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A microvisual study of viscosity and mass transfer effects on two-phase flow in porous media

Cline, John Gilbert, M.S.

Rice University, 1989
RICE UNIVERSITY

A MICROVISUAL STUDY OF
VISCOSITY AND MASS TRANSFER EFFECTS
ON TWO-PHASE FLOW IN POROUS MEDIA

by

John Gilbert Cline

A THESIS SUBMITTED
IN PARTIAL FULFILLMENT OF THE
REQUIREMENTS FOR THE DEGREE
MASTER OF SCIENCE

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Houston, Texas
April, 1989
A MICROVISUAL STUDY OF VISCOSITY AND MASS TRANSFER EFFECTS ON TWO-PHASE FLOW IN POROUS MEDIA

by

John G. Cline

ABSTRACT

A microvisual flow cell was used to observe the effects of fluid viscosity ratio and of pore size distribution on the mechanisms of steady cocurrent two-phase flow in porous media. The transition in flow mechanisms was found to have the same dependence on capillary number, $N_{ca}$, for all viscosity ratios provided $N_{ca}$ was defined in terms of the interstitial velocity of the wetting phase.

A simple theoretical model of ganglia flow through an idealized pore constriction was developed. Although velocity varied greatly during ganglion passage, the average volumetric flow rate of the nonwetting phase agreed well with relative permeability theory.

A series of displacement experiments were performed with several oil-water-alcohol systems where diffusion occurred between oil and water phases. The amount of spontaneous emulsification observed was found to be greatest in systems where both Marangoni flow and local super saturation due to diffusion were expected.
DEDICATION

To my wife Sharon;

for all of her

support, love and understanding

during my graduate experience.
ACKNOWLEDGEMENTS

I would like to take the time to express my most sincere appreciation to the people who helped make this thesis a reality. Specifically:

Professor Clarence A. Miller for his time, attention, patience and understanding. Without his help, this thesis would not have become a reality;

The United States Coast Guard for their financial and computer software support;

Professors Bedient and Zygourakis for serving on my thesis committee.
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1. INTRODUCTION

This thesis deals with certain aspects of fluid flow in porous media, a topic of interest to many areas of applied science and engineering. Some of these are groundwater hydrology and the removal of organic contaminants from soils and groundwater aquifers, oil reservoir engineering, membrane transport, deep bed filtration, and fluidized beds. Since the mechanisms of two-phase flow in porous media which are the chief concern here have special relevance to the first two topics, they are discussed further in the following sections.

1.1 Groundwater

Groundwater is a very important and vital resource. The term groundwater refers to the subsurface water that is found beneath the water table. This groundwater is located in soils and other geologic formations that are saturated with water. The primary goal of groundwater hydrology is the understanding of groundwater flow.

The study of groundwater is becoming more interdisciplinary in nature. In recent years the importance of our groundwater resource has become more evident, and this increased awareness has stimulated interest from many disciplines. I believe this trend will continue and is desirable as other disciplines have much to offer. For example, many of the flow in porous media problems that face the reservoir engineer are very similar to those faced by the hydrologist.
1.1.1 Groundwater and the Hydrologic Cycle

The hydrologic cycle is the circulation network of water in our environment. This network contains the oceans, the atmosphere and the land. Groundwater plays a very important role in this cycle. Figure 1.1 illustrates this process, and identifies the important features of the hydrologic cycle and groundwater flow for a given watershed.

When applied to the watershed in Figure 1.1, this basic cycle consists of three main components: inflow; outflow; and storage. Inflow is due to precipitation and snowmelt. Outflow is in the form of runoff, stream flow, and evapotranspiration (a combination of evaporation from water bodies and transpiration from plants). Storage is located in lakes or groundwater aquifers.

To put the importance of groundwater into perspective, Table 1.1 (1) provides an estimate of the global water balance. If we focus only on available fresh water, excluding icecaps and glaciers, it becomes apparent that 95% of the available fresh water is groundwater. With our survival dependent upon water, the importance of groundwater is obvious.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Volume (%)</th>
<th>Residence Time</th>
</tr>
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<tr>
<td>Oceans and seas</td>
<td>94</td>
<td>4000 years</td>
</tr>
<tr>
<td>Lakes and Reservoirs</td>
<td>&lt;0.01</td>
<td>10 years</td>
</tr>
<tr>
<td>Swamps</td>
<td>&lt;0.01</td>
<td>1-10 years</td>
</tr>
<tr>
<td>River channels</td>
<td>&lt;0.01</td>
<td>2 weeks</td>
</tr>
<tr>
<td>Soil moisture</td>
<td>&lt;0.01</td>
<td>2 weeks-1-year</td>
</tr>
<tr>
<td>Groundwater</td>
<td>4</td>
<td>2 weeks to</td>
</tr>
<tr>
<td></td>
<td></td>
<td>10,000 years</td>
</tr>
<tr>
<td>Icecaps and glaciers</td>
<td>2</td>
<td>10-1000 years</td>
</tr>
<tr>
<td>Atmospheric water</td>
<td>&lt;0.01</td>
<td>10 days</td>
</tr>
<tr>
<td>Biospheric water</td>
<td>&lt;0.01</td>
<td>1 week</td>
</tr>
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</table>
Figure 1.1 Schematic representation of the hydrologic cycle (1).
1.1.2 Groundwater as a Resource

Traditionally, the principal concern in the study of groundwater has been the most efficient use of this valuable resource. Groundwater is used in many industrial processes, as well as for potable (drinking) water for domestic dwellings, and for irrigation of crops.

It is important to note that while 95% of the available fresh water is groundwater, ground water is a resource of limited renewal. It make take thousands of years to establish a particular aquifer and have the formation fill with water. Water removed from an aquifer is replaced by the process of recharge. Man is able to remove water from an aquifer at a rate much higher than that by which it is naturally recharged. Without careful management, an aquifer system may become depleted over the course of several decades with little hope of the water being replaced in our lifetimes.

All of the above uses place a strain on the limited groundwater resource. In the past the emphasis has been on obtaining the maximum yield from a given groundwater supply. More recently this trend has been shifting toward long term management of a valuable and limited resource.

1.1.3 Groundwater Contamination

Over the last few decades man has been producing waste products, and disposing of them in the environment at a rate which the environment can no longer easily absorb. Many of these hazardous waste materials have found or are finding their way into our groundwater supplies. In the last decade, new and important environmental legislation has been enacted which is helping to stop this disposal
activity and to clean up past mistakes. This action is critical if we are preserve our precious groundwater resource.

When an aquifer is invaded by a liquid organic contaminant having a low solubility in water, the mechanisms of two-phase flow are important for its distribution and removal. The presence of the additional organic phase in the aquifer changes the basic hydrodynamics of the groundwater flow. Some of the results obtained from the work described in this thesis enhance our understanding concerning two-phase flow in porous media. This understanding can be applied to the problem of removing contaminants from our groundwater resource.

1.2 Reservoir Applications

An oil reservoir is typically a consolidated porous medium, which contains many small pores of irregular geometry. In any medium of this type capillary forces, related to the curvature of individual fluid drops, are very important when two or more phases are present. In oil reservoirs, under typical production conditions, capillary forces dictate which pores are occupied by which fluids and thus have a major influence on flow.

The ratio between viscous forces and capillary forces is an important dimensionless number called the capillary number, \( N_{ca} \). In simplest terms, the capillary number is defined as:

\[
N_{ca} = \frac{\nu \mu}{\gamma} \tag{1.1}
\]

where \( \nu \) is the superficial velocity of the fluid, \( \mu \) is its viscosity, and \( \gamma \) is the interfacial tension between two fluids.
There are several other related definitions that may be used to calculate $N_{ca}$ (2). These other methods are more complicated in nature, but do not yield any additional information relevant to this work.

When multiple phases are present in a particular porous medium, the location of a particular phase is determined by capillary forces and wetting properties. In many cases where two phases are present, a porous medium is wet more strongly by one fluid than by the other. When this happens, the nonwetting phase is found in the larger pores, and the wetting phase in the smaller pores. In this way the free energy of the system is minimized.

Under typical reservoir production conditions inertial and viscous forces are negligible compared to capillary forces. Under some production conditions, however, viscous forces become more important and can even dominate the flow process. One example is chemical flooding discussed in more detail below.

1.2.1 Physical Description

Oil reservoirs are typically found in sedimentary rock. By the very nature of its formation, the physical properties of sedimentary rock can vary greatly. The properties vary from one formation to another, and generally vary considerably within the same formation. Since sedimentary rocks were deposited in layers, physical properties often vary more rapidly in the vertical direction, i.e., perpendicular to the bedding planes, than in either horizontal direction.

There are three main types of sedimentary rock commonly associated with oil reservoirs. These are shales, sandstones, and carbonate rocks. The properties of
each type of rock can vary, depending upon their initial formation history and subsequent geologic history.

The two most important physical parameters that characterize a given formation are its porosity, $\phi$, and its permeability, $k$. The porosity is simply the fraction of the total formation volume that is not filled with solid, i.e. the void fraction. Effective porosity is defined in the same manner except that it includes only connected pores. Isolated voids in the rock are not counted as they have no effect upon flow processes. Values of porosity vary widely between different types of sedimentary rocks.

Permeability is a measure of how easily a fluid will flow through a porous medium and is often anisotropic in nature. Permeability is generally much lower in the direction perpendicular to the bedding planes than it is in the directions parallel to the bedding planes. The units for permeability are defined in the following section.

Some representative values of porosity and permeability are given in Table 1.2 (information compiled from (1), (3), and (8)). Sandstones and old carbonates are typical reservoir rocks while old shales are typically found over or underlying the reservoir rock.

<table>
<thead>
<tr>
<th>Type of rock</th>
<th>Porosity</th>
<th>Permeability (darcies)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Shales</td>
<td>10-25%</td>
<td>1E-03</td>
</tr>
<tr>
<td>Shales, old</td>
<td>2-10%</td>
<td>1E-05</td>
</tr>
<tr>
<td>Sandstones</td>
<td>10-40%</td>
<td>1E-03-3.5</td>
</tr>
<tr>
<td>Carbonate, young</td>
<td>20-50%</td>
<td>1000</td>
</tr>
<tr>
<td>Carbonate, old</td>
<td>0-20%</td>
<td>.01-1</td>
</tr>
</tbody>
</table>
Once the physical properties are known, they may be used to predict how the reservoir will perform, and how much oil will be produced.

1.2.2 Single Phase Flow

Single phase flow in porous media at low velocities is described by Darcy's Law. It is named after the French hydraulic engineer Henry Darcy. Darcy developed this law while studying the flow of water through sand beds and columns in 1856. One of the most commonly used forms of Darcy's Law is:

$$v = \frac{k}{\mu} (\nabla p - \rho g)$$  

where $v$ is the superficial velocity vector, $k$ the permeability, $\mu$ the fluid viscosity, $\nabla p$ the applied pressure gradient, $\rho$ the fluid density, and $g$ the gravitational vector.

This equation is valid for low Reynolds numbers, i.e. in the laminar flow regime. Typical process applications in oil reservoirs and groundwater aquifers fall in this regime.

Equation [1.2] defines the permeability, $k$. As can be seen from eqn [1.2], $k$ has the dimensions of $[\text{length}]^2$. Common units of permeability are darcies, and cm$^2$. One darcy is equal to $9.87 \times 10^{-9}$ cm$^2$.

In eqn [1.2] $k$ has been assumed to be homogeneous and isotropic, hence a constant. This assumption is valid for this work due to the physical nature of the experimental apparatus. In the general case the scalar $k$ in eqn [1.2] can be replaced by a dyadic which has nine scalar components. It is thought that the dyadic is symmetric which allows three principal directions to be defined.
1.2.3 Relative Permeability Theory

When there is more than one fluid phase present in the porous medium, eqn [1.2] must be modified (6). The modification is required because each fluid phase no longer has the entire porous medium available for flow. Relative permeability theory is currently used to describe this behavior. The permeability term in eqn [1.2] is modified to account for the effect of the additional fluid phase, and Darcy’s Law is then written for each fluid phase.

If one of the two fluids preferentially wets the porous medium, it is referred to as the wetting phase, denoted by the subscript w. The second fluid will be called nonwetting, and will be denoted by the subscript nw. Making the modification described above to eqn [1.2] yields:

\[ v_w = \frac{-k_{rw}k}{\mu_w} (\nabla p_w - \rho_w g) \] \[ v_{nw} = \frac{-k_{rnw}k}{\mu_{nw}} (\nabla p_{nw} - \rho_{nw} g) \]

where all terms are as defined for eqn [1.2], and \( k_r \) denotes the relative permeability of phase i, and \( \nabla p_i \) is the pressure gradient in phase i.

Equations [1.3] can be used to calculate the superficial or Darcy velocity of each phase. The permeability \( k \) is a constant, but the relative permeabilities are strong functions of the saturations of their respective phases. At low \( N_{ca} \) it has been generally established that relative permeabilities are independent of pressure gradients (3). Two typical graphs of relative permeability as a function of saturation are shown in Figure 1.2. The upper figure is for a consolidated sand, and the lower figure is for unconsolidated glass spheres (21).
Figure 1.2 Relative permeability curves for consolidated sands and unconsolidated glass spheres (21).
Relative permeabilities are generally independent of the individual viscosities, and the viscosity ratio. However, if the wetting fluid has a much lower viscosity than the nonwetting fluid, this is not always correct. In this case the flow of the nonwetting phase can be higher than expected near the irreducible saturation of the wetting phase. The cause of this behavior is thought to be a thin layer of the wetting phase which covers part of the rock matrix, and provides a lower resistance to flow for the nonwetting phase. This is also called the 'lubrication' effect.

The wettability of the porous medium also has an effect on relative permeability. The basic effect is that the relative permeability of the wetting phase is lower than that of the nonwetting phase when both fluids are present in comparable amounts. This effect is caused by the location of the wetting phase in the smaller pores.

The final major parameter that affects relative permeability is $N_{ca}$. As $N_{ca}$ increases, the shape of the relative permeability vs saturation curves changes. The curves tend to become straight lines intersecting at a saturation of 50%. When this occurs, the relative permeability of a phase is equal to its saturation (See Figure 1.3).

The range of $N_{ca}$'s of interest for this work is $1 \times 10^{-4}$ to $1 \times 10^{-2}$. Two-phase flow mechanisms in this $N_{ca}$ range are not completely understood.

1.2.4 Capillarity and Residual Phase Trapping

As previously stated, when more than one fluid phase is present in a porous medium, capillary forces become important. Capillary pressure between two fluid phases is defined as follows (19):

$$p_c = p_{nw} - p_w$$

[1.4]
Figure 1.3 Gas-oil "relative permeability" of Fontainebleau sandstone for very low IFT values (taken from (3) original source Bardon and Longeron, 1978, SPE-AIME).
The Laplace capillarity equation can be used to calculate the pressure change across the curved interface between two fluids A and B. The Laplace capillarity equation is:

\[ p_A - p_B = -2H \gamma \]  \hspace{1cm} [1.5]

Where \(-H\) is the mean curvature of the interface between the fluids, and \(\gamma\) is the interfacial tension between them. It is important to note that capillary pressure is directly proportional to interfacial tension and the mean curvature. The mean curvature is defined as the average value of curvature taken in the two principal directions of curvature:

\[ -2H = \frac{1}{r_1} + \frac{1}{r_2} \]  \hspace{1cm} [1.6]

where \(r_1\) and \(r_2\) are the radii of curvature in the principal directions. The negative sign associated with equation [1.6] is a convention. If we examine an interface between fluid A and fluid B, the radius of curvature is defined to be positive if the center of curvature lies on the A side of the interface, and negative if it lies on the B side of the interface. See Figure 1.4.

If the interface between fluids A and B is spherical, then the mean curvature of the interface is equal to \(1/r\) where \(r\) is the radius of the sphere. In this case equation [1.5] simplifies to:

\[ p_A - p_B = 2\gamma/r \]  \hspace{1cm} [1.7]

Equation [1.7] shows that large capillary pressures are present when interfacial tension is large and/or when the radius of curvature is small. In a typical oil reservoir, both of these conditions are often encountered.
When interfacial tension is not equal to zero, there will always be a pressure jump across a curved interface. Consider for example a slug of fluid A surrounded by fluid B inside a small capillary tube. If the drop of fluid A has the same curvature at each A - B interface, then the net pressure difference across a static drop will be zero. See Figure 1.5. The general expression for the capillary pressure change across a static drop is derived by writing Laplace's equation for each interface. For the points 1 and 2 in Figure 1.5, assuming interfaces that are spherical caps, Laplace's equation becomes:

$$\Delta p_c = p_{c2} - p_{c1} = 2\gamma \left[ \frac{1}{r_2} - \frac{1}{r_1} \right] \quad \ldots \ldots \ldots \ldots \ldots [1.8]$$

Under certain conditions $\Delta p_c$, as defined by equation [1.8], will be greater than zero. Two important situations where this condition may arise are the case of a converging axisymmetric pore throat, and that of a system with contact angle hysteresis. Both of these conditions are discussed in detail below.

