Modeling foam delivery mechanisms in deep vadose-zone remediation using method of characteristics (MoC)

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MODELING FOAM DELIVERY MECHANISMS IN DEEP VADOSE-ZONE REMEDIATION USING METHOD OF CHARACTERISTICS (MoC)

A Dissertation

Submitted to the Graduate Faculty of the Louisiana State University and Agricultural and Mechanical College in partial fulfillment of the requirements for the degree of Doctor of Philosophy

in

The Department of Petroleum Engineering

by

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B.S., Amirkabir University of Technology (Tehran Polytechnic), 2002
M.S., Colorado School of Mines, 2005
May 2013
ACKNOWLEDGEMENTS

I am very grateful to Rural Research Institute and Craft and Hawkins Department of Petroleum Engineering for their financial support without which this research wouldn’t have crafted into a dissertation. I would like to express special thanks to Chevron Inc., PNNL, and Keller professorship for the financial support.

I would especially like to thank my advisor Dr. Seung Ihl Kam who has been very patient and encouraging to take that one extra step. His insight into this topic, organization skills and dedication to research has always been inspiring to me. He has always been on my side and helped me through the tough times. This dissertation evolved from the numerous discussions we had which I would always look forward to.

I am also indebted to my committee members Dr. Karsten Thompson, Dr. Arash Dahi, and Dr. Frank Tsai for their expertise and valuable suggestions. I appreciate PNNL experimental work which motivated this modeling study. Many thanks to PNNL researchers, especially Dr. Zhong for providing the experimental data. Heartfelt thanks to my colleagues for the many enlightening conversations and my friends at LSU who made this journey of four years very pleasant and memorable.

I am forever thankful to my parents who have always believed in me and their values have helped pave the path of privileged education. My deepest gratitude to my wife for her unconditional love and support throughout these four years.
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ABSTRACT

This study investigates foam delivery mechanisms in vadose-zone remediation by using Method of Characteristics (MoC). In such applications, dry foams are introduced into a porous medium which is initially at low saturation of water ($S_w$) containing pollutants such as metals and radionuclides. For vadose-zone remediation processes to be successful, the injected aqueous phase should carry chemicals to react with pollutants and precipitate them for immobilization and stabilization purposes. Typical remediation techniques such as water and surfactant injections are not applicable, because of the concerns about downward migration. As a result, understanding foam flow mechanism in-situ is key to the optimal design of field applications.

This study mainly consists of two parts: Part 1, formulating foam model mathematically using method of characteristics (MoC) and fractional flow analysis; and Part 2, using the model to fit to experimental data.

Results from Part 1 show that foam delivery mechanism is indeed very complicated, making the optimum injection condition field-specific. The five major parameters selected (i.e., initial saturation of the medium, injection foam quality, surfactant adsorption, foam strength, and foam stability) are shown to be all important, interacting with each other linearly and non-linearly. In addition, the presence of water bank ahead of stable foams conjectured in previous studies is confirmed. Results also imply that although dry foam injection is generally recommended, too dry injection condition is found to hurt this process due to slow foam propagation.

The results from Part 2 reveals a few important insights regarding foam-assisted deep vadose zone remediation: (i) the mathematical framework established for foam modeling can fit
typical flow experiments matching wave velocities, saturation history and pressure responses; (ii) the set of input parameters may not be unique for the fit, and therefore conducting experiments to measure basic model parameters related to relative permeability, initial and residual saturations, surfactant adsorption and so on should not be overlooked; and (iii) gas compressibility plays an important role for data analysis, thus should be handled carefully in laboratory flow experiments. Foam kinetics, causing foam texture to reach its steady-state value slowly, may impose additional complications.
1.1 Background

The use of surface and subsurface storage tanks has been a common practice for waste management in order to keep liquid-based wastes. An example can be found at the Hanford site where fuels and nuclear products for the production of plutonium during the Cold War were disposed into single and double shelled tanks over decades (Gerber, 1996). Over the years, some subsurface storage tanks experienced leak problems, causing subsurface contamination of unsaturated geological formations underneath so-called vadose zone. The term deep vadose zone is used to refer these unsaturated geological layers, which are more than 100 ft below the ground surface and can go as deep as 500 ft, where open excavation remediation techniques are thought to be impractical both technically and economically.

The contaminants of major interest at the Hanford site are inorganic materials such as technetium and uranium which are radioactive and health-threatening. Other contaminants include chromium, mercury, plutonium, and strontium (Dresel et al., 2008). A vertical migration of these contaminants, often accelerated by rainfall, causes serious safety concerns because of its potential influence on the major water sources such as rivers and lakes downstream. Proper remediation actions are required with urgency to protect wild lives and human civilizations in those regions.

There are mainly two major remediation processes considered for metal and radionuclide contaminants in the deep vadose zones (Wellman et al., 2007; Szecsody et al., 2007): (i) mobilization and recovery methods such as soil flushing, electro-kinetic mobilization, and vapor extraction which actively treat the affected areas by extracting the pollutants and (ii)
sequestration and fixation methods such as precipitation, oxidation and reduction which can be viewed somewhat passive in that the pollutants are treated in place within the subsurface, not actually being taken out. The precipitation, oxidation or reduction is the result of a chemical reaction induced by the reagents which are mixed with the carrier fluid such as gas or foam at the wellhead.

One characteristic of deep vadose zone remediation at the Hanford site is that any mobilization and recovery method requiring injection of large volume of aqueous solutions, for example, water or surfactant solution injection commonly found in conventional remediation treatments (Palmer and Fish, 1992; Voudrias, 2001; Dekker and Abriolab, 2000) is not practical because of possible downward migration of pollutants. In addition, because of high level of heterogeneity with particle size ranging from clays and silts to gravels and pebbles (Saenton et al., 2002), mobility control of injected fluids is also considered as a serious concern too. The presence of preferential paths often leaves the major portion of contaminated sites un-treated and un-swept. The need to meet these challenges suggests the use of foams, long been used in petroleum industry for mobility control and shown to be applicable to non-aqueous phase liquid (NAPL) remediation (Hirasaki et al., 2000; Aarra et al., 2002; Blaker et al., 2002; Mamun et al., 2002).

Foam injection in deep vadose zone remediation is somewhat different from other foam treatments demonstrated in oil recovery and NAPL remediation. First, surfactant preflush, much needed in typical foam processes in order to satisfy surfactant absorption and help propagation of stable foams (Hong and Bae, 1990), cannot be applied due to vertical migration of contaminants. Second, this application deals with a very dry initial condition (in fact, the entire Hanford site is located within a desert where the annual precipitation is less than several inches) with injection
of foams at very high gas fraction. Third, foams are used as a delivery vehicle to transport chemical reagents in the aqueous phase so that they interact with the contaminants for immobilization and stabilization in place. These concepts are well described in Figure 1.1.

![Figure 1.1: A schematic of foam process to immobilize and stabilize subsurface contaminants in deep vadose-zone remediation (www.pnl.gov)](image)

1.2 Objectives of this study

Although previous studies captured what might happen during foam injection into porous media, the systematic investigation of foam delivery processes for vadose-zone remediation is yet to be available.

In line with a wide range of experimental studies on deep vadose remediation before, this study, for the first time, focuses on foam transport mechanism in a porous medium by using a
mathematical technique called Method of Characteristics (MoC) where surfactant preflush is not allowed. In their modeling, the gas phase is treated as a single-component and single-phase system, while the aqueous phase was treated as a two-component and single-phase system. This approach enables the model to investigate how surfactant adsorption to surrounding soils impacts displacement mechanisms such as foam propagation rate, saturation profile, production history, and pressure responses.

Therefore, the objective of Part 1 is to analyze foam delivery mechanisms using Method of Characteristics. The displacement mechanisms are interpreted in a wide range of field conditions (i.e., initial conditions in terms of water saturation ($S_w$) and injection conditions in terms of injection foam quality ($f_g$)) and foam parameters (i.e., different levels of adsorption ($D_{sf}$), mobility reduction factor (MRF), and limiting water saturation ($S_{w^*}$)) in order to understand the system responses comprehensively. The propagation of different saturation waves is examined by two fractional flow curves (surfactant-free and surfactant-present fractional flow curves) solved simultaneously. Final results are presented in a format similar to Walsh diagram (Walsh and Lake, 1989) consisting of fractional flow curves, effluent history, saturation profile, and time-distance diagram, in addition to cumulative water production.

The objective of Part 2 is to demonstrate how such a MoC-based foam fractional flow model can be applied to fit actual experimental data, by showing necessary steps to follow one by one. This study is especially important because the model fit to experimental data shows how different pieces of information collected from different types of tests (such as soil tests, petrophysical properties, residual saturation and relative permeability measurements, flooding tests, and so on) are all connected and contribute to the overall displacement efficiency relevant
to deep vadose remediation. Zhong et al.’s experimental study (2009) is selected for such a purpose.

1.3 Chapter description

The content of each chapter is summarized as follows:

Chapter 1 gives a brief introduction of the problem solved in this study followed by the objectives.

Chapter 2 provides the background for the studies conducted on the applications of foams in petroleum recovery and remediation treatment including deep vadose zone remediation.

Chapter 3 shows detailed descriptions of the mathematical model developed for foam flow in deep vadose zone remediation. This chapter also illustrates how this model can be used to quantify the process while providing insights for more accurate design.

Chapter 4 covers the steps that have to be followed to fit the developed model to experimental data. This chapter also shows how different parameters such as formation, fluids, and foam properties can affect the outcome of the process.

Chapter 5 presents the conclusions of this study followed by recommendations for future studies.
Surfactant foams have been widely applied in petroleum industry over decades to improve and enhance oil production from petroleum reservoirs (Hoefner and Evans, 1995; Casteel et al., 1988; Borchardt et al., 1985). The key mechanism associated with foams is the ability to control the mobility of injected gas phase (for example, nitrogen, carbon dioxide, and flue gas) and to overcome the heterogeneity of geological layers. If successful, foam-assisted injection processes lead to a delay of injected-gas breakthrough in production wells and improved sweep efficiency (Lee and Kam, 2012; Rosman and Kam, 2009; Kovscek et al., 1997; Rossen, 1996; Schramm, 1994). Foam’s ability to block the high-permeability layer and therefore divert subsequent fluids into the low-permeability layer is also very useful in surfactant/foam-driven groundwater clean-up in environmental remediation industry (Zhong et al., 2009; Jeong et al., 2000; Szafranski et al., 1998), even though the shallow low-pressure and low-temperature environments in remediation are quite different from petroleum reservoirs.

Numerous studies identified a variety of possible pollutants for subsurface soil and groundwater contamination. They come from many different sources such as inappropriate disposing of chemicals to the environment, failure of surface and underground storage tanks, and leaks from pipelines. Subsurface contamination can also be caused by agricultural activities, for example, pesticides and fertilizer runoff from the field treatments. Common contaminant types are inorganic contaminants (such as metals, metalloids, and radionuclides) and organic contaminants (such as polycyclic aromatic hydrocarbons, nitroaromatics, phenols and anilines, halogenated aromatic, halogenated aliphatics, pesticides and petroleum products) (Christensen et al., 2004; Trapido, 1999; Knox et al., 1999).
Many different types of remediation techniques have been invented and applied in order to remediate subsurface contaminants. In general, these techniques could be categorized into two major groups: ex-situ and in-situ remediation methods.

The ex-situ remediation method is basically taking or scooping the contaminated soils out of the affected subsurface area and then transporting them to the processing units or sites for physical and chemical treatments. In this method, the soils are flushed with chemicals which could remove the contaminants either by chemical reaction or mechanical disturbance. According to Mulligan et al. (2001), ex-situ remediation treatments such as soil washing work better for sands with less clay-mineral and organic contents, typically 10 to 20%. Although ex-situ remediation is very commonly used, it has some major limitations: (i) if the contaminated area is large or deep, the operating costs can go high and the treatment becomes uneconomical; and (ii) if the concentration of the contaminants is low in a relatively wide area, the treatment usually is very inefficient (Mann et al., 1993). These are the major motivations of using in-situ remediation methods.

