentally applied acceleration was still below (if only slightly) the theoretical unplugging level.

### 4.5 Conclusions

Increases in permeability of natural hydrological and reservoir systems induced by passing seismic waves have been reliably documented. A quantitative explanation of the
phenomenon, based on a testable physical mechanism, has nevertheless been missing. Understanding the pore-scale mechanism of the permeability enhancement is a prerequisite for possible uses of this phenomenon to achieve controlled or engineered permeability, with potentially significant economic implications.

Natural reservoirs and aquifers typically are two-phase fluid systems, filled with water as a wetting phase that suspends the non-wetting globules, such as gas or organic fluids. Capillary forces restrict the free movement of the suspended phase through the tortuous porous channels, entrapping the non-wetting globules in the narrow pore throats. The permeability of a channel with an entrapped droplet is effectively reduced to zero.

An entrapped body can be mobilized if an external pressure drop along its length exceeds a certain critical value. We compared these threshold values determined from the model of capillary entrapment with the experimentally observed ones: the agreement is excellent.

We have further experimentally demonstrated that the capillary entrapment is overcome if longitudinal vibrations of a critical amplitude are applied to the wall of the channel, “unplugging” the stuck body and restoring the permeability. The “unplugging” effect of vibrations on two-phase flow should be one of the pore-scale mechanisms by which seismic waves enhance permeability of the natural fluid-saturated porous systems. Other possible mechanisms may exist at the intermediate levels between the pore and the field scales. One of the effects under investigation in fractured laboratory samples is
unclogging of fracture apertures by the mobilization of fine particles, induced by shaking or pulsing pressure (Liu and Manga, 2009; Elhoury et al., 2011).

An analytical model of the droplet mobilization by seismic waves adequately explains the observations. It is able to predict the amplitude of the mobilizing acceleration within a factor of 1–5 of the observed value. Given the uncertainties in parameterizing the dynamic contact angle and the shape of a moving meniscus, a better theory of the seismic permeability enhancement seems to be beyond reach at present time.

**Acknowledgments**

This study was supported through the National Science Foundation (EAR-0602556) and Petroleum Research Fund (45169-AC9). The authors are grateful to R. Ewing for fruitful discussions, and to M. Broadhead and an anonymous referee for the comments. The Editor wishes to thank two anonymous reviewers for their assistance evaluating this paper.
Table 4.1 Experimental Values of Frequencies, Pressure Drops, and Ganglion Lengths

<table>
<thead>
<tr>
<th></th>
<th>5Hz</th>
<th>5Hz</th>
<th>5Hz</th>
<th>7.4Hz</th>
<th>7.4Hz</th>
<th>10Hz</th>
<th>14.2Hz</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\Delta P_e$ (Pa)</td>
<td>95</td>
<td>171</td>
<td>227</td>
<td>206</td>
<td>237</td>
<td>203</td>
<td>183</td>
</tr>
<tr>
<td>$\Delta P_{th}$ (Pa)</td>
<td>246</td>
<td>246</td>
<td>246</td>
<td>257</td>
<td>248</td>
<td>248</td>
<td>257</td>
</tr>
<tr>
<td>$l$ (mm)</td>
<td>11.8</td>
<td>11.8</td>
<td>11.8</td>
<td>6.4</td>
<td>8.9</td>
<td>8.5</td>
<td>7.9</td>
</tr>
<tr>
<td>$A_{th}$ (m/s²)</td>
<td>-</td>
<td>0.9</td>
<td>0.7</td>
<td>1.6</td>
<td>0.6</td>
<td>1.3</td>
<td>3.2</td>
</tr>
</tbody>
</table>

Figure 4.2 Comparison of the static mobilization thresholds $\Delta P_{th}$, determined from the model of capillary entrapment (equation 4.1), with the experimentally measured values.
Figure 4.3 Snapshots from Animation S1 showing the liberation of an entrapped non-wetting ganglion under the effect of vibrations.
Figure 4.4  Comparison of the threshold amplitude $A_{th}$ of the acceleration required to unplug an entrapped blob: mobilization criterion 4.2, dynamic theory, and experiment.
CHAPTER 5. CONCLUSIONS

5.1 Summary

The purpose of this dissertation is to develop further insight into the behavior of liquid-liquid flow in porous media. Several important observations were made in pursuit of this goal.