During a water flood of an oil reservoir, water is injected into the formation to drive out oil. As the flood progresses and oil is displaced by water, the saturation of the oil phase decreases until there is no longer sufficient oil to maintain a continuous oil phase. When this saturation is reached, the oil phase breaks up into discrete blobs or ganglia. Once these ganglia form, they may or may not remain mobile. A mobile ganglion will become trapped when the pressure drop across it due to viscous flow forces is exceeded by the capillary pressure increase.

An illustration of this trapping process is the converging pore throat shown in Figure 1.6. As the ganglion moves into the converging pore throat, the radius of curvature at point 2 decreases, causing an increase in $\Delta p_c$. When $\Delta p_c$ is equal to the
Figure 1.5
viscous pressure difference across the drop, the drop will no longer flow and becomes trapped.

A similar type of trapping can occur when there is contact angle hysteresis. The contact line is the line of contact of the fluid-fluid interface with the solid. The contact angle is the angle at which the fluid-fluid interface contacts the solid surface. In some systems the contact angle at the leading interface of a drop of fluid A may differ from that of the trailing interface. This difference is called hysteresis. Hysteresis can be caused by surface roughness and impurities on the solid surface (19).

Consider the oil drop in a capillary tube illustrated in Figure 1.7. Here the contact angles are measured through the water phase. The advancing contact angle, $\theta_A$ is defined by the contact line that is advancing into a region previously wet by the oil. The receding contact angle, $\theta_R$ is defined by the contact line that is receding into a region previously wet by the water. When $\theta_R$ is less than $\theta_A$, $\Delta p_c$ is greater than zero. Under these conditions a single ganglion may become trapped in a fashion similar to that described above for the converging pore throat.

1.2.5 Steady Cocurrent Flow

Most of the current literature considers the topic of displacement of one fluid by another in porous media. The work by Elsik (16) reviews the more limited literature concerning steady cocurrent flow. Elsik’s research, which is summarized below, has more clearly defined the mechanisms of steady cocurrent flow.
Figure 1.7
As with displacements, Elsik found that the mechanisms of steady two-phase flow are a function of $N_{ca}$. There are three distinct $N_{ca}$ regions, each having a unique flow mechanism which dominates the observed flow. The first region corresponds to low $N_{ca}$, $N_{ca} < 1 \times 10^{-4}$. In this region the flow field is dominated by the capillary forces. Both fluid phases flow in continuous channels or pathways which are tortuous in nature and are referred to as Continuous Tortuous Paths (CTP). The boundaries between the fluids are very stable down to the pore level. The concept of relative permeability is borne out in the micro-model Elsik used.

The second region is an intermediate $N_{ca}$ transition region from about $1 \times 10^{-4} < N_{ca} < 1 \times 10^{-2}$. In this transition region the CTP break up and discrete ganglia of the non-wetting phase are flowing. At each capillary number there is a narrow distribution of the lengths of the mobile ganglia about some mean value. The mean value is inversely proportional to the capillary number. In Elsik's work the length of the mobile ganglia was about 10 Bead Diameters, (BD) where one bead is $109 \mu$m in diameter, at a capillary number of about $1 \times 10^{-3}$. Singlets, ganglia of about 1 BD, are the mean size at $N_{ca}$ of about $1 \times 10^{-2}$.

The length of the mobile ganglia is measured in the direction of flow. At a given $N_{ca}$, ganglion size is uniform within experimental accuracy. A few longer ganglia are observed, but they are short lived and break up by either splitting or snapoff. When breakup occurs, smaller daughter ganglia are formed. If the new ganglia are larger than the average mobile value, they will be mobile. If these new ganglia are too small, they will become trapped by capillary forces, usually in a pore body. This trapping is produced by the mechanisms described previously in section 1.2.4. During cocurrent flow, however, a trapped ganglion will probably not remain trapped for long. It will be mobilized when another free flowing ganglion collides
with it and the two coalesce. The ganglion formed by this union is generally large enough to move freely.

The third $N_{ca}$ flow region is for high $N_{ca}$, where $N_{ca} > 1 \times 10^{-2}$. In this region the mobile ganglia are all less that 1 BD in length, and are called subsinglets. The subsinglet size is still inversely related to $N_{ca}$ but the relation is no longer linear. The subsinglets are elongated in the direction of flow. In this region viscous forces affect the flow pattern of the subsinglets as well as their size. The paths traveled by both phases are no longer tortuous in nature.

As $N_{ca}$ is increased even further to $1 \times 10^{-1}$, the subsinglets begin to resemble filaments which follow the streamline patterns that would exist in the pore network during single phase flow. This flow resembles classic fluid mechanics experiments where dye has been injected into single phase flow to visualize the streamlines.

As a quick summary, at low $N_{ca}$ capillary forces determine the location of the wetting and nonwetting phases in the pore structure, the nonwetting phase occupying the larger pores. As $N_{ca}$ increases, the effect of viscous forces increases. These forces first cause the breakup of the CTP, then are responsible for the decrease in mobile ganglia length.

1.2.6 Primary and Secondary Recovery

During the recovery of oil from a particular oil reservoir, a sequential process is often followed (5). The first step is well discovery and primary recovery. During primary recovery the natural forces within the reservoir, along with pumping, provide the force necessary to drive oil to the wells. These natural forces are generally due to the expansion of volatile components in the crude oil as the reservoir pressure is
reduced. During primary recovery only oil and gas are generally produced. Once the natural drive forces have been expended, the next recovery step begins.

Secondary recovery normally consists of waterflooding the oil reservoir to augment the natural driving forces. During this process, more wells are often drilled. Some of the wells are used for injecting water into the reservoir, while others are used to withdraw oil from the reservoir. There is generally a specific pattern of well placement to help insure good sweep efficiency during the flood. One of the more common well patterns is the so-called five-spot. This pattern is a square with an injection well at each of the four corners, and a single production well in the center. This basic pattern is repeated as necessary to cover the reservoir.

During secondary recovery, both oil and water are produced. There are several costs associated with this recovery. The primary costs are those of separating the recovered oil and water, of treating this water and re-injecting it into a disposal formation (or into the injection wells), of injecting the water drive, and of pretreating the injection water. When these costs exceed a certain percentage of the value of the oil produced, the process is terminated.

1.2.7 Enhanced Oil Recovery

Enhanced Oil Recovery (EOR), is often called tertiary recovery, and follows primary and secondary recovery. The motivation for EOR is that after primary and secondary recovery methods have been employed, there can still be as much as 60% of the original oil remaining in the reservoir. The potential yield of an EOR process can be greater than the amount of oil produced by both primary and secondary recovery combined.
There are two main methods of EOR. The first involves the injection of chemical energy, and the second the injection of thermal energy into the reservoir. Several examples of chemical EOR are presented in reference (5). Figure 1.8 is a diagram of a typical (alkaline) chemical flood taken from (5). A typical CO₂ flood is illustrated in (5). A third type of chemical EOR method is flooding with a micellar-polymer solution to lower interfacial tension and provide mobility control. Chemical EOR methods have great potential and are current areas of research. Even with their potential, only CO₂ is currently being used to a significant extent.

Thermal EOR is normally used for high viscosity oils. There are three main types of thermal EOR. They are steam flooding, a single well scheme called a steam soak, and in situ combustion of the reservoir. In situ combustion is a very complex process. Reference (5) contains illustrations of the above mentioned processes as well as more detailed information concerning each process. Steam processes are currently widely used.

1.3 Research Objectives

The principal objective of this work is to extend the understanding of the microscopic behavior of fluids in porous media. Much is currently known about the macroscopic behavior, but the same is not true concerning the microscopic behavior. Of particular interest is the transition of flow mechanisms at high N_{ca}. This behavior will be examined on a microscopic level by direct observation using enhanced video microscopy. The previous work of Curtis M. Elskik will be extended in certain directions.

The specific areas of interest for this work are four fold. The first area examined will be the effect of nonwetting phase viscosity on the mechanism of two-
CHEMICAL FLOODING
(Alkaline)

The method shown requires a preflush to condition the reservoir and injection of an alkaline or alkaline/polymer solution that forms surfactants in situ for releasing oil. This is followed by a polymer solution for mobility control and a driving fluid (water) to move the chemicals and resulting oil bank to production wells.

Mobility ratio is improved, and the flow of liquids through more permeable channels is reduced by the polymer solution resulting in increased volumetric sweep.

(Single 5-Spot Pattern Shown)

Figure 1.8 Schematic diagram of an enhanced oil recovery chemical flooding process (5). (Original source U.S. Department of Energy, Bartlesville, OK).
phase steady flow. Of particular interest is the effect of a high nonwetting phase viscosity, when compared to the wetting phase. This effect is important in understanding the removal process of high viscosity oils from oil reservoirs or groundwater.

The second area examined will be the effect of pore geometry on the same mechanisms of steady flow. Of particular interest here is the effect on ganglion size distribution due to a more random packing. It is expected that a more random pore size distribution will decrease the length of mobile ganglia. It is important to establish the effect of pore size distribution on the mechanisms of steady two-phase flow and to understand how this will effect oil removal from a porous media.

The third area of interest is preliminary work on displacements of one fluid phase by another, with mass transfer occurring between the fluid phases. In particular the effect of spontaneous emulsification on the displacement mechanics will be studied. In this work preliminary displacement experiments were conducted where both displaced and displacing fluids were not in chemical equilibrium prior to their contacting in the flow cell. The previous work by Elsik used only fluid pairs in chemical equilibrium. It is believed that spontaneous emulsification may increase the rate of insitu biodegradation of organic contaminants in groundwater aquifers. Therefore a basic understanding of the factors controlling the amount of spontaneous emulsification produced in a displacement is necessary before spontaneous emulsification can be used effectively to cleanup a contaminated groundwater aquifer.

The fourth and final area of interest is the formulation of a theoretical model to study the effect of nonwetting to wetting phase viscosity ratio on the movement of
an oil ganglion through an idealized pore constriction. The model is presented along with numerical examples and graphs to illustrate the results. Understanding how viscosity can effect the movement of a mobile ganglion through a pore constriction is important in reservoir engineering as the desired result is removal of the oil. Hence it is important to understand how viscosity effects the velocity of a mobile ganglion, as a sufficient reduction in the ganglion's velocity can result in its trapping.

1.3.1 Thesis Outline

Description of the fluid systems and the experimental apparatus used are detailed in Chapter 2. Two-phase steady cocurrent flow experiments are described in Chapter 3. Displacement experiments are discussed in Chapter 4. A theoretical model of ganglion flow is presented in Chapter 5.
2. EXPERIMENTAL METHODS

2.1 Fluid Systems

There are several fluid systems used in this work. Different fluid systems were chosen for each type of flow experiment. All of the chemicals used were reagent grade with the exception of the squalane, which is 2, 6, 10, 15, 19, 23-
Hexamethyldecosane. The squalane was used as received from Shell Development Company. The ultrapure water used was prepared using a SYBRON/Barnstead purification system. The local tap water was first pretreated, then distilled, and passed through a NANOpure cartridge deionization system. The resulting water conformed to ASTM Type I Reagent Grade Water standards (resistivity > 16.7 megaohm-cm).

Different dyes were used to improve the contrast between fluid phases. All physical properties were measured with the dyes present. Densities were measured with a Mettler/Par digital densitometer. Viscosities were measured with a Brookfield cone and plate viscometer. Refractive indices were measured with a Milton Roy Company refractometer. Interfacial tensions were measured using a spinning drop tensiometer.

2.1.1 Cocurrent Flow

Two fluid systems were used in the cocurrent flow experiments. Both fluid systems were allowed to come to equilibrium at room temperature before they were used in experiments. All cocurrent flow experiments were conducted at room temperature. The first fluid system was a ternary mixture of squalane, butanol, and water. This mixture was 30wt% squalane, 40wt% butanol and 30wt% water and
separated to form a three phase system. The lower phase contained mostly water and will be referred to as the aqueous phase. The middle phase was alcohol rich, the upper phase was predominantly squalane and will be referred to as the oleic phase. The equilibrium fluid phases were separated and various dyes introduced to enhance contrast. Red food coloring was added to the aqueous phase. Kodak Sudan Black dye was added to the alcohol rich phase. The interfacial tension (IFT) between the upper(U) and middle(M) phases was found to be 1.3 dyne/cm, and the interfacial tension between the U and lower(L) phase was 3.2 dyne/cm. This fluid system was chosen for its high oil viscosity, and the resulting high viscosity ratio between its U/L and U/M phases.

The second fluid system is a ternary oil-water-alcohol system. The three components are 2,3-dimethylpentane(DMP), n-propanol(NPA), and water(H₂O). This mixture has been studied by Giordano and Salter(23), Raney(24), and Elsik(16). The composition used for this work was 28.6wt% DMP and H₂O, and 42.8wt% NPA. Experiments using this fluid system were conducted at room temperature, approximately 22°C, where two phases exist at equilibrium. The lower phase is water rich and will be called the aqueous phase. The upper phase is oil rich and will be called the oleic phase. The interfacial tension for this system is 0.14 dyne/cm. This fluid system was chosen because its phase behavior and physical properties were well understood from previous research.
The physical properties of both fluid systems are listed in Table 2.1.

<table>
<thead>
<tr>
<th>Table 2.1 Fluid System Physical Properties</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fluid Composition</td>
</tr>
<tr>
<td>-------------------</td>
</tr>
<tr>
<td>SBW40</td>
</tr>
<tr>
<td>30.0wt% Squalane</td>
</tr>
<tr>
<td>40.0wt% Butanol</td>
</tr>
<tr>
<td>30.0wt% Water</td>
</tr>
<tr>
<td>DMP/NPA/H2O</td>
</tr>
<tr>
<td>28.6wt% DMP</td>
</tr>
<tr>
<td>42.8wt% NPA</td>
</tr>
<tr>
<td>28.6wt% H2O</td>
</tr>
</tbody>
</table>

2.1.2 Displacement Experiments

Various mixtures of DPM, NPA, H2O, toluene(TOL), and ethanol(ETOH) were used in the displacement experiments. Fluids in column 1 of Table 2.2 were contacted with fluids listed in column two.

<table>
<thead>
<tr>
<th>Table 2.2 Fluid Systems Used in Displacement Experiments</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fluid Composition</td>
</tr>
<tr>
<td>-------------------</td>
</tr>
<tr>
<td>40.0wt% TOL</td>
</tr>
<tr>
<td>60.0wt% NPA</td>
</tr>
<tr>
<td>100.0wt% DMP</td>
</tr>
<tr>
<td></td>
</tr>
<tr>
<td>70.0wt% DMP</td>
</tr>
<tr>
<td>30.0wt% NPA</td>
</tr>
<tr>
<td>70.0wt% TOL</td>
</tr>
<tr>
<td>30.0wt% ETOH</td>
</tr>
<tr>
<td>100.0wt% TOL</td>
</tr>
<tr>
<td></td>
</tr>
</tbody>
</table>
2.2 Experimental Apparatus

2.2.1 Flow Cell

The novel flow cell developed by Elsik was used in the majority of this work. For a historical review of flow models and a detailed description of the novel micromodel development see the work of Elsik(16).

Several additional flow cells were constructed for this work, but they were similar to those developed by Elsik. The only variation in construction was the glass bead packing arrangement. Two basic flow cell designs were used. The first type of flow cell was packed with uniform glass microspheres 109 μm (microns) in diameter. This arrangement provided a very uniform packing in a hexagonal close packed arrangement in the majority of the flow cell. The second type of flow cell was packed with a mixture of glass microspheres of two different size distributions. This packing arrangement was much less uniform than that of the 109 μm beads. A broader pore size distribution was obtained in this manner.

2.2.1.1 Micromodel Construction

A photograph of a typical flow cell is presented in Figure 2.1. Figure 2.1 shows the flow cell mounted on a glass slide using cable clamps. This is the normal configuration of the cell. Figure 2.2 is a schematic cross section of the flow cell containing uniform 109 μm glass beads. Figures 2.3(16) are photomicrographs of the flow cell. Figure 2.3a illustrates the hexagonal close packing found in the uniformly packed flow cell. Figure 2.3b is a higher magnification image of the same flow cell and illustrates the bi-layer of 109 μm glass beads found in the uniformly packed flow cell. Figure 2.3c is a photomicrograph of the randomly packed flow cell.
The micromodel is made from a 0.2x4x50 mm rectangular optical glass capillary supplied by Vitro Dynamics, Inc.. The capillary tube is sealed on one end with a 400 mesh woven polyester cloth supplied by Gilson Co. The polyester cloth is secured to the capillary using epoxy. After the polyester cloth is secured, a sleeve of 1/8" inside diameter(ID) tygon tubing is placed over the capillary and cloth and epoxied in place.