Pump-and-treat method, which is simply injecting water to recover contaminants, is perhaps the most widely used in-situ remediation method. Recent research suggests, however, that this process takes a significant, and often unacceptable, amount of time to remove a reasonable quantity of contaminants from subsurface (Voudrias, 2001; Palmer and Fish, 1992; Mackay and Cherry, 1989). This pump-and-treat method is shown to be unsuccessful and inefficient, if the solubility of contaminants in water is relatively low causing high maintenance and operation costs and if organic contaminant phases are trapped by capillary forces (Taber, 1969; Haley et al., 1991). The use of surfactant injection technique, so-called surfactant-enhanced aquifer remediation (SEAR), is shown to be very promising, because it easily dissolves
contaminants and reduces the level of capillary trapping as demonstrated in numerous one-dimensional (1D) column experiments (Brown et al., 1994; Fountain, 1992; Vignon and Rubin, 1989).

Although the SEAR method shows improved remediation efficiency over the pump-and-treat method, its performance in a multi-dimensional (2D and 3D) space is not as promising as that in 1D space (Jeong et al., 2000; Dekker and Abriolab, 2000). This dramatic difference is caused by the fact that the injected surfactant solutions are forced to contact contaminants in 1D experiment, which is not guaranteed in 2D or 3D experiment. This limitation resulting from hydrodynamics (e.g. fingering) and subsurface heterogeneity (e.g. channeling) has long been investigated in petroleum industry, and a significant progress has been made to control the mobility of injected fluids by foaming them, typically injecting gas and surfactant solutions either simultaneously or alternatively (Blaker et al., 2002).

Modeling and simulating foam transport in porous media is essential to lab-scale and field-scale treatment. Among many, there exist two major foam modeling approaches: bubble population balance modeling and local steady-state modeling. The first approach keeps track of dynamic mechanisms of bubble creation and coalescence, and puts them together to determine foam texture (i.e., the number of foam films, or bubble population) and resulting foam rheological properties as a function of time and space. The second approach is based on the pre-determined value of a reduction in gas-phase viscosity, often with “mobility reduction factor (MRF)”, assuming that attaining a local steady-state foam mobility is instantaneous. The local steady-state modeling combined with a mathematical technique called “Method of Characteristics (MoC)” has been intensively used to analyze multi-phase flow in porous media. More details about the MoC technique can be found elsewhere, and thus are not discussed here.
For example, see Lake (1989) for general discussions and derivations of the Method of Characteristics; Pope (1980) for chemical flooding; Walsh and Lake (1989) for solvent flooding; Rossen and Zhou (1995) for two-phase foam flooding; Mayberry et al. (2008) and Zanganeh et al. (2009) for three-phase foam flooding with oil; Kam and Rossen (2003), Dholkawala et al. (2005), and Afsharpoor et al. (2010) for two-phase mechanistic foam modeling.

For foam modeling purpose, the importance of capillary pressure cannot be over-emphasized (Khatib et al., 1988; Rossen, 1992). Foams cannot survive if capillary pressure ($P_c$) of a medium is so high and greater than a threshold value called “limiting capillary pressure ($P_c^*$)” (Equivalently, foams cannot survive if water saturation ($S_w$) of a medium is lower than a threshold value called “limiting water pressure ($S_w^*$)”). The magnitudes of $P_c^*$ and $S_w^*$ are shown to be material-specific, heavily depending on different types of fluid and porous medium properties.

Even with local steady-state modeling, understanding foam displacements in actual field-scale applications is not simple. The problem becomes increasingly complicated as the real-world aspects are added. Those aspects, for example, include surfactant formulations and concentrations, medium properties such as mineralogy and pore characteristics, interactions between different phases such as solubility and sorption, foam injection methods, multi-dimensional flow geometry, formation heterogeneity and so on. Among many, the effect of surfactant adsorption on foam displacement is obviously of central importance, unless surfactant adsorption is already satisfied with surfactant preflush. It is because the loss of surfactant molecules during foam injection limits the rate of foam propagation in porous media. The adsorption rate of surfactant molecules onto solid surface such as rocks and soils, which is
oftentimes described by the Langmuir-type isotherm, strongly depends on the characteristics of surfactants and rocks/soils (Grigg and Bai, 2005; Goloub et al., 1996; Trogus et al., 1977).

Deep vadose-zone foam remediation is different from other foam remediation applications, as illustrated in Figure 2.1(a), in that (i) the area of interest is very thick (as much as 300 to 500 ft of vadose zone) and desert dry (water saturation typically less than 3 to 5%); (ii) the contaminants of interest are radionuclides such as uranium (U) and technetium (Tc) leaked from storage facilities which are dissolved in aqueous phase and tend to migrate together with groundwater; (iii) the major threat is the vertical migration of contaminants along the vadose zone, which may join the flow of deep underlying groundwater zone essentially polluting the rivers and lakes downstream; and (vi) the purpose of the remediation is not cleaning up by recovering the contaminants, but “immobilizing and stabilizing” the contaminants through chemical reactions by chemical reagents in the delivering remediation fluids (Zhong et al., 2010; Zhong et al., 2009; Zhang et al., 2004; Wang et al., 2004; Hartman et al, 1999;). For example, the required chemical reactions for the precipitation of uranium and technetium are as follows, if the process works as intended (Morrison et al., 2001; Wildung et al., 2004):

\[
\text{Fe(0)}_{(s)} + \text{UO}_2(\text{CO}_3)_2^{2-} + 2\text{H}^+ = \text{UO}_2(\text{s}) + 2\text{HCO}_3^- + \text{Fe}^{2+} \quad (2.1)
\]

\[
\text{TcO}_4^{(aq)} + 3\text{Fe}^{2+} + (7+n)\text{H}_2\text{O} = 5\text{H}^+ + \text{TcO}_2.\text{nH}_2\text{O}_{(s)} + 3\text{Fe(OH)}_3 \quad (2.2)
\]

Equation 2.1 shows that the Uranium which is in the aqueous solution \((\text{UO}_2(\text{CO}_3)_2^{2-})\) precipitates as a solid phase \((\text{UO}_2(\text{s}))\) which makes it immobile. Also Technetium dissolved in water \((\text{TcO}_4^{(aq)})\) moves to the solid phase \((\text{TcO}_2.\text{nH}_2\text{O}_{(s)})\) as a result of a reaction with \text{Fe}^{2+} in presence of water (Equation 2.2).
There are two major difficulties identified. First, the delivering fluids for remediation carrying reagents should not be wet because, if so, the remediating fluids might accelerate the downward migration of contaminants. This is the main reason why conventional techniques such as pump-and-treat and SEAR are not applicable. Second, the vadose zone has a very high level of heterogeneity including pebbles, gravels, soils and clay minerals. In general, vadose-zone deposits consist of unconsolidated sands with low moisture content which show both temporal and spatial variability in saturation which complicates flow and transport processes (Alumbaugh et al., 2002). The use of foam is believed to overcome these major constraints identified in the field.

Compared to other surfactant/foam remediation treatments focusing on improved sweep efficiency in the presence of subsurface heterogeneity, this particular application has another challenge. Surfactant preflush, much needed in typical foam remediation treatments for the immediate propagation of stable foams, cannot be a possible option due to the concerns of downward migration of contaminants during surfactant pre-injection phase. If foams are injected into the medium without surfactant preflush, there are in general three different states present as conjectured by Zhong et al. (2010) and illustrated in Figure 2.1(b), because of surfactant adsorption on to the solid surfaces and subsequent coalescence of injected foams: (i) the region of high-mobility injected gas far away from the wellbore (i.e., between gas front and wetting front in Figure 2.1(b)); (ii) the region of intermediate-mobility injected water right behind (i.e., between wetting front and foam front in Figure 2.1(b)); and (iii) the region of low-mobility injected foam front near the wellbore.
Figure 2.1: Schematics of vadose-zone remediation (Zhong et al. 2010): (a) possible movement of pollutants in the vadose zone and (b) plausible foam delivery mechanisms during in-situ remediation treatments.

A series of recent experimental studies can be found for vadose-zone foam application. Zhong et al.’s study (2010) conducts an experimental study consisting of 19 column tests to investigate foam transport in different sediment packs in a range of absolute permeability values and injection foam qualities. Foam transport rates are monitored by keeping track of wet front location and pressure drop across the pack. Their study qualitatively identifies three constant regions where water saturation does not change significantly – the injection condition near the inlet, the initial condition away from the inlet, and a constant state in between where water saturation is relatively higher than other regions. Zhong et al.’s (2011) conducts another experimental study, focusing on how foam helps achieve better spatial distribution by amending flow characteristics within the contaminated zone. They perform 1-D and 2-D flow experiments by using unsaturated porous media to investigate the lateral transport mechanism of injected foams. Foam is shown to improve the sweep efficiency, even in the presence of heterogeneity, by increasing the delivery to low-permeability zones when there is a permeability contrast of 3.5.
Istok et al.’s study (2011) presents a numerical method to formulate foams to deliver polyphosphate to the deep vadose zone contaminated with Uranium. The injected polyphosphate is intended to react chemically with the pore water in vadose zone resulting in subsequent sequestration and precipitation of Uranium, which spontaneously limits the downward migration. Zhang et al. (2011) show an experimental study to look at how effective foam viscosity is affected by sediment properties and operating conditions. They also investigated the effect of different injection conditions, such as total injection rate and injection foam quality, and sediment permeability on foam effective viscosity. Their major finding is that the effective foam viscosity increases with the liquid fraction in foam, the injection rate, and sediment permeability. They also found that soil heterogeneity has less impact on the flow of foams compared to other fluids. In addition, other types of studies can be found in related areas such as adsorption (Wellman et al., 2006), visco-elastic polymer (Grate and Nelson, 2002), vapor extraction (Oostrom et al., 2005), enhanced volatilization and enhanced sorption (Dresel et al., 2008), etc.
CHAPTER 3
BUILDING FOAM MODEL FOR VADOSE ZONE REMEDIATION

This study uses concepts similar to those developed in the miscible and immiscible flooding, where different phases with multiple components in them interact with each other during the displacement process. Previous foam studies show similar approaches but not with the effect of surfactant adsorption where injected surfactants are lost continuously to the surrounding sediments. Hence there is a need to develop a mathematical formulation for vadose-zone foam-remediation process which is the focus of this chapter.

3.1 Methodology

For a multiphase flow in porous media, MoC solves the material balance equations for different chemical components present in the system (See Lake (1989) for more discussions).

For each component i, the mass conservation equation consists of accumulation $\left(\frac{\partial w_i}{\partial t}\right)$, convection $\nabla \cdot (N_i)$, and reaction ($R_i$) terms as follows:

$$\frac{\partial w_i}{\partial t} + \nabla \cdot (N_i) = R_i$$  \hspace{1cm} (3.1)

where, the first term, the overall concentration of component i ($w_i$) is defined as

$$w_i = \phi \sum_{j=1}^{N_p} \rho_j S_j w_{ij} + (1 - \phi) \rho_i w_{is}$$  \hspace{1cm} (3.2)

in which the first term is the concentration of component i in all fluid phases (i.e., phase 1 through $N_p$, expressed by subscript j) in the medium with porosity $\phi$, density of phase j $\rho_j$, saturation of phase j $S_j$, and the concentration of component i in phase j $w_{ij}$, and the second term shows the concentration of component i in the solid phase (not moving) with grain fraction (1-
\( \phi \), density of solid phase \( \rho_s \), and the concentration of component \( i \) in solid phase \( j \) \( w_{is} \); the second term, the flux of component \( i \) \( (N_i) \) is defined as

\[
N_i = \sum_{j=1}^{N_p} (\rho_j w_{ij} \vec{u}_j - \phi \rho_j S_j \vec{K}_{ij} \vec{v} w_{ij})
\]  

(3.3)

where, \( \vec{u}_j \) is the volumetric flux of phase \( j \), and \( \vec{K}_{ij} \) is the dispersion coefficient of component \( i \) in phase \( j \); and, the third term, the rate of newly created or destroyed component \( i \) \( (R_i) \) is defined as

\[
R_i = \phi \sum_{j=1}^{N_p} S_j r_{ij} + (1 - \phi) r_{is}
\]  

(3.4)

where, \( r_{ij} \) is the rate of generation or destruction of component \( i \) in phase \( j \), and \( r_{is} \) is the rate of generation or destruction of component \( i \) within the solid phase.