The initial thickness of the residual film left by an invading drop is critical to generating meaningful initial conditions for the nonlinear evolution equation used to model the dynamic “snap-off” behavior of oil ganglia. Bretherton’s half-century old theory offers a solution, but with unanswered controversy as it was developed for gas, not liquid, drops. Thus an acceptable, experimentally verified theory for one liquid displacing another has not been available. Here we develop a hydrodynamic, testable theory providing an explicit relationship between the thickness of the wetting film and the liquids’ properties.

The thickness of the film is theorized to be roughly independent of the viscosity ratio between the core and film fluids (Eq. 2.5). Data from our experiments and other researchers (Fig. 2.4), do not show any significant scatter with respect to viscosity ratio.
This supports the theory’s conclusion. At the same time, Bretherton’s theory, originally deduced for films surrounding moving gas bubbles but often believed to hold for liquids as well, fails to explain the observations. Bretherton’s derivation was explicitly dependent on the assumption of constant zero pressure in the core (gas) phase. We believe to be an oversimplification, which may explain why the suggested behavior is not seen in the experiments. To fit an experimentally observed $b-Ca$ relationship, we have used the actual variable pressure gradients in the core.

With a proper relationship describing the film thickness we can test the behavior of a long drop passing through a constricted capillary. This represents the flow of oil in petroleum reservoirs. The flow may be forced to break up when passing through a channel’s constriction and the upstream portion of the drop may become trapped by capillary pressure, thus reducing the apparent permeability of the geologic formation. Although it has long been observed that the breakup occurs near the neck of the constriction, the exact conditions for the occurrence of the forced breakup and its dynamic theory have not been understood. Thus, we formulated a “geometric breakup conjecture” stating that, regardless of the fluid properties, the channel geometry is the factor determining whether the fluid snap-off will take place. The geometric condition 3.1 is derived from simple pressure argument and is consistent with the literature on the phenomenon.

We also test a novel theory for the behavior of the interface, from post invasion to eventual breakup, using a nonlinear evolution Equation 3.3 as a quantitative model of the process. In our experiments, the breakup occurred according to the conjecture: the fluids
were continuous in the shorter tubes but disintegrated in the longer tubes. The experimentally observed core-annulus interface profiles agreed well with the theory, although the total durations of the process agreed less satisfactorily. Nonetheless, as the theory predicts, the ratio between the experimental and theoretical times of the breakup process tends to one with decreasing capillary number. The breakup condition and the dynamic theory of fluid disintegration in constricted channels can serve as quantitative models of this important natural and technical phenomenon within their limits of applicability.

With an understanding of when and how a drop may become trapped in a capillary constriction and the behavior of the drop interface within it, we can now test vibration as a means to mobilize the drop. Increases in permeability of natural reservoirs and aquifers by seismic waves are well documented (Beresnev and Johnson, 1994). However, a quantitative explanation of the phenomenon, based on a testable physical mechanism, nonetheless has been missing. If the physical causes of the permeability enhancement can be understood, technological applications would be possible for controlling the flow in hydrologic systems or enhancing production from oil reservoirs.

The reason for the increased mobility of underground fluids must be at the pore level. Natural reservoirs and aquifers typically are two-phase fluid systems, composed of water as the wetting phase and globules of gas or organic fluids as the dispersed, non-wetting phase, flowing through tortuous constricted channels. Capillary forces restrict the free motion of the suspended phase, which tend to become immobilized in narrow pore throats. The capillary entrapment significantly reduces macroscopic permeability.
We can test the theory of capillary entrapment with a controlled experiment involving a single constricted capillary channel. We measure the pressure required to immobilize a single drop; the theory agrees precisely with the experiment. We then demonstrate by direct optical pore-level observation that the vibrations applied to the wall of the channel liberate the trapped ganglia if a predictable critical acceleration is reached. When the droplet begins to progressively advance, the permeability is restored. Overcoming the capillary entrapment in porous channels is hypothesized to be one of the principal pore-scale mechanisms by which natural permeabilities are enhanced by the passage of elastic waves.