The 1/8"ID sleeve is then connected to a vacuum supply and the capillary is filled with glass microspheres (beads). This is accomplished by inserting the open end of the capillary into a container of the desired glass beads. To achieve a uniform packing arrangement the capillary is filled in stages. After partially filling the capillary the vacuum is removed and the beads are packed. The packing process entails tapping gently on the bottom of the 1/8"ID tubing while holding the flow cell in a vertical plane. Care is required here as insufficient tapping force will not pack the beads, and too much force will destroy any previous packing. The vacuum is then re-applied and more of the capillary is filled. The process is repeated until the capillary is full of beads.
Figure 2.1 Photograph of novel flow visualization cell, mounted on glass slide using cable clamps (16).
Figure 2.3a Photomicrograph of hexagonal close packing. 4X objective. Green reflected light. 109μm glass beads can be used for linear dimension calibration (16).
Figure 2.3b Bilayer of 109 \( \mu \)m glass beads in flow cell with 20x objective using green reflected and red transmitted light (16).
Figure 2.3c Random packing with non-uniform glass beads (16).
Once the capillary is filled another piece of 400 mesh polyester cloth is used to close the open end of the capillary. The cloth is attached with epoxy as before, and another 1/8"ID sleeve is placed over the capillary and cloth as before and epoxied in place. The most critical part of this operation is the attachment of the cloth to the capillary. The procedure that worked best required that the capillary be over packed, i.e. mound the glass beads out the end of the capillary, and then carefully place the cloth over the end of the capillary and attach it. This is necessary to prevent voids in the entrance region of the capillary.

The 1/8"ID tubing serves two purposes. The tubing allows convenient mounting of the flow cell onto a glass slide for ease of use and handling. The tubing also allows various fittings to be installed according to the type of experiment being performed.

When packing the capillary with the 109 μm beads, large regions of hexagonal close packing are readily formed. These regions are easily observed as they reflect light in an orderly manner. If the packing is performed carefully, large portions of the flow cell will be hexagonal close packed.

When packing the capillary to achieve a random packing a mixture of two different size distributions was used. The glass microspheres were obtained from Duke Scientific, which was also the supplier of the 109 μm beads. Various size distributions are available. In this work 40 - 70 μm and 85 - 120 μm distributions were used. The first attempt used a 50:50 mixture of these two distributions. Observation of this flow cell under the microscope revealed a very random packing with a large pore size distribution. However, the packing was prone to shifting if the flow cell was strongly agitated. The second attempt used a 25:75 ratio of the 40 -
70 μm and 85 - 120 μm distributions. Microscopic observation of this flow cell indicated a very random packing with a large pore size distribution. This packing arrangement was more stable than the first.

The two randomly packed flow cells were packed in a manner similar to the uniformly packed cells. The required amounts of each size distribution were mixed together in a small glass vial. This mixture was then used to pack the flow cell. The only difference in packing procedure was the use of a buffer region of the 109 μm beads at the flow cell inlet and outlet. This buffer region was intended to prevent any of the smaller beads from passing through the polyester cloth.

Once the flow cell has been packed and tubing sleeves have been installed, inlet and outlet tubes may be connected. Two small inlet tubes were fitted into the sleeve for the concurrent flow experiments (Chapter 3). Single inlet tubes were fitted into the sleeve for displacement experiments (Chapter 4).

The inlet tubing used by Elsik has several desirable characteristics. The tubing is translucent allowing visual tracking of the fluid interfaces as they progress toward the flow cell. Moreover, it is chemically inert and has walls rigid enough to prevent flow fluctuations when the flow cell is moved from one position to another under the microscope. The tubing used is a cross-linked ethyl vinyl acetate tubing manufactured by Micro-Line. Miniature plastic fittings were used to make the connections between sections of tubing. Luer lock fittings were used to connect the flow cell inlet tubing to the syringes containing the fluids.
2.2.1.2 Physical Parameters

Sizes of pore constrictions and pore bodies for the uniformly packed flow cell are reported by Elsik(16), and are of the order of those found in actual porous media. See Table 2.3 for values of porosity and permeability for each flow cell.

Porosity was measured by Elsik for the uniformly packed flow cell by weighing the glass beads packed into the flow cell. The density of the glass beads was supplied by the vendor allowing the volume of glass beads in the flow cell to be calculated, $V_b$. The internal volume of the capillary is readily calculated, $V_t$. Hence the porosity $\phi = 1 - V_b/V_t$. This method was also used to determine the porosity for the randomly packed flow cell.

For the uniformly packed flow cell, a theoretical calculation of the porosity is possible due to the well defined geometry of a hexagonal close pack. This calculation was performed by Elsik and yielded a value of 33.43%. The agreement with his measured value is excellent. A theoretical calculation for the randomly packed flow cell was not possible.

The absolute permeability of both flow cell types was measured in the following manner. The flow cell was fitted with a water manometer to measure the pressure drop across its length. Steady state single phase flow of water through the flow cell was established, and the pressure drop measured. Absolute permeability was then calculated from equation [1.2].
Table 2.3 Porosity and Permeability Values for Flow Cells

<table>
<thead>
<tr>
<th>Flow Cell</th>
<th>Porosity</th>
<th>Permeability (darcies)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Uniformly Packed</td>
<td>34.5%</td>
<td>16</td>
</tr>
<tr>
<td>Randomly Packed</td>
<td>46.2%</td>
<td>11</td>
</tr>
</tbody>
</table>

The porosity of the randomly packed flow cell is higher due to the looser packing obtained with the nonuniform diameter spheres. The permeability of the randomly packed flow cell is lower due to the more tortuous nature of the paths that fluids must travel.

2.2.2 Flow system

The experimental apparatus for this work is essentially the same as that reported by Elsik. The major components of the system are described below. Additional information can be found in reference (16).

A schematic diagram of the experimental apparatus is provided in Figure 2.4. The microliter syringe pump used for the majority of this work is a Harvard model 2274 dual syringe pump. This pump is designed to deliver very precise flow rates when used with Hamilton 1000 series gas tight syringes. The gear driven pump has thirty gear settings. Gear speed 1 corresponds to the highest, and gear speed 30 the lowest injection rate. For a given syringe size, there is approximately a 1.4 multiple in flow rate between consecutive gear settings. The flow rates which correspond to each gear setting and syringe size are tabulated in Table 2.4. Flow rates were converted to superficial velocities in common reservoir units of ft/day and are presented in Table 2.5. Interstitial velocities for both types of flow cell are presented in Table 2.6 and Table 2.7 respectively.
Flow Rates per Syringe (microliters/minute) for Harvard Microliter Syringe Pump Model 2274

<table>
<thead>
<tr>
<th>Gear Position</th>
<th>5 ml</th>
<th>10 ml</th>
<th>20 ml</th>
<th>30 ml</th>
<th>50 ml</th>
<th>100 ml</th>
<th>250 ml</th>
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<td>1</td>
<td>467</td>
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Table 2.5  Superficial (Darcy) Velocity (ft/day)

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Toward the completion of this work a new syringe pump was purchased. This pump is a Harvard Pump 22. This new style dual syringe pump is driven by a computer controlled electronic stepper motor. The pump 22 is a continuously variable pump and allows for finer selection of flow rates. With this pump flow rates are quickly changed without the disruption to the flow field associated with the gear driven syringe pump.
The syringes used for this work are Hamilton 1000 series gastight syringes of the 1ml, 2.5ml, 5ml, and 10ml sizes. The syringes are constructed of teflon plungers and precision bore glass barrels that provide smooth, non-pulsatile flow.

The fluids to be used in a particular experiment are drawn into separate syringes. The syringes are connected to the flow cell inlet tubing by luer lock fittings on the syringe and the inlet tubing. This connection insures that the inlet tubing is securely held in place during the experiment.

2.2.3 Optical Microscopy

The microscope used in this work is a Nikon Optiphot-Pol modified to use a High Pressure Xenon transmitted light source. Polarizing optics have been included as well as Hoffman Phase Contrast filters. Extra long working distance objectives of 4X, 10X, 20X, and 40X were used.

A review of optical microscopy can be found in reference (22). To improve contrast between the fluid pairs, one fluid phase usually contained red dye, and a blue transmitting filter was placed over the field lens.

Due to the internal reflections and refractions within the glass microspheres, high intensity light peaks are present within the flow cell. To help reduce these high intensity light peaks a diffuse filter was placed between the flow cell and the glass mounting slide. This provided better contrast between the fluid phases.

2.2.4 Enhanced Video System

The microscope was coupled to a high resolution Hamamatsu C1000-01 video camera. A 1X, 2.5X, 5X, or 10X projection lens can be inserted into the third
Figure 2.5 Video Patch Panel

Diagram of video patch panel with labels and connections:

- BTX
- 1/2" tape deck
- Time base corrector
- Monitor 1
- Frame memory
- Contour synthesizer
- Video scaler

Input ports:
- (1) Input from CCU or WS1
- (15) Input from digital USB microscope
- (17) Input from 3/4" WS M

Outputs:
- (19) Input from 3/4" WS M
- (25) Input from CCD
- (31) Input from WS4
- (21) Blank
- (23) Input from WS3
- (27) Blank
- (29) Blank

Legend:
- WS1 - Horizontal microscope
- WS2 - Miller group
- WS3 - Not used
- WS4 - Horizontal microscope
- In environmental room
microscope trinocular. This arrangement allows for simultaneous viewing through the microscope binocular eyepiece and on a video monitor. The video camera was connected directly to a camera control unit (CCU) with built in gain amplification and offset control for analog contrast enhancement.

The video signal can then be routed through many additional signal processing units which are described below. A video patch panel is provided to easily re-route the video signal path. This patch panel is schematically outlined in Figure 2.5. The patch panel introduces an incredible amount of flexibility into the signal processing path. The video signal can be completely re-routed from the front of the video component rack just by changing the patch panel connections. During normal operation each pair of vertical patch points are connected, and this is the normal signal path.

A BTX Cypher Time Code Generator is used to insert characters into the video signal. For this work the date the experiment was conducted along with the Time of Day, in 24 hour format, and an elapsed time counter were all inserted into the video signal. The BTX allows this information to become a permanent part of the experimental record. A Harris HVS 690 Digital Frame Synchronizer is used as a Time Base Corrector (TBC). The TBC is required to synchronize the video signal prior to using the digital image processor described below.

A FOR.A FM-60 Frame Memory can be used to freeze individual video frames for data analysis. The frame memory allows automatic sequential frame freezing with a user adjustable time interval between frames. This allows motion to be stopped at discrete intervals. A FOR.A IV-530 Contour Synthesizer allows gain amplification and edge enhancement. A FOR.A IV-550 Video Scaler allows objects
to be measured on the video monitor. The video scaler can be calibrated for each camera projection lens and microscope objective combination. Once calibrated the video scaler adds two vertical lines and one horizontal line to the video signal. The position of the lines can be controlled, and the distance between the vertical lines can be inserted into the video signal.

A Videotek TSM-5A wave form monitor is included in the system to allow the best analog video signal contrast to be obtained. With this unit the video signal is displayed on an oscilloscope and the optimum illumination and camera control unit settings can be determined. A Panasonic WJ-255R Video Switcher controls the input to the wave form monitor. The switcher allows all portions of the signal path to be observed on the wave form monitor.

Nine video monitors are located in the system to observe the video signal at different stages of enhancement. Four of the nine monitors are used with the digital image processor described in the next section.

Two Sony U-matic 3/4" industrial format VTRs are used to make permanent experimental records. The VO-5858 and VO-5800 recorders are connected to an RM-440 Automatic Editing Control Unit. This allows professional quality editing of experimental tapes, and the production of presentation tapes. The RM-440 is capable of assembly and insertion of audio and video signals.

A Dunn Instruments Model 635 Compact Color Camera unit produces a hard copy of the video output. It supports 8X10" Polaroid prints, and 35mm slides and prints.
2.2.5 Digital Image Processing

A Perceptive Systems PSICOM 237 Digital Image Processor (DIP) was used for contrast enhancement and data analysis. The DIP consists of an LSI-11/23 microcomputer (Digital Equipment Corp.), three 521x521x256 frame buffers each with a corresponding video display, a 14 MHz 8 bit video digitizer, and a trackball controller.

Additional system options include one multifunction board, three ITI(Image Technology Inc.) frame buffers, one four channel serial interface, an ITI arithmetic logic unit, 256k bytes of system RAM(Random Access Memory), and a 160 MB(Mega Byte) Winchester hard disk.

The microcomputer operates under the RT-11 operating system. The primary image processing software is PSIEXE. PSIEXE is a menu driven program which allows the user to digitize live or recorded video images. The digitized images are 512 pixel(picture element) x 480 pixel with 256 gray levels per pixel. This resolution provides excellent image quality while preserving fine detail. The gray levels range from black, zero, to white, 255.

The DIP is a very powerful analytical tool with many features. Some of the more useful ones are described below. Images may be scanned or digitized from live video or video tape. The digitized images may be stored on the Winchester disk for later use. There are many filter operations which enhance the contrast of the digitized image. Some of the filter operations available are the linear stretch which allows the contrast of an image to be increased over the entire gray scale, box filters, gradient filters, and Laplacian filters.
Some of the arithmetic operations include image Addition, Subtraction and Copy. There is also a calibration routine that calibrates the pixel spacing to provide for on screen measurement. Measurement routines include area, average diameter, perimeter, length and width. There is also an interactive graphics routine that allows for cutting and pasting images together as well as inserting text into an image. A paint brush routine used in conjunction with the trackball offers freehand sketching or tracing of objects in the image.

Perhaps the most useful feature of the DIP for this research is the Real Time Background Subtraction (RTBS) routine. This routine digitizes a frame from live video or tape. This image is then subtracted from the live video and the result displayed on a separate monitor. When the background is subtracted, all of the optical artifacts of the glass microspheres are removed, and any motion or change in fluid positions is readily detected.
3. TWO-PHASE STEADY COCURRENT FLOW EXPERIMENTS

3.1 Procedure

All experiments described in this chapter are conducted in a similar manner. The only differences between the experiments are in the fluids and the flow cells used. All experiments are conducted at a constant 1:1 injection ratio of the two fluids. The flow cell is configured with two inlet tubes. The procedure used is as follows. A separate gastight syringe is filled with each fluid and connected to a flow cell inlet tube. Fluid from each syringe is advanced up the injection tube to the flow cell entrance. The flow cell outlet tube is then connected to a vacuum pump, and both fluids are drawn in to the flow cell. This procedure generally removes most of the air from the flow cell. Any remaining air can be removed from the flow cell by injecting one of the fluids at a very high velocity.

Once the flow cell is full of fluid and the air has been removed, it is placed on the microscope stage. A visual check is performed with the microscope to confirm that all air has been removed from the system. If not, any remaining air is removed. The syringe pump is started at the desired flow rate and the flow field is allowed to stabilize. Once the flow is stabilized observations are recorded.

3.1.1 Explanation of Capillary Number Used

In the steady cocurrent flow work done by Elsik (16), the general capillary number defined by equation [1.1] was modified. The expression used by Elsik will be used in this work as well so that the results may be directly compared. The $N_{ca}$ defined by Elsik is:

$$N_{ca} = \frac{V\mu \nu}{\gamma} \quad \ldots \ldots \ldots \ldots [3.1]$$
Where \( V \) is the combined injection velocity of both fluids, \( \mu_w \) is the wetting phase viscosity, and \( \gamma \) is the interfacial tension between the fluid pairs.

The \( N_{ca} \) could be more suitably defined using the superficial velocity of the continuous phase as in eqn [1.1]. This is the wetting phase and \( v_w = V f_w \) (where \( f_w \) is the fractional flow of the wetting phase). In this work, \( f_w \) was held constant at 0.5 and eqn [3.1] will be a factor of two larger than the more fundamental \( N_{ca} \) defined by eqn [1.1]. In either case a form of the superficial velocity is generally used in the definition of the \( N_{ca} \) as it can be calculated without ambiguity. Use of the interstitial velocity may be more physically accurate, but generally requires detailed knowledge of the local saturations which are not commonly available in advance of the experiment. As we shall see below, however, some of our results can be best understood in terms of a capillary number based on the interstitial velocity. All velocities reported in this work are the combined velocity \( V \).

3.2 Uniformly Packed Micromodel

3.2.1 Experiments

Five experiments were conducted in this stage of the research. The motivation for these experiments was to explore the effect of the viscosity ratio, \( \mu_{nw}/\mu_w > 1 \), a matter not investigated by Elsik. Two experiments were performed using the SBW40 U/L fluid pair (see Table 2.1 for fluid properties and notation). Two experiments were performed using the SBW40 M/L fluid pair. One experiment was conducted using the DMP/NPA/H2O U/L fluid pair. Each experiment was conducted over a range of capillary numbers. The experiments will be discussed in the order in which they were performed.
Experiment 23 used the Squalane-Butanol-Water system upper and middle phases (SBW40 U/M) and the gear driven pump. The M phase was the wetting phase and the U phase was the nonwetting phase. The viscosity ratio, $\mu_{nw}/\mu_{wn}$, between the two phases was 7.52. The flow rate of the fluids was varied to cover a wide range of $N_{ca} (2 \times 10^{-4} < N_{ca} < 1 \times 10^{-3}$ as defined by eqn [3.1]). The qualitative behavior of the flow mechanisms observed is in excellent agreement with that reported by Elsik. At low flow rates stable Continuous Tortuous Paths (CTP) were observed. As the flow rate was increased, the CTP broke up into discrete ganglia which flowed in tortuous paths. As flow rate was increased further, the length of mobile ganglia decreased and the flow paths became less tortuous. Once the length of mobile ganglia decreased below one BD, the velocity of an individual ganglion was so high that detailed observation of the flow mechanisms was not possible. The length of mobile ganglia did appear to decrease further, but it was not possible to obtain length measurements.