Substituting Equations 3.2, 3.3 and 3.4 into Equation 3.1 becomes

\[
\frac{\partial}{\partial t} (\phi \sum_{j=1}^{N_p} \rho_j S_j w_{ij} + (1 - \phi) \rho_s w_{is}) + \nabla \left( \sum_{j=1}^{N_p} \rho_j w_{ij} \vec{u}_j - \phi \rho_j S_j \vec{K}_{ij} \vec{v} w_{ij} \right) =
\]

\[
\phi \sum_{j=1}^{N_p} S_j r_{ij} + (1 - \phi) r_{is}
\]  

(3.5)

For vadose-zone foam displacement mechanisms, the following assumptions are applicable: constant and uniform temperature \( (T) \), porosity \( (\phi) \) and phase densities \( (\rho_j) \); no reaction between components \( (r_{ij} = 0 \) and \( r_{is} = 0) \); no mass transfer between different phases; instantaneous local thermodynamic equilibrium; incompressible flow; and no dispersion \( (K) \). Equation 3.5 then becomes
\[ \frac{\partial}{\partial t} \left( \phi \sum_{j=1}^{N_p} \rho_j S_j w_{ij} + (1 - \phi) \rho_s w_{is} \right) + \nabla \left( \sum_{j=1}^{N_p} \rho_j w_{ij} \ddot{u}_j \right) = 0 \] \hspace{1cm} (3.6)

If the concentration of component \( i \) in phase \( j \) \( (C_{ij}) \) is defined as follows,

\[ C_{ij} = \frac{\rho_j \omega_{ij}}{\rho_j^o} \] \hspace{1cm} (3.7)

where \( \rho_j^o \) is the density of a phase at any reference conditions (pressure and temperature), then Equation 3.7 becomes

\[ \frac{\partial}{\partial t} \left( \phi \sum_{j=1}^{N_p} S_j C_{ij} + (1 - \phi) C_{is} \right) + \nabla \left( \sum_{j=1}^{N_p} C_{ij} \ddot{u}_j \right) = 0 \] \hspace{1cm} (3.8)

This equation can be further simplified for one-dimensional flow as follows:

\[ \phi \frac{\partial}{\partial t} \left( \sum_{j=1}^{N_p} S_j C_{ij} + \frac{1-\phi}{\phi} C_{is} \right) + \sum_{j=1}^{N_p} \frac{\partial}{\partial x} \left( u_t f_j C_{ij} \right) = 0 \] \hspace{1cm} (3.9)

where, \( u_t \) is the total velocity and \( f_j \) is the fractional flow of phase \( j \) (equivalent to \( u_j/u_t, u_j \) being the velocity of phase \( j \)).

Defining the concentration of component \( i \) in the solid phase per unit pore volume of rock \( (\hat{C}_i) \) as

\[ \hat{C}_i = \frac{1-\phi}{\phi} C_{is} \] \hspace{1cm} (3.10)

Equation 3.9 then becomes

\[ \phi \frac{\partial}{\partial t} \left( \sum_{j=1}^{N_p} S_j C_{ij} + \hat{C}_i \right) + \sum_{j=1}^{N_p} u_t f_j \frac{\partial C_{ij}}{\partial x} = 0 \] \hspace{1cm} (3.11)

or

16
\[ \phi \frac{\partial}{\partial t} (C_i + \hat{C}_i) + u_t \frac{\partial F_i}{\partial x} = 0, \ i = 1, 2, \ldots, N_c \]  

(3.12)

where, \( F_i \) is the overall flux of component \( i \).

In dimensionless format, this becomes

\[ \frac{\partial}{\partial t_D} (C_i + \hat{C}_i) + \frac{\partial F_i}{\partial x_D} = 0, \ i = 1, 2, \ldots, N_c \]  

(3.13)

The use of dimensionless distance and time makes it convenient to solve the differential equations for MoC (Walsh and Lake, 1989). In all equations above, the dimensionless distance \( (x_D) \) is defined as

\[ x_D(x^{'}) = \frac{\text{pore volume contained within } x \leq x^{'}}{\text{total volume of porous medium of interest}} = \frac{xA_\emptyset}{L A_\emptyset} = \frac{x}{L} \]  

(3.14)

where, \( A \) and \( L \) are the cross-sectional area and the length of the media, and \( x \) is the location of interest. Its derivative is given by

\[ dx_D = dx/L \]  

(3.15)

The dimensionless time is defined as

\[ t_D(t) = \frac{\int_0^t u_t \, dt}{A_\emptyset L} = \frac{\int_0^t \phi u_t \, dt}{\phi L} \]  

(3.16)

and its derivative is given by

\[ dt_D = \frac{u_t \, dt}{\phi L} \]  

(3.17)

Note that \( C_i = \sum_j S_j C_{ij} \) is the overall concentration of component \( i \), \( \hat{C}_i = \frac{1-\phi}{\phi} C_{is} \) is the concentration of component \( i \) in solid phase, and \( F_i = \sum_j f_j C_{ij} \) is the overall flux of component \( i \).
By using the Chain rule,

\[
\left( \frac{\partial F_i}{\partial x_p} \right)_{t_D} = \left( \frac{\partial F_i}{\partial C_i} \right) \left( \frac{\partial C_i}{\partial x_D} \right)
\]  

(3.18)

and

\[
\left( \frac{\partial \hat{C}_i}{\partial t_D} \right)_{x_D} = \left( \frac{\partial \hat{C}_i}{\partial C_i} \right) \left( \frac{\partial C_i}{\partial t_D} \right)
\]  

(3.19)

Equation 3.13 becomes

\[
\left[ 1 + \frac{\partial \hat{C}_i}{\partial C_i} \left( \frac{\partial C_i}{\partial t_D} \right) + \left( \frac{\partial F_i}{\partial C_i} \right) \left( \frac{\partial \hat{C}_i}{\partial x_D} \right) \right] = 0, \ i = 1, 2, ..., N_c
\]  

(3.20)

From Equation 3.20, the characteristic velocity could be expressed by the following equation in the absence of saturation shock:

\[
\left( \frac{dx_D}{dt_D} \right)_{char} = v_{C_i} = \frac{\left( \frac{\partial F_i}{\partial C_i} \right)}{\left( \frac{\partial \hat{C}_i}{\partial C_i} \right) + 1}, \ i = 1, 2, ..., N_c
\]  

(3.21)

If there exists a shock, Equation 3.21 changes to

\[
\left( \frac{\Delta x_D}{\Delta t_D} \right)_{char} = \frac{\left( \frac{\partial F_i}{\partial C_i} \right)}{\left( \frac{\partial \hat{C}_i}{\partial C_i} \right) + 1}, \ i = 1, 2, ..., N_c
\]  

(3.22)

These derivations are similar to those shown in miscible flooding (Walsh and Lake, 1989).

Note that there are two phases (j = 1 for aqueous phase and j = 2 for gas phase) and three components (i = w for water component, i = g for gas component, i = sf for surfactant component) involved in this process. The gas phase only consists of the gas component, and the aqueous phase consists of either water component alone or water and surfactant components.
together. It should be noted that surfactant concentration in the surfactant solution is always the same during the displacement process, because surfactant chemicals are introduced into the medium with no surfactants initially (Lake, 1989).

For the surfactant component \((i = sf)\) in the aqueous phase \((j = 1)\), Equation 3.11 could be written as

\[
\theta \frac{\partial (s_1c_{sf1})}{\partial t} + \frac{\partial \hat{c}_{sf}}{\partial t} + u_t f_1 \frac{\partial c_{sf1}}{\partial x} = 0
\]

(3.23)

and, because surfactant molecules are only present within the aqueous phase or onto solid surface (but not within the gas phase), this equation turns into

\[
\theta \frac{\partial c_{sf1}}{\partial t} \left( S_1 + \frac{\partial \hat{c}_{sf}}{\partial c_{sf1}} \right) + u_t f_1 \frac{\partial c_{sf1}}{\partial x} = 0
\]

(3.24)

If the adsorption coefficient for surfactant molecules \((D_{sf})\) is defined as

\[
D_{sf} = \frac{\partial \hat{c}_{sf}}{\partial c_{sf1}}
\]

(3.25)

Equation 3.24 becomes

\[
\theta \frac{\partial c_{sf1}}{\partial t} \left( S_1 + D_{sf} \right) + u_t f_1 \frac{\partial c_{sf1}}{\partial x} = 0
\]

(3.26)

In dimensionless format,

\[
\frac{\partial c_{sf1}}{\partial t_D} \left( S_1 + D_{sf} \right) + f_1 \frac{\partial c_{sf1}}{\partial x_D} = 0
\]

(3.27)

As a result, the characteristic velocity of surfactant component \((i = sf)\) is

\[
\frac{dx}{dt} = \left( \frac{u_t f_1}{\theta S_1 + D_{sf}} \right)^f
\]

(3.28)
or, in dimensionless form,

\[
\frac{dx_D}{dt_D} = \left( \frac{f_1}{s_3 + D_{sf}} \right)^j .
\]  

(3.29)

For characteristic velocities of phases, the solutions are similar to the Buckley-Leverett solutions (Buckley and Leverett, 1941) once the positions of interest are located on the same fractional curves (eg. either surfactant-free or surfactant-present fractional flow curve). The Buckley-Leverett solutions between surfactant-free and surfactant-present fractional flow curves are more complicated because the injected surfactant solution is miscible with the aqueous phase initially present in the medium.

For miscible flow, the overall flux of water component is expressed as follows because the gas phase does not have water component:

\[
F_w = \sum_{j=1}^{2} f_j C_{wj} = f_1 C_{w1} + f_2 C_{w2} = f_1 C_{w1} .
\]  

(3.30)

Similarly, the overall concentration of injected water component becomes

\[
C_w = S_1 C_{w1} + S_2 C_{w2} = S_1 C_{w1}
\]  

(3.31)

Then, following Equation 3.18, the Buckley-Leverett shock (Buckley and Leverett, 1941) between surfactant-free and surfactant-present fractional flow curves (V_{BL1}) is given by

\[
V_{BL1} = \frac{\Delta \left( \sum_{j=1}^{2} f_j C_{wj} \right)}{\Delta (s_1)} = \frac{\Delta (f_1)}{\Delta (s_1)} = \frac{f_{1I} - f_{1J}}{s_{1I} - s_{1J}}
\]  

(3.32)

where, the superscript IJ represents the constant state between the initial condition (I) and the injection condition (J). Note that I and J are also constant states at the same time.
It is sometimes useful to identify the front of injected water in contrast with water initially present in the media. By using both Equations 3.30 and 3.31 together with Equation 3.18, the characteristic velocity of the injected water component \((V_1)\) becomes

\[
V_1 = \left( \frac{f_1}{S_1} \right)^I \tag{3.33}
\]

\((f_1 \text{ and } S_1 \text{ represent the fractional flow and saturation of single-component aqueous phase, respectively, see } (Lake, 1989) \text{ for more discussions})\) and, if the same is applied to the injected gas phase in contrast with gas initially present in the media, the characteristic velocity of the injected gas component \((V_2)\) becomes

\[
V_2 = \left( \frac{1-f_1}{1-S_1} \right)^I = \left( \frac{f_2}{S_2} \right)^I \tag{3.34}
\]

\((f_2 \text{ and } S_2 \text{ represent the fractional flow and saturation of single-component gaseous phase, respectively, see } (Lake, 1989) \text{ for more discussions})\).

This study uses the following Corey-type relative permeability functions for aqueous and gaseous phases, \(k_{r1}\) and \(k_{r2}\), respectively:

\[
k_{r1} = 0.7888 \left( \frac{S_1 - S_{1c}}{1 - S_{1c} - S_{2r}} \right)^{1.9575} \quad \text{and} \quad \tag{3.35}
\]

\[
k_{r2} = \left( \frac{1 - S_{1c} - S_{2r}}{1 - S_{1c} - S_{2r}} \right)^{2.2868} , \tag{3.36}
\]

where \(S_{1c}\) and \(S_{2r}\) are connate water saturation and residual gas saturation, respectively.