An entrapped body can be mobilized if an external force (mainly from a pressure gradient) acting along its length exceeds a certain critical value. We compared these threshold values determined from the model of capillary entrapment with the experimentally observed ones: the agreement is excellent. We have further experimentally demonstrated that the capillary entrapment is overcome if longitudinal vibrations of a critical amplitude are applied to the wall of the channel in addition to the pressure gradient, “unplugging” the stuck body and restoring the permeability (see the shaded portion of the sinusoid in Fig. 1.2 labeled Mobilization). The “unplugging” effect of vibrations on two-phase flow should be recognized as one of the pore-scale mechanisms by which seismic waves enhance permeability of the natural fluid-saturated porous systems.
5.2 Recommendations

We have created, and verified via a series of experiments, an ensemble of capillary physics mechanisms capable of describing the complexities of two-phase flow in porous media with more accuracy and detail than previous theories, where they actually existed before. These set of tools open a new range of technical applications that can exploit the ability to temporarily modify the effective permeability of porous media.

The current three-pronged approach to the subject matter—experimental studies, numerical simulations of theory, and computational fluid dynamics—has proved very effective in producing quality results and should be continued in the future. Further work should continue exploring the dynamic behavior of vibratory mobilization, including a more detailed examination of the effects of frequency. A more modern high-speed camera with improved optics would also help in resolving the rapid collapse of the film ‘necking’ behavior just prior to snap-off and in resolving the very thin films that occur with small Ca.

So far, the mobilization mechanism studies have been restricted to single pores. While this is important in determining fundamental behavior, an extension should be made into more complex two and three dimensional models, both in theory and experimentally. Previous multi-dimensional experiments have been fraught with extremely low levels of entrapment, so fluid pairs and pore sizes need to be chosen more carefully as to create situations that have enough capillary entrapment to be usefully measured, before and after vibratory mobilization.
Looking beyond the experimental validation of theory, investigations into the feasibility of vibratory stimulation in real-world, small scale operations for EOR and possibly CO$_2$ sequestration may be of interest.

With these suggested improvements and expansions, we can further our understanding of the oil mobilization and related processes, leading to useful technical applications.
APPENDIX A. Refractive index effects

Purpose

In 1.4 it was discussed that it was desirable to have the annular fluid have a matching refractive index with respect to the glass wall of the tube in order to eliminate optical distortion. Practical reasons (being limited to a narrow capillary number range) outweighed this ideal. This appendix aims to quantify what effect non-matching refractive indices has on the measured size and observed shape of the invading drop.

Experimental cross-section

Consider the cross-section of circular glass capillary, a wetting annular fluid, and a non-wetting core fluid. Under the ideal case where the two fluid densities have been matched, thus minimizing the effects of buoyancy, the two fluids will form a circular interface and a uniform film thickness with respect to the capillary wall (Fig. A.1). $R$ is the internal diameter of the glass capillary with refractive index $n_1$, with $n_2$ as the refractive index of the wetting fluid, $r$ is the diameter of the non-wetting drop, $x$ is the distance of an incoming ray of light from the centerline of the drop/capillary tube, $x'$ the end point of the ray of light entering at $x'$, $\theta_1$ and $\theta_2$ are the incoming/outgoing angles of
the ray of light with respect to the normal of the wall of the capillary (dashed line going through the center of the drop/capillary tube and point $A$) and are determined through trigonometry A.1, Snell’s Law A.2, and a bit of algebra A.3.

\[
\sin \theta_1 = \frac{x}{R} \quad \text{(A.1)}
\]

\[
n_1 \sin \theta_1 = n_2 \sin \theta_2 \quad \text{(A.2)}
\]

\[
\theta_2 = \sin^{-1} \left( \frac{n_1 x}{n_2 R} \right) \quad \text{(A.3)}
\]

We now know the angle the light ray makes with respect to a centerline passing through $A$, but to solve for where it strikes the drop we need to know its slope. We know the slope of the centerline A.4 and using a rotation matrix A.5 we can determine the slope of the refracted ray A.6.

\[
O = (x_1, y_1) = (0, 0)
\]

\[
A = (x_2, y_2) = (x, \sqrt{R^2 - x^2})
\]

\[
m = \frac{y_2 - y_1}{x_2 - x_1} \quad \text{(A.4)}
\]
\[
\begin{vmatrix}
  x' \\
y'
\end{vmatrix} = \begin{vmatrix}
  \cos \theta & -\sin \theta \\
  \sin \theta & \cos \theta
\end{vmatrix} \begin{vmatrix}
  x \\
y
\end{vmatrix}
\]
(A.5)

\[
x' = x \cos \theta - y \sin \theta
\]
\[
y' = x \sin \theta + y \cos \theta
\]

\[
m = \frac{x_2 \sin \theta + y_2 \cos \theta - (x_1 \sin \theta + y_1 \cos \theta)}{x_2 \cos \theta - y_2 \sin \theta - (x_1 \cos \theta - y_1 \sin \theta)}
\]
(A.6)