Experiment 24 used the SBW40 system Upper and Lower phases (U/L) and the gear driven pump. The L phase was the wetting phase and the U phase was the nonwetting phase. The viscosity ratio between the two phases was 19.62. As with Experiment 23 the flow rate was varied over a wide range to cover the $N_{ca}$ range of interest ($1 \times 10^{-4} < N_{ca} < 1 \times 10^{-2}$). The flow mechanisms observed in this experiment were similar to those observed in experiment 23 and are in excellent qualitative agreement with those observed by Elsik.

Experiments 27 and 28 used the same fluid pairs and, in fact were identical to experiments 23 and 24 except that the former were conducted with the electronically controlled pump discussed further below. The qualitative behavior of the flow
mechanisms observed was identical to those of experiment 23 and 24 respectively. However there were quantitative differences.

Experiment 29 used the DMP/NPA/H2O system U/L phases and the electronically controlled pump. The viscosity ratio between the two phases was 0.6. This experiment was used to validate the accuracy of the electronically controlled injection pump. This experiment was performed to determine if the differences between experiments 23-24 and 27-28 were due to the electronically controlled injection pump. This experiment is a duplication of an experiment performed by Elsik(16) on the gear driven pump. The results obtained were essentially identical to Elsik’s, hence validating the accuracy of the electronically controlled injection pump.

The following remarks pertain to the use of the two syringe pumps. Shortly after the completion of experiments 23 and 24 a new electronically controlled pump was purchased. The gear driven pump used in experiments 23 and 24 was limited to discrete flow rates by its design. The electronically controlled pump offers a continuously variable flow rate. Over the range of flow rates of interest the gear driven pump did not provide enough discrete flow rates to yield as many data points as was desired. Also the gear driven pump greatly disturbs the flow field in the flow cell whenever pump speeds are changed. When gear speed is changed, the ram that pushes the syringe plunger will increase in speed until the new gear selected meshes. This rapid increase in injection rate drastically disrupts the flow field. The electronically controlled pump makes a much smoother transition between flow rates. I believe that this effect is more important for high viscosity ratio fluids as it takes longer for the flow field to reach steady state for these fluids. For these reasons, I am more confident in the data obtained with the electronically controlled pump for the SBW40 fluid system. Accordingly, the results obtained in experiments
27, and 28 will be emphasized. As noted above, the accuracy of the electronically controlled pump was verified for the DMP system which has a low viscosity ratio.

3.2.2 Results

The results of the five experiments detailed in section 3.2.1 along with representative data from Elsik's work are presented in Table 3.1. Column one contains the fluid pairs used for a given experiment along with the viscosity ratio for that experiment. Column two is the total superficial velocity $V$ for the experiment. Column three contains values of $N_{ca}$ calculated using equation [3.1]. Column four contains capillary numbers calculated from equation [3.1] multiplied by the viscosity ratio $\mu_{nw}/\mu_w$ (see discussion below). Column five contains the mean length of mobile ganglia observed at the given velocity.

The length of mobile ganglia were generally measured by digitizing or freezing a video image and measuring the ganglia using the video scaler or the digital image processor. This method of measurement is accurate to within a few microns. In some experiments it was not possible to measure mobile ganglia in this manner. In these experiments once the video was stopped, or an image digitized, it was not possible to see the interfaces between the ganglia and the continuous wetting phase. For these experiments the length of mobile ganglia were estimated either from the moving video or directly through the microscope during the experiment, the uniform 109$\mu$m glass beads were used as a reference to measure their length. This method is less accurate than measurement from a digitized image, and is accurate to within one half of a bead diameter.
Table 3.1 Summary of Cocurrent Flow Experiment Results

<table>
<thead>
<tr>
<th>Fluid System</th>
<th>Velocity (ft/day)</th>
<th>N\text{ca}</th>
<th>N\text{ca} &amp; \mu_{nw}/\mu_w</th>
<th>Ganglion Length (BD)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Experiment 23</td>
<td>26.2</td>
<td>2.1E-04</td>
<td>1.6E-03</td>
<td>5</td>
</tr>
<tr>
<td>SBW40 U/M</td>
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<td>3.0E-04</td>
<td>2.2E-03</td>
<td>4</td>
</tr>
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<td>\mu_{nw}/\mu_w = 7.52</td>
<td>51.5</td>
<td>4.2E-04</td>
<td>3.2E-03</td>
<td>3</td>
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<tr>
<td></td>
<td>72.0</td>
<td>5.9E-04</td>
<td>4.4E-03</td>
<td>2</td>
</tr>
<tr>
<td></td>
<td>101</td>
<td>8.2E-04</td>
<td>6.2E-03</td>
<td>1.5</td>
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<tr>
<td></td>
<td>141</td>
<td>1.1E-03</td>
<td>8.6E-03</td>
<td>1</td>
</tr>
<tr>
<td>Experiment 24</td>
<td>51.5</td>
<td>6.6E-05</td>
<td>1.3E-03</td>
<td>5</td>
</tr>
<tr>
<td>SBW40 U/L</td>
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<td>2.5E-03</td>
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</tr>
<tr>
<td>\mu_{nw}/\mu_w = 19.6</td>
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<td>1.8E-04</td>
<td>3.5E-03</td>
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</tr>
<tr>
<td></td>
<td>197</td>
<td>2.5E-04</td>
<td>5.0E-03</td>
<td>2</td>
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<tr>
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<td>2.9E-03</td>
<td>3</td>
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<td>167</td>
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<td>4.2E-03</td>
<td>1.5</td>
</tr>
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<td></td>
<td>183</td>
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<td>4.6E-03</td>
<td>1</td>
</tr>
<tr>
<td>Experiment 29</td>
<td>18.8</td>
<td>1.5E-03</td>
<td>9.6E-04</td>
<td>10</td>
</tr>
<tr>
<td>DMP/NPA/H2O</td>
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<td>2.3E-03</td>
<td>1.4E-03</td>
<td>6</td>
</tr>
<tr>
<td>\mu_{nw}/\mu_w = 0.6</td>
<td>36.7</td>
<td>3.0E-03</td>
<td>1.8E-03</td>
<td>4</td>
</tr>
<tr>
<td></td>
<td>135</td>
<td>1.1E-02</td>
<td>6.6E-03</td>
<td>1</td>
</tr>
<tr>
<td>Elsik T=30 C</td>
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<td></td>
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</tr>
<tr>
<td></td>
<td>54.2</td>
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<td></td>
<td>1.5</td>
</tr>
</tbody>
</table>

3.2.3 Correlation of the Results

The mechanisms of two phase steady flow that are being studied in this research are the transition from continuous tortuous paths at low $N_{ca}$ to ganglia flow
and the subsequent decrease in the length, L of mobile ganglia. By dimensional analysis, these mechanisms are functions of both $N_{ca}$ and the viscosity ratio $(\mu_{nw}/\mu_w)$. These two dimensionless groups should describe the transition in flow mechanisms for a particular porous medium.

Elsik’s work investigated only the effect of $N_{ca}$ (as defined by eqn [3.1]) on these flow mechanisms. The results of this research indicate that the viscosity ratio has a major effect as well. That is, neither the transition to ganglion flow nor the variation of ganglion size can be explained by variations in $N_{ca}$ alone, as the third column of Table 3.1 demonstrates. Indeed the data suggest that ganglion length, L, depends upon $(N_{ca})(\mu_{nw}/\mu_w)$ (fourth column of Table 3.1).

Dimensional analysis indicated that both $N_{ca}$ and viscosity ratio are important parameters in describing the flow mechanisms under investigation. It does not tell us what the functional dependence of these parameters is however. Elsik’s work explored the relationship between $N_{ca}$ and L. The consistency of results from the various experiments obtained in column 4 of Table 3.1 by multiplying the Nca defined by equation [3.1] by the viscosity ratio, as opposed to some other functional dependence, was determined empirically. The following derivation provides some rationale for this relationship.

Consider a trapped oil drop. Before the drop can be mobilized the pressure gradient in the continuous phase, $\nabla P_w L$, must exceed the capillary pressure $\Delta P_c$ holding the drop in place. As previously noted $\Delta P_c$ is primarily a function of the curvature of the oil-water interfaces. Since we do not know these radii of curvature we can not calculate $\Delta P_c$ from eqn [1.5]. Even though we can not calculate $\Delta P_c$ exactly, an approximation can be developed. If we assume that the interfaces
between the wetting and nonwetting phases are spherical we can use equation [1.8]. This assumption seems reasonable based on observations of the fluid in the flow cell. The maximum capillary pressure increase will occur when a ganglion's two interfaces are at their maximum and minimum curvatures. If the minimum and maximum radii of curvature for the model are defined as the radius of the pore throat, \( r_t \), and the radius of the pore body, \( r_b \) respectively, we can modify equation [1.8] to obtain:

\[
\Delta P_{c,\text{max}} = 2\gamma \left[ \frac{1}{r_t} - \frac{1}{r_b} \right] \quad \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots 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\ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldot
The following derivation develops the fractional flow relationships between viscosity and relative permeability. The volumetric flow rate for each phase may be written as the product of the velocity of each phase and the cross sectional area A.

\[ q_w = v_w A = \frac{-k_{rw} k}{\mu_w} \nabla p_w A \]  \hspace{1cm} \text{[3.5a]}

\[ q_{nw} = v_{nw} A = \frac{-k_{rnw} k}{\mu_{nw}} \nabla p_{nw} A \]  \hspace{1cm} \text{[3.5b]}

By expressing the nonwetting phase flow rate as \( q_{nw} = q_t - q_w \), and performing the subtraction, we obtain the following equation:

\[ q_w \left[ \frac{\mu_w}{k k_{rw}} + \frac{\mu_{nw}}{k k_{rnw}} \right] = q_t \mu_{nw} + \nabla p_c A \]  \hspace{1cm} \text{[3.6]}

Where \( \nabla p_c \) is the capillary pressure gradient \( \nabla (p_{nw} - p_w) \).

The fractional flow of the wetting phase can be defined as:

\[ f_w = \frac{q_w}{q_{nw} + q_w} = \frac{q_w}{q_t} \]  \hspace{1cm} \text{[3.7]}

Solving equation [3.6] for \( q_w \) using the result in equation [3.7], and simplifying, we obtain:

\[ f_w = \frac{1 + A (\nabla p_c) k k_{rnw}}{1 + \frac{\mu_t k_{nw}}{\mu_w k_{rnw}}} \mu_{nw} k_{rnw} \]  \hspace{1cm} \text{[3.8]}
If we make the assumption that the capillary pressure gradient \( \nabla p_c \) is negligible, equation [3.8] reduces to:

\[
f_w = \frac{1}{1 + \frac{\mu_w k_{rw}}{\mu_{nw} k_{rnw}}} \tag{3.9}
\]

This assumption is often made for mathematical simplicity in analyzing two-phase flow and should be reasonable provided that conditions do not change much along the flow cell.

All of the cocurrent flow experiments were conducted at an injection ratio of 1:1, hence \( f_w = 0.5 \). Reducing eqn [3.9] yields:

\[
\frac{\mu_w}{\mu_{nw}} = \frac{k_{rw}}{k_{rnw}} \tag{3.10}
\]

At high capillary numbers experiments have shown that

\((k_{rw}/k_{rnw}) \sim S_w/(1-S_w)\). If we substitute this expression into eqn [3.10] the following result is obtained:

\[
\frac{\mu_w}{S_w} = \frac{\mu_{nw}}{(1-S_w)} \tag{3.11}
\]

We can substitute eqn [3.11] into eqn [3.4] and the following result is obtained:

\[
N_{ca}^* = \frac{v_w \mu_{nw}}{\phi (1-S_w)} \tag{3.12}
\]

For large \((\mu_{nw}/\mu_w)\), \(S_w\) is small and \((1-S_w)\) is roughly constant. So

\[
N_{ca}^* = \frac{v_w \mu_{nw}}{\phi \gamma} \tag{3.13}
\]
As can be seen from eqn [3.13], $N_{ca}^*$ is just a constant multiple of $N_{ca}(\mu_{nw}/\mu_w)$, the parameter found empirically to be very good in correlating my data.

3.2.4 Discussion

The results obtained from this research are in fair quantitative agreement with the results found by Elsik. As illustrated in the Figure 3.1, the high viscosity ratio fluid pairs show some non-linearity as $(N_{ca})(\mu_{nw}/\mu_w)$ increases.

If we assume that $k_{nw}$ is approximately constant in equation [3.12], a reasonable assumption if $S_w$ (wetting phase saturation) does not vary greatly, a plot of $(N_{ca})(\mu_{nw}/\mu_w)$ vs $1/L$ should be linear. Figure 3.1 is a plot of $(N_{ca})(\mu_{nw}/\mu_w)$ vs $1/L$ for Elsik's data from Table 3.1 for the DMP/NPA/H2O system, and experiments 27, 28, and 29. As shown in Figure 3.1, the data for experiments 27 and 28 are linear for low $(N_{ca})(\mu_{nw}/\mu_w)$, but become nonlinear as $(N_{ca})(\mu_{nw}/\mu_w)$ increases. There are several possible reasons for this.

First, the data for the length of mobile ganglia for experiments 27 and 28 were taken in a different manner than those in experiment 29 and Elsik's work. The absolute velocity (superficial or interstitial) of the fluids in experiments 27 and 28 was much higher than in experiment 29 (with the exception of the last data point) or in Elsik's work. As the absolute velocity of the fluids increases, the ability to accurately measure the length of a mobile ganglion decreases. Normally the DIP is used to digitize an image where a mobile ganglion is present. Then the length of the ganglion is readily and accurately measured. As the absolute velocity increases, it becomes very difficult, if not impossible, to see the interfaces between the two fluids when an image is digitized. The motion of the mobile ganglia allows the eye and brain to
Experiments 27, 28, 29, and CME Work
Nca vs Inverse Ganglion Length

![Graph showing the relationship between Capillary Number and Inverse Ganglion Length (BD)].

- CME DMP L/U T=30
- Experiment 27
- Experiment 28
- Experiment 29

Figure 3.1
detect their presence, and measure them. But once the motion is stopped, the visual
cues necessary to identify the ganglia vanish. Since we can not digitize or freeze an
image and measure the length of an individual ganglion, measurements must be
taken from the moving video, or live through the microscope during the experiment.
This process is less accurate than measuring the length of a mobile ganglion on a
digitized image. This error in measurement will increase as velocity increases. This
error becomes more significant as L becomes small, as a small change in L produces
a larger change in 1/L. The data points for experiments 27 and 28 that show the most
deviation from the expected linear relationship occur at the higher velocities.

Second, for experiments 27 and 28 at the highest velocities it was not possible
to measure a mobile ganglion even during the experiment or from the moving video.
For these high velocities, one must find a trapped ganglion, wait until it is mobilized
by another ganglion colliding with it, and then measure the now mobile ganglion.
The ganglia measured in this manner would tend to represent a lower limit on the
length of mobile ganglia. Again this effect is more pronounced at higher velocities.
This factor will also contribute to the non-linearity of the plot for the higher velocity
data points.

A third possible reason is that $S_w$, hence $k_{nw}$ is not a constant as assumed
and varies slightly as $(N_{ca})(\mu_{nw}/\mu_w)$ increases. With the high viscosity fluids it seems
reasonable that as $(N_{ca})(\mu_{nw}/\mu_w)$ increases, $S_w$ increases. As $S_w$ increases $k_{nw}$
decreases causing the slope of the curve predicted by equation [3.12] to decrease as
$(N_{ca})(\mu_{nw}/\mu_w)$ increases. This behavior is exhibited by experiments 27 and 28 as
shown in Figure 3.1.
3.3 Randomly Packed Micromodel

There is one major difference between the randomly packed micromodel and the uniformly packed micromodel, aside from the broader bead size distribution and the packing. The uniformly packed micromodel was strongly water wet, i.e. the aqueous phase was always the wetting phase. The randomly packed micromodel was oil wet, so that the oleic phase was the wetting phase. This difference was somewhat surprising as the glass beads used in both models were supposed to be the same with the exception of size. Even after the randomly packed flow cell was carefully cleaned with acetone, ethanol and methanol, however the oil wet behavior still persisted. The beads used in the randomly packed micro-model were not treated in any manner that would alter their wettability.

3.3.1 Experiments 25 and 26

Two experiments were conducted with the randomly packed micromodel, experiments 25 and 26. Both experiments used the DMP/NPA/H2O fluid pair reported in Chapter 2. The experiments were conducted on the gear driven pump. The viscosity ratio ($\mu_{nw}/\mu_w$) for the fluid pair was 1.6. The experiments were duplicates of each other to verify the reproducibility of the data. The data for both experiments were, in fact, the same so only the data for experiment 25 will be reported.