The fractional flow of aqueous phase \((f_1)\) is determined by
\[ f_1 = \frac{k_{r1}/\mu_1}{k_{r1}/\mu_1 + (k_{r2}/MRF\mu_2)} \] (3.37)

where, \( \mu_1 \) and \( \mu_2 \) are viscosities of aqueous and gas phases, respectively, and MRF represent the mobility reduction factor in the presence of foams. Note that if MRF=1, then no foams are present and the flow becomes conventional gas-liquid two-phase flow; and if MRF > 1, then the gas mobility is reduced due to the presence of foams. A wide range of MRF values has been reported in the literature, some as high as 100,000.

It should be noted that the results in the following sections use \( f_w \) for fractional flow of aqueous phase (rather than \( f_1 \)), \( S_w \) for saturation of aqueous phase (rather than \( S_1 \)), \( S_g \) for saturation of gas phase (rather than \( S_2 \)), \( V_w \) for propagation velocity of injected water component (rather than \( V_1 \)), and \( V_g \) for propagation velocity of injected gas component (rather than \( V_2 \)), in order to follow the conventional terminology used in the literature.

### 3.2 Results

#### 3.2.1 Construction of fractional flow curves

MoC requires the construction of fractional flow curves as a first step. By using Equations 3.35 and 3.36, two fractional flow curves can be constructed as shown in Figure 3.1(a) to represent surfactant-free (solid line) and surfactant-present (dashed line) gas-water two-phase flows. For vadose-zone foam applications where foams are injected into surfactant-free porous media, the initial condition (I) is typically given by water and gas saturations (i.e., \( (S_w, S_g)^I \)) on the surfactant-free fractional flow curve, whereas the injection condition (J) is given by water and gas fractional flows (i.e., \( (f_w, f_g)^J \)) on the surfactant-present fractional flow curve. Once the level of surfactant adsorption is measured from laboratory experiments, the surfactant adsorption
coefficient $D_{sf}$ (cf. Equation 3.25) can be calculated and the resulting adsorption point $D = (-D_{sf}, 0)$ can be located in the $f_w$ vs. $S_w$ plot as shown in Figure 3.1(a). For this particular plot, $D_{sf} = 0.2$, $MRF = 100$, and $S_{w}^* = 0.2$. Note that the surfactant-present fractional flow curve is positioned far above the surfactant-free fractional flow curve for $S_w > S_{w}^*$ because MRF is much greater than 1, while the surfactant-present fractional flow curve merges into the surfactant-free fractional flow curve for $S_w < S_{w}^*$ because MRF equals to 1 (no foams). The adsorption coefficient ($D_{sf}$) typically ranges from 0.1 to 0.5 for sandstone (Mannhardt et al., 1994).

Figure 3.1(b) shows how fractional flow curves change at various MRF values. The curve shifts upward with increasing MRF, reflecting reduced gas mobility and increased water fractional flow $f_w$.
3.2.2 Fractional flow solutions: Base case

The major parameters of interest in this study include initial condition \( I = (S_w, S_g)^I \), or \((S_w^I, S_g^I))\), injection condition \( J = (f_w, f_g)^J \), or \((f_w^J, f_g^J))\), level of adsorption \( D = (-D_{sf}, 0)\), limiting water saturation \( S_w^* \), and mobility reduction factor (MRF). The base case in this study is defined by the following parameters: \( I: (S_w, S_g) = (0.20, 0.80) \); \( J: (f_w, f_g) = (0.20, 0.80) \); \( D = (-0.20, 0) \); \( S_w^* = 0.20 \); and MRF = 100. It is assumed that water viscosity is 1 cp, gas viscosity is 0.02 cp, connate water saturation \( S_{wc} \) is 0.04, and residual gas saturation \( S_{gr} \) is 0.

Figure 3.2 shows how fractional flow solutions can be constructed graphically by using the base-case fractional flow curves. The use of MoC requires two major constraints to be satisfied: first, the saturation velocity should always increase monotonically from J to I; and second, surfactant propagation has to be represented by a chemical shock from the surfactant-present to the surfactant-free fractional flow curve because the surfactant is injected into initially surfactant-free media.

The construction of graphical solutions can be made as follows as shown in Figure 3.2 (See Buckley and Leverett (1941) for more details on the similar approaches.): (i) identify the locations of both initial and injection conditions as demonstrated by point “I” and “J”, respectively; (ii) find the location of \( D = (-D_{sf}, 0) \) on the x axis that represents a predetermined level of surfactant adsorption onto solid surface. Draw a straight line from \((-D_{sf}, 0)\) to J, whose slope is given by \( V_{sf} \), in order to construct a chemical shock (cf. Equation 3.29), and extend it until the line intersects the surfactant-free fractional flow curve. This intersection point is given by a constant state, IJ. In general, this chemical shock travels together with the Buckley-Leverett saturation shock (i.e., slow BL shock given by \( V_{BL1} \); “rear edge of water bank”) as shown in
Equation 3.32; (iii) travel from the constant state IJ to the initial condition I by increasing velocity monotonically. In this particular case, there is a shock from IJ to I (i.e., fast BL shock given by $V_{BL2}$; “front edge of water bank”); and (iv) if needed, determine the velocity of injected water ($V_w$) by connecting the origin (0, 0) and the constant state (IJ) (cf. Equation 3.33), and the velocity of injected gas ($V_g$) by connecting the initial condition (I) and (1, 1) (cf. Equation 3.34).

Figure 3.2: Graphical solution for the base case by using the Method of Characteristics (I: $(S_w, S_g) = (0.2, 0.8)$, J : $(f_w, f_g) = (0.2, 0.8)$, MRF=100, $D_{sf}$=0.2, and $S_w^* = 0.2$).

Figure 3.3 shows the construction of Walsh diagram from the base-case fractional flow curves, which adds effluent history (upper right; $f_w$ vs. $t_D$ plot at $x_D = 1$), saturation profile (lower left; $S_w$ vs. $x_D$ plot at $t_D = 0.5$), and time-distance diagram (lower right). It should be noted that the entire displacement process in this base case is governed by two BL shocks travelling at $V_{BL1}$ and $V_{BL2}$, forming a water bank in between: ahead of the bank, there are saturation conditions identical to the initial condition (I); and behind the bank, there are saturation conditions identical
to the injection condition (J). It is interesting to find that (i) the injected gas phase ($V_g$ shown by dotted line) runs much faster than the injected water phase ($V_w$ shown by dashed line) because of higher gas mobility, and (ii) the injected water phase ($V_w$ shown by dashed line) travels faster than surfactants ($V_{sf}$ shown by thick dashed line) because of the loss of surfactant molecules.

The following sections show how the characteristics of this displacement process are affected by different input conditions, more specifically, case 1 to investigate the effect of initial water saturation, case 2 the effect of injection foam quality, case 3 the effect of mobility reduction factor, case 4 the effect of surfactant adsorption, and case 5 the effect of limiting water saturation.

3.2.3 Case 1: Effect of initial water saturation

Figures 3.4 and 3.5 show the effect of initial water saturation with $I: (S_w, S_g) = (0.04, 0.96)$ and $I: (S_w, S_g) = (0.50, 0.50)$, respectively, compared with the base case where $I: (S_w, S_g) = (0.20, 0.80)$, keeping other conditions identical. The comparison with the base case shows that as the initial water saturation is higher, the width of the water bank is larger (i.e., larger gap between $V_{BL1}$ and $V_{BL2}$); the front edge of water bank moves faster (i.e., high $V_{BL2}$); and the injected gas propagates faster (i.e., high $V_g$). However, because there are no changes in injection condition (J) and constant state (IJ), water saturation at the water bank (i.e., $S_w$ at IJ), velocity of rear edge of water bank (i.e., $V_{BL1}$), and the velocity of injected liquid ($V_w$) are not altered. By the same token, there are no changes in the propagation of surfactant chemical (i.e., $V_{sf}$), exhibiting the same break-through time irrespective of initial conditions.
Figure 3.3: Walsh diagram for the base case (I: \((S_w, S_g) = (0.20, 0.80)\), J: \((f_w, f_g) = (0.2, 0.8)\), MRF=100, \(D_{sf} = 0.2\), and \(S_w = 0.2\)).
Figure 3.4: Walsh diagram for case 1 with lower initial water saturation I: \((S_w, S_g) = (0.04, 0.96)\), keeping other conditions the same as the Base Case \((J: (f_w, f_g) = (0.2, 0.8), \text{MRF}=100, D_{sf}=0.2, \text{and } S_{w^*}=0.2)\).

Figure 3.6 compares the effect of initial water saturation by using cumulative water production. Also included are the breakthrough times of injected gas \((V_g)\), injected water \((V_w)\), and surfactant chemical \((V_{sf})\). The observations from Figure 3.3 through 3.5 are consistently shown in this figure: the earlier breakthrough of front-edge water bank at higher initial \(S_w\) (due to
Figure 3.5: Walsh diagram for case 1 with higher initial water saturation I: \((S_w, S_g)=(0.50, 0.50)\), keeping other conditions the same as the Base Case (J: \((f_w, f_g)=(0.20, 0.80)\), MRF=100, \(D_{sf}=0.2\), and \(S_w =0.2\)).

higher \(V_{BL2}\); the earlier breakthrough of injected gas \((V_g)\); and unaltered breakthrough time for rear-edge water bank \((V_{BL1})\), injected water \((V_w)\), and surfactant chemicals \((V_{sf})\). The plots showing cumulative water production are not included in the following cases, because they can be constructed easily in the same manner.
Figure 3.6: Cumulative water production at different initial conditions: (a) I: (S_w, S_g)=(0.04, 0.96), (b) I: (S_w, S_g)=(0.2, 0.8), and (c) I: (S_w, S_g)=(0.5, 0.5).
3.2.4 Case 2: Effect of injection foam quality

Figures 3.7 through 3.9 show the effect of injection foam quality with $J(\mathbf{f}_w, \mathbf{f}_g) = (0.05, 0.95)$, $J(\mathbf{f}_w, \mathbf{f}_g) = (0.52, 0.48)$, and $J(\mathbf{f}_w, \mathbf{f}_g) = (0.80, 0.20)$ respectively, compared with the base case where $J(\mathbf{f}_w, \mathbf{f}_g) = (0.20, 0.80)$. The graphical solutions show that the change in injection foam quality has a significant effect on the velocity of surfactant chemical ($V_{sf}$) and rear-edge water bank ($V_{BL1}$), showing a dramatic reduction at drier foam injection conditions (i.e., very gentle slope of the line connecting D and J in Figure 3.7) and a dramatic increase at wetter foam injection conditions (i.e., very steep slope of the line connecting D and J, in Figures 3.8 or 3.9). This impacts the overall displacements in two ways: (i) as the injected foam becomes drier (i.e., low $f_w^J$ or high $f_g^J$), the velocities of surfactant chemical ($V_{sf}$) and rear-edge water bank ($V_{BL1}$) become lower; and (ii) as the injected foams are wetter (i.e., high $f_w^J$ or low $f_g^J$), all three waves (i.e., surfactant chemical, and front-edge and rear-edge water banks represented by $V_{sf}$, $V_{BL2}$, and $V_{BL1}$) propagate faster, the size of the water bank is larger, and the water saturation behind the water bank ($S_w^J$) is higher. Note that if injection water fraction ($f_w^J$) is greater than 0.52 (cf. Figure 3.9), rear edge of water bank ($V_{BL1}$) and surfactant ($V_{sf}$) do not travel at the same velocity. Instead, $V_{sf}$ is greater than $V_{BL1}$.