Simplifying A.6

\[
m = \frac{x_2 \sin \theta + y_2 \cos \theta}{x_2 \cos \theta - y_2 \sin \theta}
\]
(A.7)

and substituting trigonometric identities

\[
m = \frac{\frac{n_1}{n_2} x^2 + \sqrt{R^2 - x^2} \sqrt{1 - \left(\frac{n_1}{n_2} \frac{x}{R}\right)^2}}{x \sqrt{1 - \left(\frac{n_1}{n_2} \frac{x}{R}\right)^2 - \sqrt{R^2 - x^2 \left(\frac{n_1}{n_2} \frac{x}{R}\right)}}}
\]
(A.8)

From here it is trivial to determine where the light ray strikes the drop, but in this general case this arbitrary point has little meaning. In relation to the experiment, the point of interest is where the light ray strikes the very edge of the drop. Such a light ray would strike the drop at a tangent (Fig. A.2).

First, it is useful to define reduced variables.

\[
n^* = \frac{n_1}{n_2}
\]
\[
x^* = \frac{x}{R}
\]
\[
r^* = \frac{r}{R}
\]
The slope of the refracted light ray.

\[ m = \frac{n^*x^2 + \sqrt{1-x^2}\sqrt{1-(n^*x^2)^2}}{x^*(\sqrt{1-(n^*x^2)^2} - n^*\sqrt{1-x^2})} \]  

(A.9)

Equations for the capillary wall, drop, and slope of the drop.

\[ R(x) = \sqrt{R^2 - x^2} \]
\[ r(x) = \sqrt{r^2 - x^2} \]  

(A.10)

\[ \frac{\partial r(x)}{\partial x} = \frac{-x}{r(x)} \]

And in reduced form.

\[ R^*(x^*) = \sqrt{1-x^*^2} \]
\[ r^*(x^*) = \sqrt{r^*^2 - x^*^2} \]  

(A.11)

\[ \frac{\partial r^*(x^*)}{\partial x^*} = \frac{-x^*}{r^*(x^*)} \]

The light ray must intersect the curve of the drop at exactly one point A.12 and also have matching slope A.13

\[ r(x') = R(x) + (x' - x)[m] \]
\[ \sqrt{r^*^2 - x'^*^2} = \sqrt{1-x'^2} + (x'^* - x^*) \frac{n^*x'^2 + \sqrt{1-x'^2}\sqrt{1-(n^*x'^2)^2}}{x^*(\sqrt{1-(n^*x^2)^2} - n^*\sqrt{1-x^2})} \]  

(A.12)

\[ \frac{-x'}{r(x')} = [m] \]
\[ \frac{-x'^*}{\sqrt{r^*^2 - x'^*^2}} = \frac{n^*x'^2 + \sqrt{1-x'^2}\sqrt{1-(n^*x'^2)^2}}{x^*(\sqrt{1-(n^*x^2)^2} - n^*\sqrt{1-x^2})} \]  

(A.13)
Using MATLAB’s symbolic solver the following (4) solutions are found.

\[
x^* = \pm \left( \frac{2r^5 \sqrt{n^2 - 2r^2}(r^2\sqrt{1 - r^2} \pm r^2\sqrt{n^2 - r^2})}{n} \right) - \frac{r^3\sqrt{n^2 - 2r^2}(r^2\sqrt{1 - r^2} \pm r^2\sqrt{n^2 - r^2})}{n^2r^4 - n^2r^2 + r^4}
\]

\[
\cdots + \frac{r\sqrt{1 - r^2}(r^2\sqrt{1 - r^2} \pm r^2\sqrt{n^2 - r^2})\sqrt{n^2 - 2r^2}(r^2\sqrt{1 - r^2} \pm r^2\sqrt{n^2 - r^2}) + r^2 - n^2r^2}{n^2r^4 - n^2r^2 + r^4}
\]

(A.14)