3.3.1.1 Results

The results for experiment 25 are summarized in Table 3.2 below. Column one of the table identifies the fluid system for the experiment. Column two contains the velocity, $V$ for each data point. Column three contains the capillary numbers
calculated from equation [3.1]. Column four contains the capillary numbers in column three multiplied by the viscosity ratio, $\mu_{nw}/\mu_w$. Column five contains the length of the mobile ganglia observed at each velocity. These numbers are not multiples of actual bead diameters as have been reported in section 3.2.2. The reason for this is that there is no uniform scaling parameter for the randomly packed model. The ganglia were measured on the video screen using the video scaler, and their length divided by 109μm for easy comparison with the data in section 3.2.

The basic flow mechanisms observed in experiments 25 and 26 were the same as those observed in the preceding section, i.e. at low flow rates stable Continuous Tortuous Paths (CTP) were observed. As the flow rate was increased, the CTP broke up into discrete ganglia which flowed in tortuous paths. As flow rate was increased further, the length of mobile ganglia decreased and the flow paths became less tortuous. As the length of mobile ganglia decreased even further, the velocity of an individual ganglion was so high that detailed observation of the flow mechanisms was not possible. The length of mobile ganglia did appear to decrease further, but it was not possible to obtain length measurements.

Large mobile ganglia were not observed in this flow cell at any flow rate. In fact the length of mobile ganglia observed in this flow cell were approximately a factor of 2 smaller than those observed in the uniformly packed model at the same $N_{ca}$ with the same fluids. This result was expected due to the wide bead size distribution used in this flow cell. The wide size distribution increases the aspect ratio of the pore throat compared to the pore body. A high aspect ratio favors snap off, and any large ganglion would quickly be broken into several smaller daughter ganglia by this mechanism. Indeed, snap off was readily observable in the randomly
packed flow cell. The transition to ganglia flow occurred at roughly the same $N_{ca}$ as in the uniformly packed micromodel.

<table>
<thead>
<tr>
<th>Fluid System</th>
<th>Velocity (ft/day)</th>
<th>$N_{ca}$</th>
<th>$N_{ca} \mu_{nw}/\mu_w$</th>
<th>Ganglion Length (BD)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Experiment 25</td>
<td>18.8</td>
<td>9.5E-04</td>
<td>1.5E-03</td>
<td>2.29</td>
</tr>
<tr>
<td>DMP L/U</td>
<td>26.2</td>
<td>1.3E-03</td>
<td>2.1E-03</td>
<td>1.83</td>
</tr>
<tr>
<td></td>
<td>36.7</td>
<td>1.9E-03</td>
<td>3.0E-03</td>
<td>1.37</td>
</tr>
<tr>
<td>$\mu_{nw}/\mu_w = 1.6$</td>
<td>51.5</td>
<td>2.6E-03</td>
<td>4.2E-03</td>
<td>1.05</td>
</tr>
</tbody>
</table>

3.3.1.2 Discussion

Figure 3.2 is a plot of the data in column four in Table 3.2 vs 1/L. As can be seen from this figure the data are very linear. The slope of the line is different from that of experiment 29 in Section 3.2. This is expected as the permeability $k$, and the pore size distribution (related to $r_1$ and $r_p$) are different in this micromodel.

Figure 3.3 is a plot of $N_{ca}$ vs 1/L for experiments 25, 27, 28, 29 and Elsik's DMP/NPA system at 30°C. This figure has been provided so that all of the experimental results of this chapter may be more readily compared. Figure 3.4 is a plot of $N_{ca}$ vs 1/L for all of the experiments using DMP. The interesting feature of this figure is that the slopes for all of the DMP experiments are similar.

3.4 Conclusions

Based upon the results of this research, the mechanisms of steady two phase cocurrent flow for fluids of high viscosity ratio ($\mu_{nw}/\mu_w$) have been shown to be qualitatively similar to those of low viscosity ratio fluids. Capillary numbers calculated in the usual manner do not provide quantitative agreement in the flow
mechanisms observed. However, the correlation is reasonable if $N_{ca^*}$, based on the interstitial velocity, is used.
DMP Experiments
Nca vs Inverse Ganglion Length

Capillary Number

Inverse Ganglion Length (BD)

0.0E+00 0.1 0.2 0.3 0.4 0.5 0.6 0.7 0.8 0.9 1

0.0E+00 1.0E-03 2.0E-03 3.0E-03 4.0E-03 5.0E-03 6.0E-03 7.0E-03

CME DMP L/U T=30
Exp 25

CME DMP L/U T=30
Experiment 25

Figure 3.2
Experiments 25, 27, 28, 29, and CME Work
Nca vs Inverse Ganglion Length

Figure 3.3
Figure 3.4
4. DISPLACEMENT EXPERIMENTS

This chapter describes how the novel flow cell was used to observe nonequilibrium displacement experiments, and is preliminary in nature. Previously reported work using the novel flow cell, including Chapter 3 of this work and the work done by Elsik (16), used pre-equilibrated fluid phases. The focus of this chapter is the study of displacement experiments which exhibit mass transfer between the fluid phases.

Before proceeding further, it will be helpful to define two important terms that will be used in this chapter. The first is spontaneous emulsification. Spontaneous emulsification is the spontaneous formation of an emulsion near the interface of two fluid phases caused by the local supersaturation of one phase (17). In this work we are particularly interested in oil-in-water emulsions. When spontaneous emulsification of oil in water occurs, the interfacial area between oil and water is greatly increased. This should be useful in the biodegradation process, as anything that increases the surface area of the oil should increase its rate of biodegradation (this assumes that the rate of biodegradation is surface area dependent).

The second term that will be used in this chapter is Marangoni flow or Marangoni effect. Marangoni flow is spontaneously generated interfacial convection produced by interfacial tension gradients (19). The practical importance of this phenomenon is that it promotes interfacial turbulence and mixing and this increases mass transfer rates. It has been observed in this work that when both Marangoni flow and spontaneous emulsification are present, the rate of spontaneous emulsification is increased over that where Marangoni flow does not occur.
4.1 Nonequilibrium Effects and Displacement

The main purposes of the research reported in this chapter were to establish if it was possible to obtain significant spontaneous emulsification of oil in our flow cell, and to observe its effects on the displacement process. As stated above, this behavior is of interest because if spontaneous emulsification can be made to occur in a porous medium, it could speed up biodegradation of trapped oil even if the oil is not mobilized. The biodegradation of oil in a contaminated ground water aquifer is of great interest as a potential mechanism for cleanup.

Somewhat similar work which focuses on mobilization of trapped oil ganglia in porous media has been conducted by Lam, Schechter, and Wade (18). The experiments described in this chapter do not correspond directly to those in reference (18). However they are close enough to add some new insight.

Lam, Schechter, and Wade conducted flow experiments in a microvisual flow cell. Their flow cell was not the same construction as the novel flow cell reported in this work (see their paper for details). The main focus of Lam, Schechter, and Wade's work was the observation and identification of nonequilibrium effects on the mobilization of an isolated trapped oil ganglion. They confined their work to the ternary system cyclohexane, water, and n-propyl alcohol.

All of their work used nonequilibrium fluid pairs whose compositions were on the binodal curve. Their basic experimental procedure was to fill their flow cell with the cyclohexane rich mixture (it also contained some water and n-propanol since it was an equilibrium composition). The water rich fluid was then injected into the flow cell at a low velocity, so that the displacement would be incomplete and oil ganglia would be trapped. Then they switched the water rich phase to a fluid having a higher
alcohol concentration which was not in equilibrium with the trapped oil, and observed the results.

The purpose of their work was to isolate which nonequilibrium effects were important in the mobilization of a trapped oil ganglia. They focused their attention on dynamic interfacial tension (IFT), spontaneous emulsification caused by diffusion and stranding, mass transfer into an oil drop, and Marangoni flow.

Their most important results that apply to this work are as follows. First they observed that mobilization is primarily controlled by the capillary number as for pre-equilibrated phases. Second they found that in some cases the $N_{ca}$ required for mobilization was reduced by as much as a factor of two in the non-equilibrium case. Their explanation for this reduction in $N_{ca}$ was the onset of Marangoni flow. Third they calculated that at higher injection velocities, the concentration gradient of alcohol near the oil rich/water rich interface increases, and that this effect favors Marangoni flow. They also established a lower limit on velocity of around 5 to 6 ft/day required to produce the critical concentration gradient in their system. Below this velocity they believed that Marangoni flow did not occur. Fourth, they ruled out spontaneous emulsification due to diffusion and stranding as a mobilization mechanism although emulsification did occur in some experiments.

The fluid systems used in the work reported in this system were similar to that of Lam et al, i.e. oil-water-alcohol systems, the principal difference being that this work has mass transfer both into and out of the oil. The same nonequilibrium effects noted by Lam et al were present in this work. The nonequilibrium effects observed were spontaneous emulsification, Marangoni flow, varying IFT and mass transfer. While we did not calculate the alcohol concentration gradient near the oil phase-
aqueous phase interface, our flooding velocities were kept in the 5 ft/day range. At this velocity, it is likely that the difference between the equilibrium and nonequilibrium $N_{ca}$ required for mobilization is small. However, as indicated later, we do feel that Marangoni flow did occur in some of our experiments.

4.2 Procedure

The basic experimental procedure for all of the experiments reported on in this chapter is as follows. The micromodel packed uniformly with 109 $\mu$m diameter beads was used exclusively for these experiments. The flow cell was configured with a short Primary Injection Tube (PIT), and a long Secondary Injection Tube (SIT) (see Figure 4.1). In this configuration only one injection tube is connected to the flow cell inlet. The SIT is connected to the PIT by means of a "T" fitting located close to the flow cell inlet. This arrangement was found to be most suitable as it allows the fluid pairs to remain separated from each other until they are in the flow cell.

A typical experiment proceeds as follows. The fluid that is to be displaced, fluid A, is placed in a Hamilton Gas Tight syringe and connected to the PIT. The SIT is left open, and both the PIT and SIT are filled with fluid A. Once the SIT is full of fluid A, it is plugged. The flow cell outlet is connected to the vacuum pump, and fluid A is introduced into the flow cell. Once the flow cell is full of fluid A, it is removed from vacuum, and placed under the microscope. There the flow cell is visually inspected to insure that there is no trapped air in the pores. Any trapped air is removed at this time by injecting fluid A at a very high velocity. The syringe pump is
started, and several pore volumes of fluid are allowed to flow through the flow cell to ensure that it contains only fluid A.

The next part of the procedure requires close attention to detail and some practice to achieve success on a regular basis. A second Hamilton Gas Tight syringe is filled with the displacing fluid, fluid B. The displacing fluid is always the aqueous phase and contains red dye. The pump is stopped, and the syringe containing fluid A is removed. The second syringe containing fluid B is then attached to the PIT. This must be done carefully to prevent the two fluids from mixing. Mixing of the two fluids at this point is not desired. Since we are interested in studying nonequilibrium effects in these experiments, we want the fluids to mix in the flow cell, not in the tubing leading to the flow cell. We are looking for a step change in concentration inside the flow cell as opposed to a ramp or gradual change in concentration. An air bubble is intentionally allowed to separate the two fluids in the PIT. At this point the SIT is unplugged, and fluid B is slowly injected into the PIT. The SIT is left unplugged during this process so that the remaining fluid in the PIT will travel out through the SIT rather than through the flow cell.

When the air bubble that is separating the two fluids nears the "T" fitting, one of several things must happen. First one must look at the flow cell entrance. There is a gap inside the flow cell between the end of the PIT and the actual entrance to the bead pack (see Figure 4.2). This area is inside the 1/4" OD tygon tubing that connects the PIT and the bead pack. This area is called the plenum. There is almost always air trapped in the plenum from the filling process. Since it is buoyant, the air rests along the upper wall of the 1/4" tygon tubing and does not normally interfere with the experiment. However, if the air bubble that is between the two fluids and the air trapped in the plenum combine, often the resulting bubble is large enough
that it will enter the flow cell. When this happens, the entire flow field in the flow cell is disturbed and the experiment must be redone.

There are two principal ways to prevent this from happening. The first is to make sure that the air bubble that is separating fluids A and B is small enough that the combined air bubbles will not be too large and enter the flow cell. This sounds reasonable, but is difficult to achieve and is risky. By this time a fair amount of time has been invested which will be lost if too large an air bubble results from the combination. The preferred method is more difficult, but it is more reliable. When the air bubble separating the two fluids nears the "T", stop advancing it (stop injecting fluid B) before it reaches the "T". A third syringe is filled partially with air and connected to the SIT. The third syringe is used to manipulate the size and placement of the air bubble separating the two fluids. Once the syringe is connected to the SIT, the leading edge of the air bubble separating the fluids is pushed beyond the "T", and a small amount of this air is drawn into the SIT. Then fluid B is advanced further down the PIT. Once the trailing interface of the air bubble separating the fluids is down stream of the "T", some of the fluid B is drawn up into the SIT. This is a cautionary measure to prevent fluid A from coming back out of the SIT and mixing with fluid B.

The air bubble separating the fluids is advanced slowly down the PIT until the leading interface of the air bubble separating the two fluids contacts the air bubble in the plenum. Once these two air bubbles contact, they will coalesce. The combined bubble can then be slowly drawn back up the PIT. This is accomplished by slowly withdrawing fluid through the third syringe connected to the SIT. Care must be taken to avoid pushing air into the flow cell, or disturbing the flow cell with large pressure gradients.
Once the air bubble reaches the "T", most of the air is drawn up the SIT using the syringe connected to the SIT. A small air bubble should be maintained between the fluids. Once the correct size air bubble has been achieved (I found 1 to 3 mm in length worked best), the air bubble is advanced down the PIT toward the flow cell past the "T" using the syringe filled with fluid B.

The final step is to remove any air from the SIT. This is done by slowly injecting fluid B while carefully withdrawing fluid with the syringe connected to the SIT. Failure to remove this air makes the experiment more difficult to conduct. When there is a large amount of air in the SIT, it behaves in a manner similar to a compression tank in a water system. The pump spends several (or many depending upon the injection velocity) minutes pressurizing the air column in the SIT before any fluid is forced into the flow cell. This also makes it harder to estimate the actual pressure drop across the flow cell.

Once the air has been removed from the SIT, disconnect the syringe connected to the SIT and gently insert the plug. This must be done carefully as the air bubble separating the two fluids will be forced toward the flow cell during plug insertion. Now place the syringe containing the displacing fluid into the syringe pump and start the pump.

Depending upon the particular experiment, the fluid changing procedure outlined above may have to be repeated. For example experiments 16, and 21 each required the use of three fluids. In these experiments the flow cell was first filled with an oil, then water flooded (a displacement using pure water) to obtain trapped oil drops, and then a second displacement with a water/alcohol solution was conducted. During the second fluid change, it is important to be very careful while
changing the fluids and manipulating the air bubble so that none of the oil drops that were trapped by the water flood will be displaced. The flow cell was visually examined during the fluid changing process to insure that any trapped oil drops were not disturbed.

4.3 Experiments and Results

There are seven experiments reported in this chapter. A brief summary is provided in Table 4.1. The first column contains the number of the experiment. The second column contains the fluids used in the experiment. The fluid abbreviations are listed in the notes section following the table and in Section 2.1.2. The third column indicates the sequence in which the fluids were contacted. A '/' indicates that the fluids separated by the '/' were mixed together before they were contacted with another fluid. Fluids separated by a '-' were contacted together in that order. For example consider experiment 16. The flow cell was first filled with DMP, then water flooded with pure water (to trap regions of oil), then a mixture of water and NPA (30 wt %) was introduced into the flow cell. The fourth column contains the velocity used to displace the fluids. Where more than one velocity is listed, as in experiment 16, multiple displacements were involved. The first velocity corresponds to the first displacement, and the second velocity to the second displacement. The fifth column indicates the level of spontaneous emulsification that was observed. Each experiment is discussed in more detail below.
<table>
<thead>
<tr>
<th>Experiment</th>
<th>Fluids</th>
<th>Sequence</th>
<th>V (ft/day)</th>
<th>S.E.</th>
</tr>
</thead>
<tbody>
<tr>
<td>14</td>
<td>TP1, H2O</td>
<td>TP1-H2O</td>
<td>5.04</td>
<td>slight</td>
</tr>
<tr>
<td>16 (1)</td>
<td>DMP, H2O</td>
<td>DMP - H2O -</td>
<td>9.39</td>
<td>substantial</td>
</tr>
<tr>
<td></td>
<td>H2O/NPA(30W%)</td>
<td>H2O/NPA(30W%)</td>
<td>5.04</td>
<td></td>
</tr>
<tr>
<td>17 (1)</td>
<td>DMP/NPA(30W%)</td>
<td>DMP/NPA(30W%) -</td>
<td>5.04</td>
<td>slight</td>
</tr>
<tr>
<td></td>
<td>H2O</td>
<td>H2O</td>
<td></td>
<td></td>
</tr>
<tr>
<td>18</td>
<td>TE1, H2O</td>
<td>TE1-H2O</td>
<td>5.04</td>
<td>very vigorous</td>
</tr>
<tr>
<td>19</td>
<td>DMP</td>
<td>DMP-</td>
<td>5.04</td>
<td>substantial</td>
</tr>
<tr>
<td></td>
<td>H2O/NPA(30W%)</td>
<td>H2O/NPA(30W%)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>20</td>
<td>Toluene, WP1 (2)</td>
<td>Toluene -</td>
<td>5.04</td>
<td>vigorous</td>
</tr>
<tr>
<td></td>
<td></td>
<td>WP1</td>
<td></td>
<td></td>
</tr>
<tr>
<td>21</td>
<td>Toluene, H2O, WP1 (2)</td>
<td>Toluene-H2O -</td>
<td>12.6</td>
<td>vigorous</td>
</tr>
<tr>
<td></td>
<td></td>
<td>WP1</td>
<td>5.04</td>
<td></td>
</tr>
</tbody>
</table>

NOTES
(1) The video room felt warm. Perhaps > 25°C.
(2) Complete oil displacement was obtained.
TE1 Toluene/Ethanol(30W%).
TP1 Toluene/normalpropanol(NPA)(60W%).
WP1 Water/normalpropanol(NPA)(45W%).
/ Denotes that fluids indicated are mixed together.
- Denotes order in which fluids were contacted.