In actual deep-vadose zone remediation treatments, foams at low foam quality (or, wet foams) are not appropriate because of downward migration; the example of wet foam injection (e.g. $f_w = 0.52$), shown in this study, is to show the capability of the model to handle a wide range of injection foam qualities for demonstration purpose, however.
Figure 3.7: Walsh diagram for case 2 with higher injection foam quality $J$: $(f_w, f_g) = (0.05, 0.95)$ keeping other conditions the same as the Base Case (I: $(S_w, S_g) = (0.20, 0.80)$, MRF=100, $D_{sf}=0.2$ and $S_w^*=0.2$).
Figure 3.8: Walsh diagram for case 2 with lower injection foam quality J: \((f_w, f_g) = (0.52, 0.48)\), keeping other conditions the same as the Base Case \((I: (S_w, S_g) = (0.20, 0.80), \text{MRF}=100, D_{sf}=0.2, \text{and } S_w = 0.2)\).
Figure 3.9: Walsh diagram for case 2 with very low injection foam quality $J$: $(f_w, f_g)=(0.80, 0.20)$, keeping other conditions the same as the Base Case $(I: (S_w, S_g)=(0.20, 0.80), \text{MRF}=100, D_{sf}=0.2 \text{ and } S_w^*=0.2)$.

3.2.5 Case 3: Effect of mobility reduction factor

Figures 3.10 and 3.11 show the effect of mobility reduction factor (MRF) where MRF is 10 and 1000, in order to represent weak-foam and strong-foam states respectively, compared with the base case of MRF = 100. The fractional flow curve is located closer to the gas-water two-phase fractional flow curve if MRF becomes lower. Because the injection condition is
specified by injected foam quality (i.e., \( f_g^J = 0.80 \)), the waves propagate more slowly (i.e., smaller \( V_{sf}, V_{BL1}, V_{BL2} \)) as the MRF becomes lower. In addition, lower MRF values tend to cause the \( S_w \) and \( f_w \) of the water bank to be lower and the water saturation at the injection condition higher.

Figure 3.10: Walsh diagram for case 3 with low mobility reduction factor of MRF=10, keeping other conditions the same as the Base Case (I: \((S_w, S_g)=(0.20, 0.0007)\), J: \((f_w, f_g)=(0.2, 0.20)\), \(D_{sf}=0.2\), and \(S_w =0.2\)).
Figure 3.11: Walsh diagram for case 3 with high mobility reduction factor of MRF=1000, keeping other conditions the same as the Base Case (I: \((S_w, S_g)=(0.20, 0.80)\), J: \((f_w, f_g)=(0.2, 0.8)\), \(D_{sf}=0.2\), and \(S_w^* = 0.2\)).

3.2.6 Case 4: Effect of surfactant adsorption

Figures 3.12 and 3.13 show Walsh diagram where \(D:(-D_{sf}, 0) = (-0.40, 0)\) and \((0, 0)\) respectively, compared with the base-case \(D:(-D_{sf}, 0) = (-0.20, 0)\). Note that \(D_{sf}\) expresses the effect of chemical retention in pore volume units, therefore, for example, \(D_{sf} = 0.2\) means 0.2 PV of surfactant solutions should be injected in order to satisfy surfactant adsorption. As expected, a
larger value of $D_{sf}$ means a higher level of surfactant adsorption onto the soil and rock surfaces, and therefore a more delay in surfactant and foam propagation. The $S_w$ of water bank tends to reduce at higher $D_{sf}$ because the rear edge velocity ($V_{BL1}$) decreases, and the width of water bank tends to increase at higher $D_{sf}$ because the difference between front edge and rear edge velocities.

Figure 3.12: Walsh diagram for case 4 with higher adsorption level of $D_{sf}$=0.4, keeping other conditions the same as the Base Case (I: $(S_w, S_g)=(0.20,0.80)$, J: $(f_w, f_g)=(0.2, 0.8)$, MRF=100, and $S_w^*=0.2$).
(i.e., $V_{BL2} - V_{BL1}$) increases slightly. It should be noted that if there is no surfactant adsorption ($D_{sf} = 0$), the injected water travels together with surfactant chemicals at the same velocity (Figure 3.13).

Figure 3.13: Walsh diagram for case 4 with no adsorption; $D_{sf}=0.0$, keeping other conditions the same as the Base Case ($I: (S_w, S_g)=(0.20, 0.80)$, $J: (f_w, f_g)=(0.2,0.8)$, MRF=100, and $S_{w*}=0.2$).
3.2.7 Case 5: Effect of limiting water saturation

Figures 3.14 and 3.15 show the effect of limiting water saturation where $S_w^* = 0.05$ and 0.4, respectively, compared with the base case where $S_w^* = 0.2$. An increase in $S_w^*$ tends to move the position of J to the right (i.e., higher $S_w^J$ at the same $f_w^J$). Therefore, as $S_w^*$ increases, water

Figure 3.14: Walsh diagram for case 5 with lower limiting water saturation $S_w^* = 0.05$, keeping other conditions the same as base case (I: $(S_w, S_g) = (0.20, 0.80)$, J: $(f_w, f_g) = (0.2, 0.8)$, MRF=100, and $D_{sf}=0.2$).
Figure 3.15: Walsh Diagram for Case 5 with higher limiting water saturation $S_w^* = 0.40$, keeping other conditions the same as the Base Case (I : $(S_w, S_g) = (0.20, 0.80)$, J: $(f_w, f_g) = (0.2, 0.8)$, MRF=100, and $D_{sf}=0.2$).

Bank and surfactant chemicals propagate more slowly (i.e., low $V_{BL1}$, $V_{BL2}$, and $V_{sf}$), causing a delay in the breakthrough of injected water and surfactant chemicals. Note that a high $S_w^*$ value represents a poor foamer, or a medium which does not allow an efficient propagation of foams.
3.3 Discussion

The results presented above provide interesting and useful insights into the vadose-zone foam remediation. Because the remediation process is carried out at relatively dry conditions in order to avoid a downward migration of liquid, the following conditions should be satisfied in general: (i) the surfactant solution to be selected should be a good foamer so that it could provide low gas mobility (i.e., large MRF) and maintain a good stability even at dry conditions (i.e., low $S_w^*$). These two properties typically come together because stable foam films tend to reduce gas mobility; (ii) the loss of surfactant chemicals should be minimal, not only because of chemical costs involved, but also because of propagation of surfactants and foams. The more the surfactant chemicals are adsorbed onto the soil surface (i.e., large $D_{sf}$), the more slowly the surfactant and foam fronts move; and (iii) the injection condition should not be too dry (i.e., high $f_g$) or too wet (i.e., low $f_g$). If too wet, the size of water bank becomes too large causing concerns about downward movement in actual applications, and if too dry, the remediation process takes a significant amount of time due to very low wave velocities.

It should be noted, however, that the difficulty and complexity of the optimum design lie in the fact that most of these design parameters depend on many different test conditions, and the dependence is interconnected in a complicated manner. For example, if one proposes the use of higher surfactant concentration or addition of polymer into surfactant solution in order to achieve high MRF and low $S_w^*$, that tends to increase the level of surfactant adsorption (i.e., high $D_{sf}$). The effect of these foam parameters interrelated (i.e., MRF, $S_w^*$, $D_{sf}$, and $f_w^I$) not only depends on surfactant chemistry (e.g. type, formulation, concentration, disjoining pressure), but also strongly depends on other conditions such as soil type and mineralogy, groundwater composition, pore characteristics, wettability and so on. As a result, the optimum design of foam
application is very field specific, and thus the optimum condition in one site is not necessarily applicable successfully to other sites.

Although this study shows a framework for foam delivery mechanism in deep-vadose zone remediation, the model in its current form is not readily available to help and guide field applications. It is believed that two more crucial steps are still required – (i) a fit of this model to foam flood experimental data, at conditions similar to the field (eg., surfactant formulations and concentrations, soils and sediments, injection rates and methods and so on), in order to extract model parameters; and (ii) an extension of this study to multi-dimensional space to see how foam characteristics and displacement patterns change as foams are delivered deeper into the contaminated formation. These tasks remain as future topics.

It should also be noted that the results in this study are based on the assumption that gas compressibility is negligible, which is required for MoC analysis. Gas compressibility may play an important role in actual field applications because it affects the velocity of saturation waves and, more important, foam rheological properties due to the change in foam quality and total velocities.

In a 1D system, the size of water bank grows proportionately with time as it is illustrated in Figure 3.16 (a). However, when the system is radial, there is another factor coming into play - the cross-sectional area contacted by the injected foams increases with time, which results in reducing the width of water bank. Putting these two counteracting factors together, Figure 3.16 shows the size of water bank in a radial system, where the growth of water-bank width is less significant compared to that in linear geometry. Note that time t in Figure 3.16(b) is calculated by $t = \frac{t_D \pi r_e^2 H \rho}{q_t}$ with the following values: total flow rate ($Q_t$) = 0.66 m$^3$/hr, $\phi$=0.3, $r_e$=10 m and H=6 m.
Figure 3.16: Change in the size of the water bank in linear vs. radial geometry
CHAPTER 4
MODEL FIT TO EXPERIMENTAL DATA FOR FOAM-ASSISTED DEEP VADOSE ZONE REMEDIATION

Among those earlier studies, the experimental study of Zhong et al. (2009) is especially noteworthy because of detailed experimental data from laboratory flow tests during which foams at very dry conditions are injected into different soil columns. The displacement fronts for liquid bank and foams are monitored in conjunction with pressure measurement. It is also reported how the average liquid saturation changes with time and when wet liquid front and foam front break through the soil column. Figure 4.1 shows their flow apparatus in which air and 1 wt. % CS-330 (sodium lauryl ether sulfate) surfactant solution are injected simultaneously into a foam generation column followed by a vertically mounted soil column through which the position of displacement fronts can be visualized as shown in Figure 4.2.

In this chapter the mathematical foam model developed in Chapter 3 is compared to the experimental data of Zhong et al. (2009).

Figure 4.3, which is used as a basis for the initial discussion in this chapter, shows an example from chapter 3 where the initial water and gas saturations are 0.2 and 0.8 (i.e., I:($S_w^I$, $S_g^I$) = (0.2, 0.8)), injection water and gas fractions are 0.2 and 0.8 (i.e., J:($f_w^J$, $f_g^J$) = (0.2, 0.8)), gas-phase mobility reduction factor (MRF) is 100 (i.e., meaning that gas viscosity increases by a factor of 100 by foaming), level of surfactant adsorption ($D_{sf}$) is 0.2 (i.e., meaning that 0.2 pore volume of surfactant solution is required to satisfy surfactant adsorption), and limiting water saturation ($S_w^*$) is 0.2 (i.e., meaning that foam completely collapses if the media is too dry with $S_w < 0.2$). Solving two fractional flow curves (one with surfactant and the other with no
Figure 4.1: A schematic of experimental set up in Zhong et al.’s study (2009).

Figure 4.2: Foam delivery experiment for deep vadose-zone remediation showing the propagation of wet front (Zhong et al., 2009).
Figure 4.3: An example result of MoC based foam modeling for deep vadose-zone remediation (I: \((S_w, S_g) = (0.2, 0.8)\), J: \((f_w, f_g) = (0.2, 0.8)\), MRF = 100, \(D_{sf} = 0.2\), and \(S_w^* = 0.2\)).

Surfactant in water) in the \(f_w\) vs. \(S_w\) domain simultaneously, the MoC-based fractional flow analysis produces effluent history, saturation profile, and time-distance diagram. The general output from the modeling study is consistent with that from experimental study of Zhong et al. (2010) – the migration of three constant states such as initial condition, injection condition, and intermediate state (denoted by I, J, and IJ in Figure 3.3 respectively) is governed by two shock fronts (denoted by slow-moving Buckley-Leverett shock, \(V_{BL1}\), and fast-moving Buckley-
Leverett shock, $V_{BL2}$), one surfactant chemical front (denoted by chemical shock, $V_{sf}$) and propagation of injected water and gas (denoted by $V_w$ and $V_g$ respectively). But their model has never been applied to actual laboratory or field pilot test data.

4.1 Methodology

Although detailed derivations are given in chapter 3, important equations are reiterated here.