Collecting like terms

\[
x^* = \pm \left( \frac{2r^5 - r^3 + r\sqrt{1 - r^2}(r^2\sqrt{1 - r^2} \pm r^2\sqrt{n^2 - r^2})}{n^2r^4 - n^2r^2 + r^4} \right)
\]

\[
\cdots \times \frac{n^2 - 2r^2(r^2\sqrt{1 - r^2} \pm r^2\sqrt{n^2 - r^2}) + r^2 - n^2r^2}{n}
\]

(A.15)

Distributing and canceling terms in the first half; distributing, cancelling, and collecting terms to form a perfect square allowing us to ‘de-nest’ the radical on the second half.

\[
x^* = \pm \left( \frac{r^5 \pm r^3\sqrt{1 - r^2}\sqrt{1 - r^2} \sqrt{1 - r^2} \sqrt{n^2 - r^2} \mp r^3}{n^2r^4 - n^2r^2 + r^4} \right)
\]

(A.16)

As a result of multiplying a \pm by a \mp the products of the first terms and last terms of each half of the equation sum to zero, leading to

\[
x^* = \pm \left( \frac{n^2r^4 - n^2r^2 + r^4}{n} \right)
\]

(A.17)

The \(r^7\) terms in the numerator and several powers of \(r\) between the numerator and denominator cancel out.
\[ x^* = \pm \left( \frac{\pm(-n^2 r^2 + n^2 - r^2)r}{(n^2 r^2 - n^2 + r^2)n} \right) \]  \hspace{1cm} (A.18)

Yielding the final solution of

\[ x^* = \pm \frac{r}{n} \]  \hspace{1cm} (A.19)

Where the positive solution is the one we are looking for.

**Discussion**

As we can see from A.19, the effect from a mismatch between the refractive indices of the capillary tube and the wetting fluid is that of simple magnification, and that the magnification has a value of \( M = \frac{n_2}{n_1} \). That is to say when the index of refraction of the wetting fluid is less than that of the capillary tube the drop will appear smaller. This effect is determined only by the ratio of the two indices of refraction and is irrespective of the capillary or drop size, or the radial position of the incoming light ray. This means that even along a capillary (and consequently drop) of varying radius the magnification effect is consistent.

Figure A.3 shows the magnification effect on multiple rays of light spaced at \( \frac{1}{10} \) tube diameter intervals. The first six rays \( (x = 0.1-0.6) \) are refracted inside the capillary, while the last three \( (x = 0.7-0.9) \) are reflected out, as predicted by Snell’s Law A.2 and the law of reflection. The point at which this behavior switches is the apparent edge of the capillary tube. Its value is equal to the magnification equation \( (M = x = 0.667) \).
The ray entering at \( x = 0.5 \) is refracted and hits the edge of the drop \( (r = 0.75) \). Thus the apparent size of the drop is 0.5 and the apparent size of the capillary is 0.667.

In figures A.1 A.2 A.3 the ratio of refractive indices is \( 1.5/1.0 \) in order to exaggerate the effects of the magnification \( (M = 0.667) \). The wetting fluid used in the experiments (glycerol \( n_2 = 1.473 \)) more closely matches the refractive index of the glass used in the capillary tubes (borosilicate \( n_1 = 1.474 \)). The glycerol was diluted with water to lower its viscosity to achieve a large range of capillary numbers, this also lowered its refractive index. The refractive index of the most dilute glycerol solution (75% glycerol 2.2) was \( n_2 = 1.438 \). From A.19 this is a magnification of \( M = 0.9756 \times \) or relative error of 2.44%. However, the film thicknesses \( (b \) or \( 1 - r \)) in Table 2.4 are normalized by the radius of the capillary, so the reported \( b \) value for Figure A.3 would be \( b = 1 - \frac{0.500}{0.667} = 0.25 \), which is the correct value. In the experiments, both the apparent film thickness and capillary diameter were measured and since they were both affected by the same magnification the reported \( b \) values are in fact correct, notwithstanding that the relative error for either measurement is \(< 2.5\% \) for all of the experiments.
Figure A.1  Effects of non-matching refractive indices on an incoming ray of light, general case.

Figure A.2  Effects of non-matching refractive indices on an incoming ray of light, apparent edge of the drop.
Figure A.3 Effects of non-matching refractive indices on an incoming ray of light, incoming rays spaced at $\frac{1}{10}$ tube diameter intervals.


**BIBLIOGRAPHY**