Experiment 14 used the TP1 solution displaced by pure water. The injection velocity for the displacing fluid was 5.04 ft/day. A slight amount of spontaneous emulsification was observed. There were large areas of trapped TP1 remaining after the experiment had reached equilibrium. These fluids were contacted in a test tube prior to the displacement experiment and Marangoni flow was observed in the test tube. No evidence of extensive Marangoni flow was observed in the flow cell. However, some Marangoni flow likely developed since one small oil drop (approx 2
\( \mu m \) was observed kicking or twitching. This behavior was very short lived, on the order of 2 to 3 seconds. No other 'kicking' drops were found.

In the ternary system in this experiment NPA is preferentially soluble in toluene compared to water. Little or no spontaneous emulsification would be expected based on the diffusion path analysis presented by Ruschak and Miller (17). None was observed in their work with an initial alcohol content of 40 wt% in the toluene phase. However the higher alcohol content used here could cause a little emulsification to occur. In any case the results of this experiment are in basic agreement with Ruschak and Miller's predictions.

Experiment 16 used DMP displaced by pure water which, in turn, was displaced by a mixture of water and NPA (30 wt%). The intent of this experiment was to see if trapped oil ganglia would be mobilized when contacted by the water/NPA solution. The first water flood was conducted at 9.39 ft/day and resulted in large areas of trapped oil. The second displacement was conducted at 5.04 ft/day. There was a substantial amount of spontaneous emulsification observed during this displacement. This spontaneous emulsification was time dependent in nature. As the water/NPA mixture was being injected into the flow cell, the rate of spontaneous emulsification was observed to initially be zero, then increase to a maximum, and subsequently decrease back to zero. At the low injection velocity used here, there is sufficient time before the main water/NPA front moves into the flow cell that diffusion and/or dispersion are important. In this fluid system, the alcohol (NPA) is preferentially soluble in water compared to the oil (DMP). Between 60% and 70% of the oil trapped by the first water flood was mobilized as emulsified drops. As the second displacement progressed there was some movement and rearrangement of the trapped oil ganglia, but the large trapped ganglia were not mobilized. These
fluids were contacted in a test tube prior to the displacement experiment and
Marangoni flow and spontaneous emulsification were observed in the test tube.
Slight spontaneous emulsification is predicted by the diffusion path analysis
presented by Ruschak and Miller.

Experiment 17 used a mixture of DMP and NPA (30 wt%) displaced by
water. The displacing fluid velocity was 5.04 ft/day. Only a slight amount of
spontaneous emulsification was observed in this experiment. In this fluid system the
alcohol (NPA) is preferentially soluble in water compared to the oil (DMP). There
was no Marangoni flow in this system, and some spontaneous emulsification is
predicted by the diffusion path analysis. The relatively low level of spontaneous
emulsification observed is attributable to the lack of Marangoni flow.

Experiment 18 used a mixture of TOL and ETOH (ethanol) (40 wt %)
displaced by water. The displacement velocity was 5.04 ft/day. Previous work with
this system (17, 20) indicated that spontaneous emulsification and Marangoni flow
were both observed when these fluids were brought into contact. When the
displacing fluid and the fluid to be displaced made contact in the flow cell plenum
there was rapid mixing and what appeared to be spontaneous emulsification. It is
very difficult to observe much detail concerning what happens in the plenum, but the
mixing was quite noticeable. The fluid in the plenum took on a very cloudy
appearance indicating that an emulsion had been formed. This fluid system
produced the most spontaneous emulsification of any tested. The spontaneous
emulsification was especially vigorous in nature. However, even with this very
vigorous spontaneous emulsification, not all of the oil was displaced. After
equilibrium was reached there were still large regions of trapped oil remaining.
Experiment 19 used the same fluid system as experiment 16, the only difference being that the pure water flood performed in experiment 16 was omitted in this experiment. In experiment 16 we were interested in seeing if trapped oil drops would be mobilized when contacted by the water/NPA mixture. Often it is more difficult to mobilize trapped oil ganglia than it is to displace a region saturated with oil. As in experiment 16 a substantial amount of spontaneous emulsification was observed. The time dependent nature of the spontaneous emulsification noted in experiment 16 was not observed here. This seems reasonable as in experiment 16 the alcohol could diffuse through the pure water ahead of the main water/NPA front. In this experiment there was no similar water pathway for diffusion. This displacement experiment did not produce a complete displacement of the oil. However about 80% of the oil was displaced.

Experiment 20 used TOL and a water/NPA (45 wt%) mixture. The displacement velocity was 5.04 ft/day. In this fluid system the alcohol (NPA) is preferentially soluble in the oil (TOL). Marangoni flow and spontaneous emulsification were observed in test tube contacting experiments. Spontaneous emulsification is predicted by the diffusion path analysis. This fluid system produced a very interesting displacement. After the water/NPA mixture had been connected to the flow cell and had entered the flow cell plenum, there was no noticeable change in the fluid in the plenum. As time progressed regions of emulsified drops began to appear in the TOL, but the main displacement front had not entered the visible part of the flow cell. These drops were only found in the entrance region of the flow cell. They were most likely emulsified drops of water in the TOL. As time passed, more of these drops began to appear, and their presence was noted further into the flow cell. Then the main displacement front moved into the flow cell. There was a
tremendous amount of activity as the front passed through. There was a great deal of spontaneous emulsification, and large regions of TOL were trapped. This sequence was repeated over and over in different parts of the flow cell. One could jump ahead of the displacement front and observe the same sequence of events. After the entire flow cell had been swept by the displacement front, the entrance region of the flow cell was checked to see if there were still regions of trapped TOL. The regions of trapped TOL were being broken up into smaller ganglia, and small drops ranging in size from 5 to 10 \( \mu m \) to 600 to 800 \( \mu m \). From the behavior of these now mobile ganglia, it appears that ultra low interfacial tension existed between the fluid phases. The interfaces between the phases were very flexible and would readily deform as the ganglia passed through pore constrictions. During a period of approximately 30 minutes this behavior was observed throughout the entire flow cell. The result was a total mobilization of all of the previously trapped TOL.

Experiment 21 used the same fluids as experiment 20, and was performed in the same general manner. The difference was that an intermediate water flood was added in experiment 21 to trap large regions of TOL before the water/NPA mixture was injected. The results of this experiment were essentially the same as those reported for experiment 20. The time dependent behavior of the displacement process was still present, and the sequence of events was the same. This displacement also mobilized 100% of the trapped TOL.

4.4 Discussion and Recommendations for Further Work

As stated in the beginning of this chapter, the work presented here was preliminary in nature. The nonequilibrium effects on the displacement and mobilization of oil from a porous media are complex. The work presented in this
chapter contributes to the understanding of this process, and identifies other areas where more work is necessary.

4.4.1 Discussion

In general, the work presented in this chapter is consistent with the work of Lam, Schechter, and Wade. Dynamic IFT is an important factor in nonequilibrium displacements. Any phenomenon that decreases the IFT at a given velocity, and increases the capillary number should promote higher oil recovery. The results of this work indicate that the dynamic IFT can be decreased to a point where all of the oil is mobilized in certain oil-water-alcohol systems. As the IFT becomes lower, interfaces are more deformable, and some smaller oil drops may be produced by the break-up of moving ganglia. The effects of dynamic IFT could be studied further to more rigorously explore its effect.

Spontaneous emulsification is an important phenomenon in nonequilibrium displacement experiments. In the work reported in this chapter, there was at least some spontaneous emulsification in most of the experiments. Lam, Schechter, and Wade dismissed its importance in increasing the mobilization of trapped oil ganglia. In my experiments where spontaneous emulsification was most vigorous, more oil was displaced. It was not possible to tell if particular isolated trapped ganglia were mobilized, and certainly there were still large regions of trapped oil in most of the experiments. But it was also apparent that a fair amount of oil was displaced from the flow cell in the form of emulsified oil drops carried in the aqueous phase. Spontaneous emulsification caused by diffusion and stranding may not be the critical factor in the mobilization of trapped oil ganglia, but it certainly did account for an
overall increase in the amount of the oil displaced from the flow cell in experiments where spontaneous emulsification was pronounced.

As previously noted, spontaneous emulsification in and of itself may be enough to enhance biodegradation through the increase in surface area associated with spontaneous emulsification. This work demonstrates that spontaneous emulsification can be maximized by choosing a system where the diffusion path predicts spontaneous emulsification, and where Marangoni flow also occurs. Consider experiments 14, 17, 18, and the information contained in Table 4.2. As can be seen from Table 4.2, the system having both Marangoni flow and spontaneous emulsification is experiment 18. This system had the most vigorous spontaneous emulsification of any examined. Diffusion path analysis predicts some spontaneous emulsification in experiment 17, but little was observed because there was no Marangoni flow. Similarly, little spontaneous emulsification was observed in experiment 14 where there was Marangoni flow but little or no spontaneous emulsification predicted by the diffusion path.

<table>
<thead>
<tr>
<th>Experiment</th>
<th>Marangoni Flow</th>
<th>Spontaneous Emulsification Predicted</th>
<th>Spontaneous Emulsification Observed</th>
</tr>
</thead>
<tbody>
<tr>
<td>14</td>
<td>yes</td>
<td>no</td>
<td>slight</td>
</tr>
<tr>
<td>17</td>
<td>no</td>
<td>yes</td>
<td>slight</td>
</tr>
<tr>
<td>18</td>
<td>yes</td>
<td>yes</td>
<td>very vigorous</td>
</tr>
</tbody>
</table>
As noted above, there were few direct signs of Marangoni flow in the flow cell, but I believe that some Marangoni flow did occur with the resulting increase in emulsion formation. This conclusion differs somewhat from that of Lam et al, who considered that Marangoni flow was not important in their system at the low flooding velocities used in this work. However, our velocities are only slightly below their calculated values for the initiation of Marangoni flow. I feel that Marangoni flow did occur in my experiments, although I do agree with Lam et al that Marangoni flow would be more pronounced at higher velocities. More work could be done to better define the role of spontaneous emulsification and Marangoni flow in nonequilibrium displacements.

Mass transfer is certainly an important phenomenon in any study of nonequilibrium effects. Lam, Schechter, and Wade determined that interfacial turbulence was caused by sufficiently high concentration gradients inside of the trapped oil ganglia. When interfacial turbulence begins, it should increase the mobilization of trapped oil ganglia. While no effort was made during the this work to obtain precise values of the $N_{ca}$ required for mobilization of trapped drops, the results obtained are basically consistent with those of Lam et al. However, in this work mass transfer was not limited strictly to transfer from the continuous water rich phase into the oil rich phase. In experiment 17 for example, the alcohol was initially placed in the oil, but as the alcohol is preferentially soluble in water we expect mass transfer to occur from the oil into the water. As the work reported in this chapter was conducted at low velocities, the mobilization of trapped oil ganglia, and the presence and rate of spontaneous emulsification were both time dependent as expected.
On a macroscopic level Marangoni flow can generally be detected when suitable pairs of fluids are contacted. When a drop of one fluid is carefully lowered into the second, the drop will twitch and kick if there is Marangoni flow. Marangoni flow was observed in test tube contacting experiments with most of the fluid pairs used in this work. In these contacting experiments it was observed that there was considerably more spontaneous emulsification when Marangoni flow was present then when it was not. In the displacement experiments reported in this chapter we were not able to directly observe additional flow at the interface which normally accompanies Marangoni flow. During displacement of one fluid by another interfaces are in motion and the Marangoni effect may not be visible in the flow cell. The extensive spontaneous emulsification observed in many experiments is probably indirect evidence that Marangoni flow was occurring.

In summary, it seems that anything that promotes the lowering of IFT or increases spontaneous emulsification is a desirable effect regarding the mobilization and/or removal of oil from porous media. Lowering the IFT can allow the oil to be mobilized or removed from the porous medium by normal flow related mechanisms. Some oil will also be mobilized in the form of emulsified oil drops in the aqueous phase. Even if all of the oil is not displaced or mobilized, an increase in spontaneous emulsification can enhance the rate of biodegradation through the accompanying increase in surface area resulting from the spontaneous emulsification. Predictions made using the diffusion path analyses presented by Ruschak and Miller regarding the likelihood of spontaneous emulsification occurring can help guide the choice of fluids to be used with a given oil contaminant.
4.4.2 Further Work

Some interesting topics to explore in future work would include the effect of velocity on the amount of spontaneous emulsification. Once spontaneous emulsification is shown to occur for a given fluid system, it should be possible to increase the rate of spontaneous emulsification by increasing the velocity of the displacement. When bulk fluids are contacted and spontaneous emulsification occurs, the rate of spontaneous emulsification can be increased by mixing the bulk fluids. Increasing the displacement velocity should have a similar effect.

Also of interest would be to repeat these experiments using aqueous phase compositions which are on the coexistence curve. This would allow the calculation of the non-equilibrium $N_{ep}$ and the final IFT. It should also promote spontaneous emulsification of water in the oil phase according to the Ruschak and Miller scheme. An example of this could be experiments 16 and 19. If the flooding phase composition is changed to contain a small amount of DMP, the diffusion path analysis would predict the formation of a water-in-oil emulsion as well as the oil-in-water emulsion already observed. The ability to emulsify the aqueous phase in the oil phase may also be useful in insitu biodegradation.
5. THEORETICAL MODEL

This chapter describes the development of a numerical model which simulates the flow of a oil ganglion through an idealized pore. The usual model of relative permeability adequately describes two phase flow when both fluid phases are continuous. In the work presented in chapter 3 and (16) it was observed that at higher \( N_{ca} \)'s one phase becomes discontinuous. This model is a first step toward modeling the flow behavior of discontinuous fluid drops. A description of the model is presented, followed by the formulation of the governing equations. Several numerical examples are considered, and the results presented and discussed. Also included is a discussion on possible extensions to the model that would improve its ability to more fully describe the conditions found in real porous media.

5.1 Development of Model

This section describes the physical parameters of the model used, and the assumptions made in the formulation of the model.

5.1.1 Description of the Model

The model that has been developed consists of a series of pore throats or constrictions. At any given point in time some of these pore throats will be filled with only water, some with only oil, and others partially filled with water and oil. One such pore throat is shown in Figure 5.1. The model parameters are defined as follows: \( R_o \) is the left hand radius of curvature of the oil ganglion; \( r_1 \) is the inside radius of the capillary tube; \( R_{or} \) is the radius of curvature of the right hand oil-water interface; \( x_1 \) is the leading edge of the right hand interface; and \( L \) is the length of the capillary tube.
Initial Condition

Figure 5.1
The physical nature of the flow of an oil ganglion through the model capillary tube allows the flow process to be broken into discrete stages. These stages are outlined in the next section.

5.1.2 Physical Assumptions

To simplify the development of the model, the following assumptions have been made. First, it is assumed that the tube is completely water wet. This means that the oil will not wet the capillary tube. This condition requires that the contact angle between the capillary tube wall and the oil ganglion be zero degrees when measured through the water. Second, it is assumed that all oil-water interfaces are spherical, or spherical caps. Third, it is assumed that there is no contact angle hysteresis. That is to say that the advancing and retreating contact angles are equal. The fourth assumption is that the volume of the oil drop remains constant. The fifth assumption is that all fluids involved are constant property, Newtonian fluids. The sixth assumption is that the volume of the drop is greater than the volume of the tube.

5.1.3 Breakdown of the Flow Process into Stages

Flow of the oil ganglion through the capillary tube has been divided into six stages. The divisions between the stages are described below. These stages are detailed in the Figures 5.2 through 5.7.

5.1.3.1 Stage 1

The starting point for Stage 1 is the initial contact of the oil ganglion with the capillary tube. After initial contact, the outside radius $R_O$ will decrease from its initial value $R^*$. As $R_O$ decreases, the volume inside the capillary tube will increase,
Stage 1

Figure 5.2
Stage 2

Figure 5.3
Stage 3

Figure 5.4
Stage 4

Figure 5.5
Stage 5

Figure 5.6

X1
Figure 5.7

Stage 6

$r_1$

10.0000

15.8884

$L$

$X_1$

$R_3.0000$
and the value of \( R_{Or} \) will decrease from \( R^* \) to \( r_1 \). The end point of stage 1 is the point where the volume of the spherical cap inside the capillary tube is equal to the volume of a hemispherical cap of radius \( r_1 \). This value follows from assumptions one, two and four.