The governing mass conservation equation for multiphase flow in porous media can be written as follows in general for phase $j$ (Lake, 1989):

$$\phi \frac{\partial c_{ij}}{\partial t}(S_j) + u_t f_j \frac{\partial c_{ij}}{\partial x} = 0, \quad i = 1,2,...,N_c$$

(4.1)

where, subscript $i$ represents a component and $\phi$, $S_j$, $c_{ij}$, $u_t$, $f_j$, and $N_c$ represent the porosity of the media, saturation of phase $j$, concentration of component $i$ in phase $j$, total injection velocity, fractional flow of phase $j$ and the total number of components in the system respectively. For deep vadose zone foam remediation, the subscript $j$ represents either aqueous ($j = w$) or gaseous ($j = g$) phase. The aqueous phase consists of water component ($i = 1$) with or without surfactant component ($i = sf$), and the gaseous phase consists of only gas component ($i = 2$). For surfactant component in the aqueous phase ($i = sf$ and $j = w$), the delay in surfactant propagation slightly modifies Equation 4.1 as shown below because of the loss of surfactant molecules to soil surface.

$$\phi \frac{\partial c_{sfw}}{\partial t} \left( S_w + D_{sf} \right) + u_t f_w c_{sfw} \frac{\partial c_{sfw}}{\partial x} = 0$$

(4.2)

where, $c_{sfw}$ is the concentration of surfactant chemicals in the aqueous phase and $D_{sf}$ is the pore volume of surfactant solution required to satisfy surfactant adsorption for one pore volume of
soils. The water and gas fractional flow \( f_w \) and \( f_g \) can be expressed as follows in typical gas-water two phase flow:

\[
f_w = \frac{k_{rw}}{\mu_w} = \frac{1}{1 + \frac{k_{rg}}{k_{rw}} \frac{\mu_w}{\mu_g}}; \quad f_g = 1 - f_w \quad . \tag{4.3}
\]

In the presence of foams, the equation becomes as follows by using a mobility reduction factor (MRF) to account for reduced gas-phase mobility.

\[
f_w = \frac{k_{rw}}{\mu_w} = \frac{1}{1 + \frac{k_{rg}}{k_{rw}} \frac{\mu_w}{\mu_g \text{MRF}}} \quad ; \quad f_g = 1 - f_w \quad . \tag{4.4}
\]

Note that the MRF value is equal to 1 when there is no foam present (i.e., conventional gas-liquid two-phase flow) and can go as high as tens of thousands or more (Lee et al., 1991).

According to Equations 4.3 and 4.4, knowing the relative permeability functions for aqueous and gaseous phases is essential to computing water and gas fractional flows. A common technique to express relative permeability mathematically is using Corey-type functions by using two end points (A, B), two exponents (m, n), residual water saturation \( S_{wr} \), and residual gas saturation \( S_{gr} \) (Corey, 1954), which leads to

\[
k_{rw} = A \left( \frac{S_w - S_{wr}}{1 - S_{wr} - S_{gr}} \right)^m \tag{4.5}
\]

and

\[
k_{rg} = B \left( \frac{1 - S_w - S_{gr}}{1 - S_{wr} - S_{gr}} \right)^n \tag{4.6}
\]

The transport equations for aqueous and gaseous phases in porous media, so-called Darcy equations, can be written as follows, respectively:
Darcy’s equation for the gaseous phase in presence of foam then becomes

\[
\begin{align*}
\frac{u_g}{\mu_g} &= \frac{kk_{rg} \Delta P}{MRF L} = \frac{kb \left( \frac{1-S_w - S_{gr}}{1-S_{wr} - S_{gr}} \right)^n}{\mu_g MRF} \frac{\Delta P}{L} \\
\end{align*}
\] (4.8)

For this particular type of foam applications where there exist three constant states (i.e., initial condition (I), injection condition (J), and intermediate state (IJ) in between), there are two shock waves associated with displacement process – dimensionless saturation velocity \( V_{BL1} \) (traveling together with dimensionless surfactant propagation rate \( V_{sf} \)) between IJ and J, and dimensionless saturation velocity \( V_{BL2} \) between I and IJ. Note that \( V_{BL2} \) is a fast wave representing the front-edge of water bank (referred to as “wet front” below), while \( V_{BL1} \) is a slow wave representing the rear-edge of water bank (referred to as “foam front” below). The average water saturation inside the pack \( S_{wavg} \) can then be expressed as follows, if the dimensionless time \( (t_D) \), in pore volume injected (PV), is less than the wet front breakthrough time, \( t_{DwBT} \) (or, equivalently, the dimensionless distance \( (x_D) \) of wet front is less than one) as shown in the saturation profile in Figure 4.3:

\[
S_{wavg} = S_w^I t_D V_{BL1} + S_w^{IJ} (t_D V_{BL2} - t_D V_{BL1}) + S_w^I (1 - t_D V_{BL2})
\] (4.9)

where, \( t_D V_{BL1} \) and \( t_D V_{BL2} \) are the dimensionless distances \( (x_D) \) of foam front and wet front at the dimensionless time of \( t_D \) respectively. (for example, in the saturation profile of Figure 4.3, they are about 0.20 and 0.30 at \( t_D = 0.5 \)). Note that \( s_w^I, s_w^j, \) and \( s_w^{ij} \) represent water saturations
at the initial, injection, and intermediate states respectively (for example, in the fractional flow curves and saturation profile of Figure 4.3, they are about 0.20, 0.84, and 0.30, respectively). If $S_w^I$, $S_w^J$, and $S_{wavg}$ are known experimentally, $S_w^H$ can be estimated from the following equation:

$$S_w^H = \frac{S_{wavg} - S_w^I t_D V_{BL1} - S_w^J (1-t_D V_{BL2})}{t_D V_{BL2} - t_D V_{BL1}}.$$  

(4.10)

It should be noted that the three dimensionless velocities (see the straight line construction in the fractional flow curves of Figure 4.3) are defined as follows:

$$V_{BL1} = \frac{f_w^{IJ} - f_w^I}{S_w^{IJ} - S_w^I},$$  

(4.11)

$$V_{BL2} = \frac{f_w^{IJ} - f_w^I}{S_w^{IJ} - S_w^I}, \text{ and}$$

(4.12)

$$V_{sf} = \frac{f_w^I}{S_w + D_{sf}}$$

(4.13)

where, water fractional flow at the intermediate state ($f_w^{IJ}$) and water fractional flow at the initial condition ($f_w^I$) can be calculated as follows, if other parameters are given:

$$f_w^{IJ} = f_w^I + (S_w^I - S_w^J) V_{BL1}, \text{ and}$$

(4.14)

$$f_w^I = f_w^{IJ} + (S_w^I - S_w^{IJ}) V_{BL2}.$$  

(4.15)

See chapter 3 for more details about related mathematical definitions and derivations.

In order to follow common terminology, the term “water” is used to represent the aqueous phase, and “gas” is used to represent the gaseous phase below. The following parameters are employed in all calculations below: water viscosity ($\mu_w$) = 1 cp, gas viscosity ($\mu_g$) = 0.02 cp, and residual gas saturation ($S_{gr}$) = 0.
4.2 Results

Before discussing model fit to experimental data, it is valuable to understand how fractional flow solutions based on MoC calculations can first be translated into a format consistent with laboratory flow tests. Three different types of experimental data are commonly collected and reported – pressure response of the system at different locations, cumulative water production from water volume measurement at the outlet, and average water saturation inside the pack from weight measurement. Figures 4.4(a) and 4.4(b) show the cumulative water production and the average water saturation as a function of time for the case shown in Figure 4.3, which can be calculated from effluent history and saturation profile, respectively.

Figure 4.4: Construction of (a) cumulative water production and (b) average water saturation from Figure 4.3 (I: \((S_w, S_g) = (0.2, 0.8)\), J: \((f_w, f_g) = (0.2, 0.8)\), MRF = 100, \(D_{sf} = 0.2\), and \(S_w^* = 0.2\).
Figure 4.5: Water saturation profile at different times showing the propagation of wet front and foam front in conjunction with Figure 4.4: (a) at the initial condition; (b) before wet front breakthrough; (c) at wet front breakthrough; (d) at foam front breakthrough.

Figures 4.5(a) through 4.5(d) show a series of snapshots illustrating the change in water saturation profile at $t_D = 0.0, 1.35, 1.52,$ and $2.50$ PV, respectively, with $S_w^I = 0.20$, $S_w^{IJ} = 0.85$, and $S_w^J = 0.30$. Note that Figure 4.5(c) corresponds to when the constant state IJ forming water
bank displaces the initial condition “I” out of the system completely (i.e., “wet front breakthrough” or “front-edge water-bank breakthrough”), while Figure 4.5(d) corresponds to when the injection condition “J” displaces the water bank completely (i.e., “foam breakthrough” or “rear-edge water-bank breakthrough”). It should be pointed out that the width of water bank (i.e., \((V_{BL2} - V_{BL1})t_D\)) increases proportionally with time.

### 4.2.1 Analysis of experimental data from Zhong et al. (2009)

The sands used in Zhong et al. (2009) to pack the column are from the sediments in the Hanford site, at approximately 1 m below surface. The original moisture content of the Hanford sediment is estimated to be around 0.4 wt%, which is equivalent to \(S_w = 0.023\), assuming water density \(\rho_w = 1 \text{ g/cm}^3\), sediment solid density \(\rho_s = 2.6 \text{ g/cm}^3\), and one pore volume of soil column = 49.8 cm\(^3\). Because of additional 1 wt% moisture content added during packing (i.e., a total of 1.4 wt% moisture), the initial water saturation of the pack is estimated to be \(S_w = S_w^1 = 0.082\) (corresponding to actual water volume of 4.09 cm\(^3\)) which is believed to be close to the gas-flooded residual water saturation \(S_{wr}\). Other reported information is summarized in Table 4.1.

Figure 4.6 shows the original experimental data that Zhong et al. (2009) collected from laboratory flow test as a function of dimensionless time (i.e., \(t_D\) in pore volume injected (PV)) by using the experimental set-up as shown in Figure 4.1. The figure, which is updated and
Table 4.1: Experimental data reported in Zhong et al. (2009)

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Gas Phase</td>
<td>air</td>
</tr>
<tr>
<td>Concentration of CS-330 surfactant solution as liquid phase, wt%</td>
<td>1.0</td>
</tr>
<tr>
<td>Solid phase</td>
<td>Hanford sands</td>
</tr>
<tr>
<td>Absolute permeability (k), Darcy</td>
<td>47.9</td>
</tr>
<tr>
<td>Injection water fraction (f_w)*</td>
<td>0.05</td>
</tr>
<tr>
<td>Total injection rate (Q_t), cm³/min*</td>
<td>8.08</td>
</tr>
<tr>
<td>Gas injection rate (Q_g), cm³/min*</td>
<td>7.68</td>
</tr>
<tr>
<td>Liquid injection rate (Q_w), cm³/min</td>
<td>0.404</td>
</tr>
<tr>
<td>Pack length (L), cm</td>
<td>30.2</td>
</tr>
<tr>
<td>Pack diameter (D), cm</td>
<td>2.5</td>
</tr>
<tr>
<td>Porosity ((\phi))</td>
<td>0.31</td>
</tr>
<tr>
<td>Pore volume ((V_p)), cm³</td>
<td>49.8</td>
</tr>
<tr>
<td>Steady-state pressure drop during foam injection ((\Delta p_{\text{ss}})), kPa (gauge)</td>
<td>229.2</td>
</tr>
<tr>
<td>Back Pressure ((P_{\text{out}})), kPa (absolute)</td>
<td>101.3</td>
</tr>
</tbody>
</table>

* (All flow rates and fractions are reported at the back pressure)

overwritten in bold by the terms and analysis used in this study on the original plot, consists of wet front location (or, where the position of leading-edge water bank is located; \(V_{BL_2} \times t_D\)), liquid uptake (or, how much the weight of pack holder gains by introducing foams into the pack compared to the initial weight, which is essentially the same as the gain of water weight in the pack; \(\Delta W_w\)), and pressure drop (or, how the inlet pressure changes at the fixed outlet pressure of atmospheric pressure \(\Delta P = P_{\text{in}} - P_{\text{out}}\)).