5.1.3.2 Stage 2

The beginning point for stage 2 is the end point of stage 1. During stage 2, the hemispherical cap formed at the end of stage 1 will propagate down the capillary tube. The end point for stage 2 is when the contact line between the oil drop and the capillary tube reaches the end of the tube, or when \( x_1 = L + r_1 \). During stage 2, \( R_O \) will decrease continuously, and \( R_{Or} \) will remain constant at \( r_1 \). See Figure 5.3.

5.1.3.3 Stage 3

The starting point for stage 3 is the end point of stage 2. During stage 3 the hemispherical cap on the right hand side of the capillary tube will grow until \( R_O = R_{Or} \). This is the end point for stage 3. See Figure 5.4.

5.1.3.4 Stage 4

The beginning point for stage 4 is the end point of stage 3. During stage 4, \( R_O \) will decrease until it reaches the value \( r_1 \). \( R_{Or} \) will increase continuously during this stage. See Figure 5.5.

It is interesting to note that the constant drop volume and no contact angle hysteresis assumptions force symmetry on the model. The decrease of \( R_O \) during this stage is analogous to the growth of \( R_{Or} \) during stage 3. This inherent symmetry was exploited during the numerical simulation.
5.1.3.5 Stage 5

The beginning point of stage 5 is the end point of stage 4. During stage 5 the contact line of the hemispherical cap on the left hand side of the capillary tube moves from its initial position at \( x = 0 \), to its final position at \( x = L \). During this stage the value of \( R_O \) remains constant and equal to \( r_1 \), the value of \( R_{or} \) will increase continuously. See Figure 5.6.

Again, the constant drop volume assumption requires that the increase of \( R_{or} \) in this stage be analogous to the decrease of \( R_O \) in stage 2.

5.1.3.6 Stage 6

The beginning point of stage 6 is the end point of stage 5. During this stage, the volume inside the capillary tube will decrease from that of a hemispherical cap until it reaches the value corresponding to the initial condition. This requires that \( R_O \) increase in value from \( r_1 \) to \( R^* \). The value of \( R_{or} \) will increase continuously during this stage until it attains the value of \( R^* \). See Figure 5.7.

The constant volume assumption requires that the increase in \( R_O \) and \( R_{or} \) be analogous to the decrease in \( R_{or} \) and \( R_O \) in stage 1, respectively.

5.2 Formulation of Equations

In this section, the governing equations of flow and capillary pressure are presented. The derivation of the dimensionless groups used in this work are also presented.
5.2.1 Governing Equations Defined

The model system chosen most closely resembles that of laminar flow in a cylindrical tube. The solution of the equation of motion is well known in this case, i.e. Poiseuille flow. The expression for the average velocity is given by (12):

\[ v = \frac{(P_0 - P_L) r_1^2}{8 \mu L} \quad \cdots \cdots \cdots \cdots [5.1] \]

For this model, \( P_0 - P_L \) is replaced by \( \Delta P \). \( \Delta P \) is defined as the total pressure drop due to viscous flow through a water filled tube. In the numerical calculations that follow, \( \Delta P \) is a parameter that will be specified. If we invoke assumption five, then [5.1] becomes:

\[ v_w = \frac{(\Delta P) r_1^2}{8 \mu_w L_w} \quad \cdots \cdots \cdots [5.2] \]

One can show that eqn [5.2] is consistent with Darcy's law for flow in porous media, where the permeability \( k \), is equal to \( (r_1^2)/8 \) (1).

\[ v_w = \frac{k (\Delta P)}{\mu_w L_w} \quad \cdots \cdots \cdots [5.3] \]

Here \( v_w \) is the average water velocity. When using eqn [5.3] we have neglected the dimensions of the meniscus in computing \( L_w \) as \( L >> r_1 \). A similar equation also holds for the flow of oil in the same tube.

5.2.2 Derivation of Capillary Pressure Equation

The actual pressure drop across a flowing oil ganglion in the model will be composed of two parts. The first part will be the pressure drop due to viscous flow, and the second part will be the pressure drop due to capillary pressure. The pressure
drop due to viscous flow may be obtained from equation [5.2] and the standard continuity equation. The pressure drop due to capillary pressure will be derived.

The capillary pressure is derived from the Laplace equation in the usual manner. First we assume that the normal components of viscous stresses are negligible at the low flow rates under consideration. The form of the Laplace equation, for spherical interfaces is:

\[ P_o - P_w = \frac{2\gamma}{R} \]  \hspace{1cm} \text{[5.4]}

where the subscripts o, and w denote oil and water respectively. R is the radius of curvature of the oil-water interface. From eqn [5.4], it can be seen that there is a pressure jump across a curved interface. So for our model, there will be a pressure jump across each oil-water interface. The capillary pressure jump across the drop is basically the sum of the pressure drops across the two oil-water interfaces and can be expressed by:

\[ \Delta P_c = 2\gamma \left( \frac{1}{R_{or}} - \frac{1}{R_o} \right) \]  \hspace{1cm} \text{[5.5]}

Here the subscript c denotes capillary pressure, and \( \gamma \) is the interfacial tension between oil and water.

5.2.3 Derivation of Dimensionless Groups

For the evaluation of numerical results, it is desirable to have quantities expressed in dimensionless groups. If the equations can be expressed in dimensionless terms, only the dimensionless parameters and variables need to be varied to study new conditions. For this model there are five dimensionless groups of interest. These dimensionless groups were created using the initial drop radius, \( R^* \), as a scaling factor. Table 5.1 lists the dimensionless variables used.
<table>
<thead>
<tr>
<th>Variable</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>B</td>
<td>Dimensionless curvature of the left hand Interface (R_o/R*)</td>
</tr>
<tr>
<td>C</td>
<td>Dimensionless curvature of the right hand Interface (R_0r/R*)</td>
</tr>
<tr>
<td>Z</td>
<td>Ratio of inside tube diameter to initial drop radius (r_1/R*)</td>
</tr>
<tr>
<td>Y</td>
<td>Length of capillary tube divided by initial drop radius (L/R*)</td>
</tr>
<tr>
<td>W</td>
<td>Dimensionless position of the right hand interface (x_1/R*)</td>
</tr>
<tr>
<td>ΔPe*</td>
<td>Dimensionless capillary pressure (1/C)-(1/B)</td>
</tr>
<tr>
<td>Nca</td>
<td>Capillary number ( (r_1^2ΔP)/(γL) )</td>
</tr>
<tr>
<td>v*</td>
<td>Dimensionless velocity of the right hand interface see eqn [5.11]</td>
</tr>
<tr>
<td>l_0</td>
<td>Dimensionless oil saturation in tube ( (L_o/L) )</td>
</tr>
<tr>
<td>μ*</td>
<td>Viscosity Ratio</td>
</tr>
<tr>
<td>t*</td>
<td>Dimensionless time step</td>
</tr>
</tbody>
</table>

The first dimensionless group derived was the dimensionless capillary pressure. As can be seen from eqn [5.5], the capillary pressure is primarily a function of the model geometry. It is most useful to form the dimensionless capillary pressure in a manner such that it only depends upon geometry and not any fluid properties. This dimensionless parameter is readily determined from eqn [5.5], and is given by:
\[ \Delta P_c^* = \frac{(\Delta P_c) R_o^*}{2\gamma} = \frac{R^*}{R_{or}} - \frac{R^*_o}{R^*} \] ........................ [5.6]

By defining two dimensionless variables, \( B = R_o/R^* \) and \( C = R_{or}/R^* \) eqn [5.6] becomes:

\[ \Delta P_c^* = \left[ \frac{1}{C} - \frac{1}{B} \right] \] ........................ [5.7]

The second dimensionless group derived was a dimensionless distance parameter. This parameter tracks the location of the right hand interface \( x_1 \), and is given by \( W = x_1/R^* \). The actual equation that calculated \( W \) is dependent upon the stage under consideration, and the details are given in Appendix A. By looking at the geometry of the model, general values of \( W \) may be established. For example in stage 1 \( W \) will start at the initial value of the small spherical cap inside the tube when contact is made, and will increase to \( r_1/R^* \) (Z).

The third dimensionless group is the dimensionless capillary number given by:

\[ N_{ca} = \frac{v_w \mu_w}{\gamma} \] ........................ [5.8]

Here \( v_w \) can be replaced by eqn [5.2]. Then eqn [5.8] becomes:

\[ N_{ca} = \frac{r_1^2 \Delta P}{\gamma L} \] ........................ [5.9]

The fourth dimensionless group is a dimensionless velocity. The velocity of the right hand interface is a function of the pressure gradient in the water filled tube (eqn [5.9]), the capillary pressure (eqn [5.7]), and the viscosity of both fluids. The average velocity of the right hand interface is given by:

\[ v_{tube} = \frac{(\Delta P - \Delta P_c^*) r_1^2}{8 \left( \frac{\mu_o L_o}{\mu_w L_w} \right)} \] ........................ [5.10]
Where \( L_o \) and \( L_w \) are the length of the capillary tube filled with oil and water respectively. The values of \( L_o \), \( L_w \), and \( \Delta P \) are dependent upon the position of \( x_1 \).

If eqn [5.10] is divided by the average velocity for the water filled tube (eqn [5.2]), and some simplification is performed, the following is obtained:

\[
v^* = \frac{v_{\text{tube}}}{v_w} = 1 - \frac{\left[ \frac{\Delta P \gamma}{4N_{ca}} \right]}{L_o (\mu^* - 1) + 1} \quad \text{[5.11]}
\]

where \( Z = \frac{r_1}{R^*} \), \( Y = \frac{L}{R^*} \), \( L_o = \frac{L_o}{L} \), \( \mu^* = \frac{\mu_o}{\mu_w} \). The quantity \( v^* \) is the dimensionless velocity plotted in the figures presented in this chapter. As can be seen from eqn [5.11], \( v^* \) is a function of the variables \( B, C, L_o \) and the dimensionless parameters \( Z, Y, N_{ca} \) and \( \mu^* \).

The fifth dimensionless group is a dimensionless time parameter, \( t^* \). This is simply a dimensionless time step used to calculate velocity as a function of time. No new parameters are involved in this group.

5.3 Numerical Examples

The following sections will define, present and discuss the numerical simulations performed using the model.

5.3.1 Numerical Values Defined

Ten cases have been studied. These cases cover a range of viscosity ratios at two capillary numbers. Numerical values were selected that are representative of actual conditions that might be found in a porous media. The following is a list of the values used for each case.
Table 5.2 Numerical Values Used in Calculations

<table>
<thead>
<tr>
<th>Case</th>
<th>Capillary Number</th>
<th>Viscosity Ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>1.667E-02</td>
<td>0.50</td>
</tr>
<tr>
<td>2</td>
<td>1.667E-02</td>
<td>0.95</td>
</tr>
<tr>
<td>3</td>
<td>1.667E-02</td>
<td>4.00</td>
</tr>
<tr>
<td>4</td>
<td>1.667E-02</td>
<td>10.00</td>
</tr>
<tr>
<td>5</td>
<td>1.667E-02</td>
<td>40.00</td>
</tr>
<tr>
<td>6</td>
<td>5.000E-02</td>
<td>0.50</td>
</tr>
<tr>
<td>7</td>
<td>5.000E-02</td>
<td>0.95</td>
</tr>
<tr>
<td>8</td>
<td>5.000E-02</td>
<td>4.00</td>
</tr>
<tr>
<td>9</td>
<td>5.000E-02</td>
<td>10.00</td>
</tr>
<tr>
<td>10</td>
<td>5.000E-02</td>
<td>40.00</td>
</tr>
</tbody>
</table>

For each of the above cases, the physical parameters of oil drop volume, capillary tube length, and capillary tube radius were held constant. In terms of the dimensionless parameters already discussed, the values are $Z=(1/3)$ and $Y=(10/3)$. Some physical values which correspond to these dimensionless parameters are $R^* = 30 \mu m$, $r_1 = 10 \mu m$, and $L = 100 \mu m$. The first $N_{ca}$ used is large enough to make the numerator of eqn [5.11] positive. A capillary number much lower that this value would allow the oil ganglia to become trapped in the tube. The minimum capillary number required is $1.662 \times 10^{-2}$.

5.3.2 Method of Calculation

The first step in the calculation process is to specify $Z$ and $Y$. With these physical characteristics defined, $\Delta P e^*$ can be calculated as a function of $W$. This is important as solving for $C$ at a given value of $B$ requires an iterative solution ($B$ and $C$ are related through the constant drop volume assumption which is a cubic relation).
Once \( \Delta P^* \) as a function of the dimensionless meniscus position \( W \) is known, \( v^* \) can be calculated as a function of \( W \) by specifying \( N_{ca} \) and \( \mu^* \) (\( l_0 \) is calculated from \( W \)). To obtain velocity as a function of time the following method was used. At \( t^*=0, v_0^* \) is assigned the value for \( W_0=\text{initial contact} \). A dimensionless time step, \( (\Delta t^*) \), is specified, and \( W_1 \) is calculated from \( W_1=W_0+v_0^*\Delta t^* \). The value of \( v_1^* \) is then calculated based on the position \( W_1 \). Then \( W_2=W_1+v_1^*\Delta t^* \), \( v_2^* \) is calculated, and the process continues until the oil drop has passed completely through the tube.

5.3.3 Presentation and Discussion of Results

The easiest and most informative way to present the results is in graphical form. Various graphs are presented in Figures 5.8 through 5.18. The results of the numerical simulation will be discussed on a figure by figure basis.

5.3.3.1 Dimensionless \( \Delta P^* \) vs \( W \)

Figure 5.8 is a plot of the dimensionless capillary pressure change, \( \Delta P^* \), vs the dimensionless position of the right hand interface, \( W \). The most important feature of this figure is that \( \Delta P^* \) is a function only of the geometrical parameters \( Z \) and \( Y \). The general characteristics of the figure are discussed below. The most prominent feature of the figure is the rapid increase of \( \Delta P^* \) in stage 1. This indicates that the maximum value of \( \Delta P^* \) is obtained when the hemispherical cap is first formed inside the tube.
As the hemispherical cap moves down the capillary tube, $\Delta Pc^*$ slowly decreases (stage 2). The decrease in $\Delta Pc^*$ in this range is due solely to the decrease in $B$ ($C$ remains constant).

Once $W$ exceeds $Z + Y$ (when the contact line of the hemispherical cap is just in contact with $x = L$), then the primary contribution to the reduction in $\Delta Pc^*$ is both the increase of $C$ and the decrease in $B$. At about $W = 4.7$, $B = C$ and $\Delta Pc^* = 0$ at this point (stage 3).

Beyond this point, $B$ is decreasing and $C$ is increasing. Both of these factors cause a sharp decrease in $\Delta Pc^*$ making $\Delta Pc^*$ negative. The rate of decrease changes sharply at about $W = 5.0$. This corresponds to the point where $B = Z$ (stage 4).

During stage 5, $B$ remains constant, and $\Delta Pc^*$ increases slightly. The magnitude of the increase in $\Delta Pc^*$ during stage 5 is identical to the magnitude of the decrease during stage 2. The reason the curve is much sharper in the stage 5 region is due to the dimensionless distance used. Since we are tracking the right hand interface, $W$, the change in $W$ is much greater during stage 2 than it is in stage 5.

Finally in stage 6, $B$ is increasing from $Z$ to 1, while $C$ is only slightly increasing to 1. This rapid increase in $B$ is the dominating contribution to the increase in $\Delta Pc^*$ in this region. At the end of stage 6, $B = C$, hence $\Delta Pc^*$ returns to zero.

5.3.3.2 Dimensionless Velocity vs Distance $N_{ca} = 1.667E-02$

Figure 5.9 is a plot of the dimensionless velocity, $v^*$, vs dimensionless position of the right hand interface, $W$, for $N_{ca} = 1.667E-02$. Figure 5.9 illustrates the effect of viscosity ratio on velocity at a fixed $N_{ca}$. As can be seen from the figure the initial
Dimensionless Velocity vs Distance
Capillary Number = 0.01667

Figure 5.9
velocity of the ganglion decreases sharply in stage 1. This is required by the rapid increase in $\Delta P_c^*$ in stage 1.

During stage 2, $\Delta P_c^*$ decreases slowly allowing a corresponding increase in $v^*$. In stages 3 and 4 where $\Delta P_c^*$ decreases rapidly to zero and then becomes negative, $v^*$ increases rapidly. As can be seen from the numerator of eqn (5.11), when $\Delta P_c^*$ becomes negative, the capillary pressure difference actually helps move the oil ganglion through the tube. In fact when $\Delta P_c^*$ is negative, the ganglion will move through the tube even if the external pressure gradient is removed. During stage 4 $v^*$ reaches its maximum for $\mu^* = 0.5$ and $\mu^* = 0.95$.