A few important parameters can be extracted from Figure 4.6 for model fit. First, the wet front location follows a linear trend as expected by the MoC model (note the slope is no other
than $V_{BL2}$), and the wet front breakthrough time ($t_{DwBT}$) is expressed by $1/V_{BL2}$ which is around 15 PV. Second, the liquid uptake increases with time until $t_{DwBT} = 1/V_{BL2}$, but declines after that until foam breakthrough time ($t_{DFBT}$) is reached at $t_{DFBT} = 1/V_{BL1}$ as the water bank leaves the pack. This reduction in liquid uptake takes place between $t_D = t_{DwBT} = 15$ and $t_D = t_{DFBT} = 18.2$ PV. Third, the almost constant liquid uptake after $t_{DFBT} = 18.2$ PV reflects the steady-state injection water saturation, more precisely, $(S_w^J - S_w^I)$ which is no other than $(\Delta W_w/\rho_w)/V_p$, i.e.,

$$(S_w^J - S_w^I) = (7.48 \text{ g})/(1 \text{ g/cm}^3)/(49.8 \text{ cm}^3) = 0.150,$$

leading to $S_w^J = 0.232$ (or, actual water volume of 11.57 cm$^3$ equivalently). Fourth and last, the fact that the steady-state pressure drop ($\Delta P_{ss}$) is about $2.29 \times 10^5$ Pa, more than two times higher than the back pressure ($P_{out} = 1.01 \times 10^5$ Pa in terms of absolute pressure), implies that the gas compressibility also plays a significant
role. Such an effect can impact wave velocities drastically, for example, slowing down $V_{BL1}$ and $V_{BL2}$ gradually as the pressure drop across the core increases.

It should be noted that exactly when the system reaches a steady state is not clear. In an ideal world with no gas compressibility and immediate attainment of steady-state foam texture, the steady state during foam injection reaches when foam front breaks through. As shown in Figure 4.6, foam breakthrough time ($t_{DIBT}$) is about 18.2 PV in liquid uptake data while about 23 PV or more in pressure drop data. This study first moves on with $t_{DIBT} = 18.2$ PV as Base Case, and the implication of reading much higher $t_{DIBT}$ is covered with Case 1, Case 2, and Case 3.

With the initial and injection saturations ($S_w^I$ and $S_w^J$) determined, the liquid uptake information in Figure 4.6 can be translated into the water saturation profile at different $t_D$’s (Figure 4.7(a)) and average water saturation history (Figure 4.7(b)). For example, when the wet front reaches the outlet at $t_{DwBT} = 15$ PV, the average water saturation at that particular moment

![Diagram](image)

Figure 4.7: Change in water saturation manipulated prior to model fit: (a) saturation profile at the time of wet front breakthrough, $t_{DwBT}=15$ PV; (b) history of average water saturation inside the pack.
is $S_{\text{avg}} = 0.276$ (Figure 4.7(b)). This allows $S_{w}^{\text{IJ}}$ to be calculated ($S_{w}^{\text{IJ}} = 0.478$) by using water material balance (Equation 4.10) as shown in Figure 4.7(a). Table 4.2 shows more details about how to convert weight-based liquid uptake data to saturation information.

Table 4.2: Translation of liquid uptake data to water saturation

<table>
<thead>
<tr>
<th>Zhong et al. (2009)</th>
<th>Calculated water volume ($V_{w}$), cm$^3$</th>
<th>Calculated water saturation ($S_{w}$), fraction</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nominal PV injected</td>
<td>Liquid uptake, gram</td>
<td></td>
</tr>
<tr>
<td>0</td>
<td>0</td>
<td>4.09</td>
</tr>
<tr>
<td>1.31</td>
<td>0.90</td>
<td>4.99</td>
</tr>
<tr>
<td>1.9</td>
<td>2.54</td>
<td>6.63</td>
</tr>
<tr>
<td>4.05</td>
<td>3.89</td>
<td>7.98</td>
</tr>
<tr>
<td>6.07</td>
<td>5.08</td>
<td>9.17</td>
</tr>
<tr>
<td>8.09</td>
<td>6.28</td>
<td>10.37</td>
</tr>
<tr>
<td>10.12</td>
<td>7.63</td>
<td>11.72</td>
</tr>
<tr>
<td>12.14</td>
<td>8.52</td>
<td>12.61</td>
</tr>
<tr>
<td>14.17</td>
<td>8.97</td>
<td>13.06</td>
</tr>
<tr>
<td>16.19</td>
<td>7.48</td>
<td>11.57</td>
</tr>
<tr>
<td>18.21</td>
<td>7.77</td>
<td>11.86</td>
</tr>
<tr>
<td>20.24</td>
<td>7.62</td>
<td>11.71</td>
</tr>
<tr>
<td>22.26</td>
<td>7.47</td>
<td>11.56</td>
</tr>
<tr>
<td>24.29</td>
<td>7.33</td>
<td>11.42</td>
</tr>
<tr>
<td>26.31</td>
<td>7.48</td>
<td>11.57</td>
</tr>
<tr>
<td>28.45</td>
<td>7.48</td>
<td>11.57</td>
</tr>
</tbody>
</table>

*(Initial moisture content is estimated to be 4.09 cm$^3$ before the flow experiment)*

4.2.2 Equations and unknowns

The biggest uncertainty for model fit is that the relative permeability functions are not available. This uncertainty also prevents $f_{w}$ values from being calculated even though $S_{w}^{1}$, $S_{w}^{J}$, and $S_{w}^{\text{IJ}}$ are determined already as described in the previous section.
It is worth mentioning that even though \( f_w^J \) is given by the experimental conditions, the value cannot be used for the modeling purpose directly. It is because the injected gas phase compresses as the system pressure builds up during foam injection, and therefore the reported values of \( f_w^J \) evaluated at the back pressure is not a true representation of actual injection condition. Because the adsorption point \((-D_{sf}, 0)\), injection condition \((S_w^J, f_w^J)\), and wet front velocity \((V_{BL1})\) are interrelated (Equation 4.12) and because \( S_w^J \) and \( V_{BL1} \) are provided by the experiments, this study specifies \( D_{sf} \) as an input to calculate \( f_w^J \). \( D_{sf} \) for the soil samples tested is unknown, but this study assumes \( D_{sf} = 0.2 \) based on the typical range for soils, sands, and sandstones (about 0.1 to 0.5; see Mannhardt et al. (1994) and Mannhardt (1993) for more details).

By putting these together, there are seven unknowns such as \( A, B, m, n \) and MRF (cf. Equations 4.7a, 4.7b and 4.8) together with \( f_w^U \) and \( f_w^I \) (cf. Equations 4.14 and 4.15) and seven equations as follows:

- Darcy’s equation for water at the steady state \((J)\): Equation 4.7a with \( A \) and \( m \);

- Darcy’s equation for gas at the steady state in presence of foam \((J)\): Equation 4.8 with \( B, n \) and MRF;

- Calculation of \( f_w^U \) using \( V_{BL1} \): Equation 4.14 for \( f_w^U \);

- Calculation of \( f_w^I \) using \( V_{BL2} \) and \( f_w^U \): Equation 4.15 for \( f_w^I \);

- Positioning the initial condition \((I: (S_w^I, f_w^I))\) on the water-gas fractional flow curve: Equation 4.3 with \( A, B, m, \) and \( n \);
- Positioning the intermediate condition (IJ: \((S_w^I, f_w^I)\)) on the water-gas fractional flow curve: Equation 4.3 with A, B, m, and n;

- Positioning the injection condition (J: \((S_w^J, f_w^J)\)) on the foam fractional flow curve: Equation 4.4 with A, B, m, n, and MRF.

Please note that since only two of the last three equations are independent (i.e., these three equations are connected through \(V_{BL1}\) and \(V_{BL2}\); cf. Equations 4.11 and 4.12), there are seven unknowns but six independent equations. As a result, this study assumes that the end-point relative gas permeability is one (i.e., \(B = 1\)) due to the nature of highly dry and unconsolidated sandpack, and solve for other six unknowns for simplicity. This does not limit the robustness of this modeling approach, however, as discussed more below.

4.2.3 Construction of fractional flow solutions

Before constructing fractional flow solutions, it must be decided how the experimental data in Figure 4.6 should be manipulated to accommodate the effect of gas compressibility. This study defines the Base Case as follows: gas phase assumed to be incompressible (meaning that the nominal dimensionless time (PV) in Table 4.2 and Figure 4.6 remain identical); input parameters such as described above (\(\Delta p_{ss} = 229.2\) kPa, \(S_w^I = 0.082\), \(S_w^J = 0.232\), \(S_w^{IJ} = 0.478\), \(D_{sf} = 0.2\), \(t_{DwBT} = 15\) PV, \(t_{DBT} = 18.2\) PV, \(Q_t = 8.08\) cm\(^3\)/min, \(f_w^J = 0.0238\), \(B=1\)); and output parameters such as \(f_w^I = 0.0109\), \(f_w^{IJ} = 0.03725\), MRF = 325.43, \(A = 8.72 \times 10^{-5}\), \(m = -0.38135\), and \(n = 17.17\) as summarized in Table 4.3. A graphical construction of fractional flow solutions is shown in Figure 4.8.

Although Figure 4.8 may look reasonable satisfying all input conditions overall, it should be noted that the value of m is negative when a certain number of constraints are imposed (eg. 0
< f_w^J < f_w^J < f_w^J < 1.0, MRF > 1). This implies that even though there might be multiple solutions satisfying those six equations “mathematically”, they may not be necessarily correct “physically”. Among numerous numerical calculations, including Goal-Seek and Solver functions in Microsoft Excel, none of them have produced a set of six parameters physically meaningful when the same input parameters are applied.

In order to move forward, the next step is taken such that a physically meaningful value of m is enforced to the fractional flow model. As an easiest guess assuming a linear relative permeability, m = 1 is selected randomly. This requirement of m = 1 (in addition to B = 1) inevitably forces a condition to be relaxed for data fit as a trade-off. Thinking of the uncertainty in foam breakthrough time (Figure 4.6), the calculation procedure now allows t_{DBT} to be calculated as an output rather than input. (In reality, once A, B, m, and n values are measured in lab experiments, fractional flow solutions can be used to determine wave velocities and breakthrough times; this study proceeds in opposite way, however, because relative permeability functions are not available.)

This study further deals with gas compressibility more systematically: Case 1 assuming that the gas phase is incompressible and input parameters just like Base Case with the exception of m=1 as an input and t_{DBT} as an output; Case 2 the same as Case 1 with the exception of the gas phase being compressed with time as foam is injected, by using the ideal gas law at the average system pressure (i.e., the average of inlet and outlet pressures); and Case 3 the same as Case 1 with the exception of the gas phase being compressed instantaneously at the steady-state pressure (Δp_{ss}). Note that, as a result, the total injection rates in Case 1 and Case 3 are fixed at Q_t = 8.08 and 4.01 cm³/min, while the total injection rate in Case 2 varies with time. For the
Figure 4.8: Fractional flow solutions (Base Case): See Table 4.3 for inputs and outputs.

The purpose of Darcy’s equation calculation for Case 2, an averaged total injection rate of $Q_t = (8.04 + 4.01)/2$ cm$^3$/min is used. More details about inputs and outputs are summarized in Table 4.3.

How dimensionless times in Base Case, Case 1, Case 2, and Case 3 are re-scaled is shown in Table 4.4.