During stage 5 $v^*$ is decreasing as $\Delta P_c^*$ becomes more negative. When $\mu^* < 1$, this has the effect of reducing $v^*$. When $\mu^* > 1$, the result is a continued increase in $v^*$. This results from the magnitude of the first product in the denominator of eqn (5.11). $v^*$ reaches a maximum for $\mu^* = 4.0$ and $\mu^* = 10.0$ at the conclusion of stage 5.

When $\mu^* = 40.0$, the denominator of eqn (5.11) becomes small enough that $v^*$ does not reach its maximum until the drop detaches itself from the tube and moves with the free stream velocity $v_w$.

5.3.3.3 Dimensionless Velocity vs Time. $N_{ca} = 1.667E-02$.

Figures 5.10 and 5.11 show the relationship between $v^*$ and $t^*$ for $N_{ca} = 1.667E-02$. As would be expected from looking at Figure 5.9, $v^*$ vs $t^*$ for all $\mu^*$ except $\mu^* = 40.0$ exhibit a maximum value before the left hand interface of the oil ganglion reaches the end of the tube. This is consistent with the velocity vs position
Dimensionless Velocity vs Time
Capillary Number = 0.01667

Figure 5.10
Dimensionless Velocity vs Time
Capillary Number = 0.01667

Figure 5.11
profiles for each viscosity ratio. For $\mu^*=40.0$ the lack of this maximum is also consistent with its velocity vs position profile.

The important feature of these figures is that for all values of $\mu^*$, the ganglion spends the majority of its time inside the tube in stage 2. Also important to note is that as $\mu^*$ increases, the time required for the ganglion to transit the tube also increases. This is not surprising given the inverse relationship between $v^*$ and $\mu^*$.

5.3.3.4 Dimensionless Distance vs Time. $N_{ca}=1.667E-02$.

Figures 5.12 and 5.13 illustrate the relationship between $W$ and $t^*$ for $N_{ca}=1.667E-02$. The important feature for all viscosity ratios is that the ganglion spends the majority of its time in stage 2. This is consistent with velocity vs position profiles described previously. Another important feature is that for all viscosity ratios, very little time is spent in stage 1.

5.3.3.5 Dimensionless Velocity vs Distance $N_{ca}=5.000E-02$

Figure 5.14 is a plot of the dimensionless velocity, $v^*$, vs dimensionless position of the right hand interface, $W$, for $N_{ca}=5.000E-02$. As in Figure 5.9, Figure 5.14 also illustrates the effect of viscosity ratio on velocity at a fixed $N_{ca}$. As in Figure 5.9 the initial velocity of the ganglion decreases sharply in stage 1. This is required by the rapid increase in $\Delta P e^*$ in stage 1.

As can be seen by comparing Figures 5.9 and 5.14, the relationship between velocity and position is very similar. One main difference between the two figures is that the minimum value of velocity obtained is higher for the larger $N_{ca}$. Another difference is that maximum in the velocity prior to the left hand interface of the ganglion reaching the end of the tube is missing for both $\mu^*=10.0$ and $\mu^*=40.0$. 
Dimensionless Distance vs Time
Capillary Number = 1.667E-02

Figure 5.12
Dimensionless Distance vs Time
Capillary Number = 1.667E-02

Figure 5.13
Dimensionless Velocity vs Distance
Capillary Number = 0.05

Figure 5.14
During stages 5 and 6 the value of $l_o$ is decreasing. The relationship between $l_o$ and $\mu^*$ appears to change as $N_{ca}$ increases. In stages 5 and 6 the numerator of eqn [5.11] is decreasing as is the denominator. The amount of decrease in the numerator increases as $N_{ca}$ increases. This follows from the fact that the denominator of eqn [5.11] is independent of the $N_{ca}$. It is expected that this trend would continue as $N_{ca}$ is increased.

5.3.3.6 Dimensionless Velocity vs Time. $N_{ca}=5.000E-02$

Figures 5.15 and 5.16 are plots of $v^*$ vs $t^*$ for $N_{ca}=5.000E-02$. These plots are very similar to Figures 5.10 and 5.11, and the comments made describing Figures 5.10 and 5.11 apply here as well. The major differences between the plots are the magnitude of the minimum velocity achieved, and the amount of time required for the ganglion to traverse the tube. At the larger $N_{ca}$ the minimum velocity is higher, and the time required to traverse the tube is much shorter. Both of these results are expected based on eqn [5.11].

5.3.3.7 Dimensionless Distance vs Time. $N_{ca}=5.000E-02$.

Figures 5.17 and 5.18 are plots of $W^*$ vs $t^*$ for $N_{ca}=5.000E-02$. These plots are very similar to Figures 5.12 and 5.13, and the comments made describing Figures 5.12 and 5.13 apply here as well. The major differences between the plots are the smoothness of the curves and the amount of time required for the ganglion to traverse the tube. The amount of time required to transit the tube is substantially shorter for the higher $N_{ca}$. The shape of the curves for the higher $N_{ca}$ are smoother than for the lower $N_{ca}$. 
Dimensionless Velocity vs Time
Capillary Number = 0.05

Figure 5.15
Dimensionless Velocity vs Time
Capillary Number = 0.05

Figure 5.16
Dimensionless Distance vs Time
Capillary Number = 0.05

Figure 5.17
Dimensionless Distance vs Time

Capillary Number = 0.05

Figure 5.18
The minimum values of velocity obtained for each case studied are contained in Table 5.3 below.

<table>
<thead>
<tr>
<th>Case</th>
<th>Dimensionless Velocity v*</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>0.003068</td>
</tr>
<tr>
<td>2</td>
<td>0.002929</td>
</tr>
<tr>
<td>3</td>
<td>0.002242</td>
</tr>
<tr>
<td>4</td>
<td>0.001534</td>
</tr>
<tr>
<td>5</td>
<td>0.000595</td>
</tr>
<tr>
<td>6</td>
<td>0.701520</td>
</tr>
<tr>
<td>7</td>
<td>0.670923</td>
</tr>
<tr>
<td>8</td>
<td>0.171607</td>
</tr>
<tr>
<td>9</td>
<td>0.068642</td>
</tr>
<tr>
<td>10</td>
<td>0.017160</td>
</tr>
</tbody>
</table>

5.4 Possible Extensions to the Model

This model was intentionally kept simple to study the effect of viscosity ratio on the displacement of an oil ganglion. In addition to the cases presented in this work, Z and Y can be varied to evaluate the effects of drop volume and the length of the tube (pore throat).

There are some modifications that could be made to extend the applicability of this model to actual porous media. The first possible modification would be to include the effect of non-zero contact angles in the derivation of the capillary pressure equation. This improvement is desirable as not all porous media of interest are water wet in nature. This improvement would change the way that B and C are calculated.
A second possible improvement would be to include the effects of contact angle hysteresis in the derivation of the capillary pressure equation. This phenomenon has been observed in many actual porous media, and the inclusion of its effects is known to be important (13), (14), (15). This modification would require a change in the methods used to calculate B and C. Some of the symmetry used to calculate C from a given value of B would no longer be applicable.

The third possible improvement concerns the uniform capillary tube assumption. It is clear that this geometry is an oversimplification of real porous media. To make this model more applicable to real world situations, we would need to take a more realistic pore geometry. Several possibilities were presented in references (13, 14).

5.5 Relation of Model Results to Relative Permeability Theory

In general the results obtained in this work agree with relative permeability theory, although the agreement is not complete. For this model to agree with relative permeability it must agree with Darcy’s Law, and the superficial velocity should be proportional to the relative permeability, viscosity ratio, and a constant driving force. These two concepts will be addressed separately starting with Darcy’s law. The following derivation will show the conditions that must be met to satisfy Darcy’s law. The average flow rate of oil through a tube, $Q_o$, is simply the volume of the oil drop divided by the total time required for the drop to move through the tube, $t_T$, and is given by:

$$Q_o \sim \frac{\frac{4}{3} \pi (R^*)^3}{t_T} \quad \cdots \cdots \cdots \cdots \quad [5.12]$$
According to Darcy's law, the flowrate $Q_0$ is proportional to the pressure gradient across the tube and the oil viscosity. This relationship is given in [5.13].

$$Q_0 \sim \frac{1}{\mu_0} \frac{\Delta P}{L} \quad [5.13]$$

If eqn [5.12] is substituted into eqn [5.13] and some simplification if performed, the following is obtained:

$$t_T \sim \frac{4}{3} \frac{\pi (R^*)^3}{\Delta P} \mu_0 \quad [5.14]$$

In the model developed in this chapter only one value of $R^*$ was used. So for a given $N_{ca}$ the first term on the right hand side of the equation is a constant, and there should be a linear relationship between $t_T$ and $\mu_0$. There should be a corresponding relationship between their dimensionless counterparts. Figures 5.19
Total Time (for Ganglion Transit) vs Viscosity Ratio

Figure 5-19
Total Time (for Ganglion Transit) vs Viscosity Ratio

Figure 5–20
and 5.20 are plots of dimensionless $t_T$ vs $\mu^*$. As illustrated in these figures, the linear relationship is excellent. This behavior is in agreement with relative permeability theory.

As noted above, for these results to be in agreement with relative permeability theory the superficial velocity should be proportional to the relative permeability, viscosity ratio, and a constant driving force. As can be seen from eqn [5.11] this requirement is not strictly met. The relative permeability is a strong function of saturation, and the $I_Q$ term satisfies this requirement as it is the saturation in one pore constriction. What is not satisfied is the constant driving force requirement. In this model $\Delta P c^*$ is not constant at any point in the model and this violates one of the basic assumptions of relative permeability theory. If we confine ourselves to considering only stage 2, then $\Delta P c^*$ does not change much, and perhaps relative permeability theory can be used to describe the flow in this stage. Basically this model is reasonably consistent with relative permeability theory.

Other pore structures could be considered that would provide large regions where $\Delta P c^*$ would be constant. In these regions the model would agree well with relative permeability theory. There are no model geometries that I can think of that would allow a constant driving force term at all points in the model. However, as $N_{ca}$ increases for any given model, the effect of small changes in $\Delta P c^*$ will become less important.

5.6 Conclusions

In conclusion the model presented in this chapter provides a good starting point for a numerical model of oil ganglion flow in an idealized porous medium. The model developed has increased our understanding of the effects of viscosity and
capillary pressure on the movement of an oil ganglion through a capillary tube. This model provides a good foundation for further work using more complex geometries, and more complex models.

The basic relationship between viscosity and velocity has been shown to be inverse in nature. The results presented here illustrate this relationship. They also illustrate the basic change in velocity as the oil ganglia moves through the porous medium. These relationships help us understand how oil ganglia move through porous media, and help us see the effects of increasing viscosity ratio and increasing $N_{ca}$.

It has been shown that the flow described by the model is basically consistent with relative permeability theory and that it satisfies Darcy's law. The agreement between the model and relative permeability theory should improve as $N_{ca}$ increases.

The extension of this model to more complex systems should allow us to broaden our understanding of the effects of viscosity and pore geometry on the movement of oil through a porous media.
6. REFERENCES


APPENDIX A  METHOD USED IN THEORETICAL MODEL CALCULATIONS

The purpose of this appendix is to provide sufficient detail to enable an interested reader to reproduce the calculations presented in chapter 5.

The starting point for the calculations is the constant drop volume assumption stated in chapter 5 and the assignment of the dimensionless geometric parameters \( Z_1(r_1/R^*) \), and \( Y_1(L/R^*) \) (refer to Figure 5.1 for the model geometry). Once the system geometry has been defined by \( Z \) and \( Y \), the total drop volume is readily calculated from the standard equation for the volume of a sphere. In this work a dimensionless volume was used and was obtained by dividing the total drop volume by \((R^*)^3\). Hence the total dimensionless drop volume is given by \( V_T = (4/3)\pi \).

The next step is to calculate the values of \( B_1(R_0/R^*) \) and \( C_1(R_{ol}/R^*) \) as a function of the dimensionless position of the right hand interface \( W_1(x_1/R^*) \). Here \( R_0 \) is the radius of curvature of the left hand interface, \( R_{ol} \) is the radius of curvature of the right hand interface, and \( x_1 \) is the distance of the extreme most point of the right hand interface from the tube inlet. The methods used to determine these values are detailed below.

The value of \( B \) was used as the primary variable in all of the calculations pertaining to drop volume, curvature, and position in stages 1 through 4. The value of \( B \) starts at 1 and is reduced in small increments. The corresponding dimensionless volume outside of the pore constriction was calculated for each value of \( B \). As can be seen from Figure 5.1 the dimensionless volume outside of the pore constriction is:

\[
V_{ol} = \frac{4}{3} \pi B^3 - V_{sc} \quad \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \quad [A.1]
\]
where $V_{sc}$ is the volume of the spherical cap of radius $B$ which protrudes into the capillary tube. The volume of a spherical cap of radius $R_0$ and height $H$ is given by $V = (1/3)\pi H^2 (3R_0 - H)$ (see Figure A.1). As can be seen from Figure A.1, $H$ can be expressed in terms of $R_0$ and $r_1$. This transformation is desirable as $V_{sc} = V/R^*$ can be expressed in terms of the dimensionless variable $B$ and parameter $Z$.

If the transformation is made the following result is obtained:

$$V_{sc} = \frac{1}{3} \pi \left[ 2B^3 - (2B^2 + Z^2) \left( B^2 - Z^2 \right) \cdot 5 \right] \quad \ldots \ldots \ldots \ldots \ldots [A.2]$$

The volume inside of the tube, $V_{inside}$, is simply $V_{inside} = V_T - V_{ol}$. For each value of $B$, the corresponding values of $V_{ol}$ and $V_{inside}$ were calculated. $B$ is reduced until it reaches the value of $Z$ at the end of stage 4.

The calculation of both $W$ and $C$ are stage dependent. In stage 1, $W$ starts at its initial value (the height of the initial spherical cap) and increases to $Z$. $W$ is calculated for a given value of $V_{inside}$ by an iterative solution method until the following equation is satisfied:

$$V_{inside} = \frac{1}{6} \pi W \left( 3Z^2 + W^2 \right) \quad \ldots \ldots \ldots \ldots \ldots [A.3]$$

This equation is simply another form of the volume of a spherical cap where $W$ takes the place of $H$. Once the value of $W$ is known for a given value of $B$, $C$ can be readily calculated. $C$ starts stage 1 with a value of 1 and decreases to the value of $Z$. $C$ is calculated from the following relation:

$$C = \frac{(Z^2 + W^2)}{2W} \quad \ldots \ldots \ldots \ldots \ldots [A.4]$$

In stage 2 the value of $C$ is constant at $Z$, and $W$ is readily calculated from the relation $W = [V_{inside} - (2/3)\pi Z^3] / (\pi Z^2) + Z$. 

In stages 3 and 4 things are a bit more complicated, and these two stages are considered together. B is still the key variable. Values of $V_{ol}$ are still tabulated for each value of B. The constant drop volume assumption forces the decrease in B to directly correspond to the increase in C during these two stages. This forced symmetry was used to calculated C in these two stages in the following manner. For each value of B and $V_{ol}$ the value of the dimensionless volume outside of the tube on the right hand side of the tube was also calculated. These volumes were used to look up the curvature in the tabulated values of B vs $V_{ol}$. This is possible because the volume of the drop outside the tube is a function of its radius only. Therefore it does not matter which side of the tube the drop is protruding from, only the volume of the drop outside of the tube is important. For a given volume the radius of curvature must be the same on either end of the tube. W was calculated from C in this region by the following relation:

$$W = Y + C + (C^2 - z^2)$$ ........................................ [A. 5]

For stages 5 and 6 a different method was used. In these two stages the variable used was C instead of B. C was allowed to assume the same values that were previously use for B in stages 1 and 2. During stage 5, B is constant at Z. During stage 6, B was determined from values of C in stage 1. The value of W was calculated in the same manner as stages 3 and 4.

Once the values of W, B, and Z have been tabulated, the dimensionless capillary pressure $\Delta P_c^*$ can be calculated from eqn [5.7]. Values of $\Delta P_c^*$ were calculated for each position value W.

The only remaining variable necessary to calculate the velocity from eqn [5.11] is the dimensionless portion of the tube filled with oil, $l_0$. The equation used to
determine $l_0$ is also stage dependent. The value used to specify the amount of the tube filled with oil is $x_1$. In terms of the dimensionless variables already defined $l_0 = (W/Y)$ in stage 1. During stage 2, $l_0$ is calculated in the same manner until it obtains the value of 1. Then it remains at 1 for the remainder of stage 2, and through stages 3 and 4.

During stages 5 and 6, the value of $l_0$ decreases from 1 to its initial value. The values of $l_0$ obtained in stages 2 and 1 are used to determine $l_0$ in stages 5 and 6. They correspond directly to values of $l_0$ determined in stages 2 and 1 respectively. For example if in stage 1 the values of B and $l_0$ are .95 and .125 respectively, in stage 6, $l_0$ will be .125 when C is .95. This scheme is the same one used to determine C in stages 3 and 4.

All of the values of B, C, W, and $l_0$ were compiled in a table, and the dimensionless velocity was calculated at each point from eqn [5.11]. Once the dimensionless velocity was known as a function of position, the dimensionless time information was calculated. The method for determining the time dependence of velocity and position is described in chapter 5.