Following re-scaled $t_D$’s as shown in Table 4.4, Figure 4.9 shows how the experimental data in Figure 4.6 can be interpreted. The values on the y axis are not affected while the values on the x axis are. The breakthrough times of wet front and foam front are affected significantly.
Table 4.3: Summary of the input and output parameters for different cases investigated

<table>
<thead>
<tr>
<th>parameter</th>
<th>Base Case</th>
<th>Case I</th>
<th>Case II</th>
<th>Case III</th>
</tr>
</thead>
<tbody>
<tr>
<td>wet front breakthrough time ($t_{DwBT}$), PV</td>
<td>15</td>
<td>15</td>
<td>12.8</td>
<td>8</td>
</tr>
<tr>
<td>foam front breakthrough time ($t_{DfBT}$), PV</td>
<td>18.2</td>
<td>N/A</td>
<td>N/A</td>
<td>N/A</td>
</tr>
<tr>
<td>total injection rate ($Q_t$), cm$^3$/min</td>
<td>8.08</td>
<td>8.08</td>
<td>6.045</td>
<td>4.01</td>
</tr>
<tr>
<td>injection water fraction ($f_w$)</td>
<td>0.024</td>
<td>0.013</td>
<td>0.015</td>
<td>0.024</td>
</tr>
<tr>
<td>end point for gas rel. perm. function (B)</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>exponent for water rel. perm. function (m)</td>
<td>N/A</td>
<td>1</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>end point for water rel. perm. function (A)</td>
<td>0.000087</td>
<td>0.000577</td>
<td>0.000506</td>
<td>0.000538</td>
</tr>
<tr>
<td>exponent for gas rel. perm. function (n)</td>
<td>17.17</td>
<td>30.04</td>
<td>31.06</td>
<td>32.54</td>
</tr>
<tr>
<td>mobility reduction factor (MRF)</td>
<td>325.43</td>
<td>32.27</td>
<td>36.00</td>
<td>42.10</td>
</tr>
<tr>
<td>initial water fractional flow ($f_w^I$)</td>
<td>1.09E-02</td>
<td>1.23E-15</td>
<td>1.09E-15</td>
<td>1.13E-15</td>
</tr>
<tr>
<td>intermediate water fractional flow ($f_w^{IJ}$)</td>
<td>0.03725</td>
<td>0.01522</td>
<td>0.01784</td>
<td>0.02854</td>
</tr>
<tr>
<td>exponent for water rel. perm. function (m)</td>
<td>-0.38135</td>
<td>N/A</td>
<td>N/A</td>
<td>N/A</td>
</tr>
<tr>
<td>foam breakthrough time ($t_{DfBT}$), PV</td>
<td>N/A</td>
<td>33.54</td>
<td>28.62</td>
<td>17.89</td>
</tr>
</tbody>
</table>

as the gas compressibility is manipulated differently: $t_{DwBT} = 15.0$ PV, $t_{DfBT} = 33.54$ PV in Case 1; $t_{DwBT} = 12.8$ PV, $t_{DfBT} = 28.62$ PV in Case 2, $t_{DwBT} = 8.0$ PV, $t_{DfBT} = 17.89$ PV in Case 3. In all three cases, m value of 1 requires foam breakthrough time ($t_{DfBT}$) much higher than 18.2 PV in Base Case. Saturation profiles in Figure 4.10 seem pretty much the same at $t_D = t_{DwBT}$ in all three cases even though actual $t_{DwBT}$ values are very different, keeping almost identical $S_w^{IJ}$ values.

Figure 4.11 shows the change in average water saturation in Case 1 through 3, compared with experimental data.

Figures 4.12 through 4.14 show the fractional flow solutions for Cases 1, 2, and 3, respectively. These graphical solutions are consistent with the inputs and outputs shown in Table 4.3.
Table 4.4: Modifying the experimental data for pressure change: Case 1, incompressible gas; Case 2, gas being compressed as pressure builds up; and Case 3, gas instantaneously compressed at the steady state pressure

<table>
<thead>
<tr>
<th>Zhong et al.’s data (2009)</th>
<th>Base Case or Case 1</th>
<th>Case 3</th>
<th>Case 2</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>nominal pore volume</strong> t₀</td>
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<td><strong>wet front location</strong></td>
<td><strong>pore volume</strong> t₀</td>
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*Evaluated at the injection rate of Qᵢ=8.08 cm³/min (incompressible)

**Evaluated at the injection rate of Qᵢ=4.01 cm³/min (compressed instantaneously at the steady state pressure)
Figure 4.9: Reconstruction of Figure 4.6 using 3 different ways of handling gas compressibility: (a) Case 1, incompressible gas; (b) Case 2, gas being compressed as pressure builds up and (c) Case 3, gas instantaneously compressed at the steady state pressure.

Figure 4.10: Saturation profile at the time of wet front breakthrough: (a) Case 1, incompressible gas; (b) Case 2, gas being compressed as pressure builds up and (c) Case 3, gas instantaneously compressed at the steady state pressure.
Figure 4.11: Change in average water saturation: (a) Case 1, incompressible gas; (b) Case 2, gas being compressed as pressure builds up and (c) Case 3, gas instantaneously compressed at the steady state pressure.

Figure 4.12: Fractional flow solutions (Case 1): See Table 4.3 for inputs and outputs.
Figure 4.13: Fractional flow solutions (Case 2): See Table 4.3 for inputs and outputs.
4.3 Discussion

Figure 4.15 shows the detailed procedures followed in this study to calculate parameters such as \( m, n, A, \text{MRF, } f_w^1, \text{ and } f_w^\text{f}. \) Although this study tries to fit the model into flow experimental data (average water saturation, wet front propagation, foam front propagation, pressure buildup, etc), what happens in reality is to conduct basic laboratory measurements (fluid and sandpack properties, relative permeability functions, surfactant adsorption test, etc) and then predict what might take place during flow experiments. Once experimental parameters such as \( m, n, A, \text{B, and } D_{sf} \) are already available from basic experiments, then the same algorithm, but in
different order, can be used to determine the propagation of foam front and wet front \( (V_{BL1} \text{ and } V_{BL2}) \), pressure history, effluent fluid production, average water saturation and so on.

Overall the outcome of this study based on four cases reproduces Zhong et al.’s data (2009) qualitatively. More accurate fit quantitatively is believed to be hindered by the following reasons. First, flow test data as shown in Figure 4.6 is in some sense a “macroscopic” response (average saturation, front locations, etc.) while the model requires “microscopic” input parameters such as residual saturations, exponents and end-point relative permeability values for Corey-type relative permeability functions, level of surfactant adsorptions and so on. In other words, what this study demonstrates is how important it is to measure all those fundamental parameters before actual laboratory and field-scale flow tests. It is because there are possibly many different combinations of input parameters showing a similar flooding response. The relative permeability at low water saturations (near the initial and injection conditions) should be especially captured well because the overall process heavily counts on the flow physics within that narrow region. Second, gas compressibility is an important experimental parameter and may cause a significant amount of errors, if not handled properly. Even though deep vadose zone remediation is not typically associated with high back pressure, flow experiments for the purpose of extracting foam propagation mechanisms are highly recommended to be performed at high back pressure. Lake (1989) suggests the ratio between steady-state pressure drop and back pressure less than 0.5 for reliable use of fractional flow analysis, but Zhong et al. (2009) has the ratio around 2.27 (i.e., 229 kPa/101 kPa). Third, there are concerns about the accuracy of measured data, not because of experimental errors (which is also possible) but because of difficulties of measurement. For example, the wet front propagation monitored during the
Figure 4.15: The procedure followed to calculate six unknown parameters.

Experiments may not be a true representation because the injected fluid runs faster through the large opening size near the wall (this is why a pack with larger diameters (more than 2 inches) is preferred for visualization experiments), fluid injection has been halted intermittently to measure the weight of the pack during which fluid redistribution might have occurred inside the pack, cumulative water production data – easy to measure and conveniently allowing the accuracy of average water saturation measurement to be checked using material balance – is lacking, initial water saturation was not measured carefully although the change in water saturation (or pack weight) was carefully followed during the flow test. Fourth and last, although
all of the above-mentioned complexities are determined crystal clear, there is still a challenge in determining foam properties for modeling purpose. It is because foam texture builds up slowly with time, which impacts gas-phase mobility reduction factor (MRF), breakthrough time, determination of when to reach the steady state, and so on. This time-dependent foam kinetics should be carefully treated when laboratory experimental data and foam modeling results are extended into field scale tests.

Figure 4.16: Fractional flow solutions for the case with $B = 1.0$ and $m = 1.31$, keeping other conditions identical to Case 1.
Although the end-point gas relative permeability and the exponent of water relative permeability function are assumed to be $B = 1$ and $m = 1$ in Case 1 through 3, they are arbitrarily selected to illustrate how the entire framework of model fit to data works for demonstration.

Figure 4.17: Fractional flow solutions for the case with $B = 0.5$ and $m = 1.34$, keeping other conditions identical to Case 1.

Two other cases are further examined as shown in Figure 4.16 with $B = 1.0$ and $m = 1.31$ and Figure 4.17 with $B = 0.50$ and $m = 1.34$ in order to show the robustness of the model, by keeping other conditions identical to Case 1. The fractional flow solutions in Figs. 4.16 and 4.17 satisfy all input constraints, producing output of $n = 29.59$, $A = 1.01 \times 10^{-3}$, $MRF = 35.0$ and $n = 27.11$, $A = 1.07 \times 10^{-3}$, $MRF = 27.2$, respectively.
The procedure followed so far to fit the model to experimental data uses a weighting to the history prior to the wet-front breakthrough time \( (t_{\text{DwBT}}) \), and let the remaining part determined. Other approaches are also possible, for example, providing equal weighting before and after \( t_{\text{DwBT}} \), dealing with all data points and coming up with least errors involved. In the following paragraphs, such results are reported in the three cases discussed earlier with \( R^2 \) values. The closer \( R^2 \) value to one, the better match exists between the modeling results and experimental data.

![Graphs showing average water saturation](image)

Figure 4.18: Average water saturation of Case 1 with early-time weighting ((a) \( t_{\text{DwBT}}=15, R^2=0.9325 \)) vs. all-time weighting ((b) \( t_{\text{DwBT}}=14.5, R^2=0.9608 \)).
Figure 4.19: Average water saturation of Case 2 with early-time weighting ((a) $t_{DwBT}=12.8$, $R^2=0.9246$) vs. all-time weighting ((b) $t_{DwBT}=11.8$, $R^2=0.9286$).

Figure 4.20: Average water saturation of Case 3 with early-time weighting ((a) $t_{DwBT}=8$, $R^2=0.9236$) vs. all-time weighting ((b) $t_{DwBT}=7.1$, $R^2=0.9310$).
CHAPTER 5
CONCLUSIONS AND RECOMMENDATIONS

This chapter consists of major conclusions and recommendations from this study.

5.1 Conclusions

Chapter 3 shows how the Method of Characteristics can be used to analyze displacement mechanisms of foams in vadose-zone remediation for immobilization and stabilization. The results are presented graphically in order to investigate various design parameters such as initial water saturation ($S_{w1}$), injection foam quality ($f_g^I$), foam mobility reduction factor (MRF), limiting water saturation ($S_{w^*}$), and surfactant adsorption ($D_{sf}$). The major achievements of this study are summarized as follows:

- The Method of Characteristics based on material balance equations is shown to successfully capture the foam delivery mechanisms in vadose-zone remediation, where the initial condition is surfactant-free and dry, and the injection condition is co-injection of gas and surfactant solutions. The presence of three distinct regions (i.e., dry region with mostly gas phase far away from the injection inlet; near wellbore region with injected foams; and a region with high water saturation (i.e., water bank) in between) qualitatively conjectured in previous studies is confirmed.

- The use of surfactant solution exhibiting high MRF, even at high $f_g^I$, and low $S_{w^*}$ is crucial to the overall design process. If these conditions are not satisfied, the water saturation in the foam zone near the well would be high enough to cause downward migration of liquid phase, which might do potentially harm to the entire remediation process.
- A high level of surfactant adsorption (i.e., large $D_{sf}$) was shown to hurt this process not only due to its high chemical costs, but also due to slow propagation of foam front. This implies that the ideal surfactant for vadose-zone remediation should be a good foamer with high stability with a minimal level of surfactant adsorption onto the soil and rock surfaces.

- Although a dry injection condition (i.e., high $f_{g}^{J}$) is required in general in order to reduce water bank size and foam-zone water saturation, it might deteriorate the process significantly due to too slow propagation of foam front. The optimum injection foam quality should be designed taking these two conditions (i.e., water bank size and propagation rate) into consideration. This optimum injection condition is found to depend strongly on other field and design parameters. For example, as the level of adsorption increases, the optimum injection foam quality is shown to decrease (i.e., wetter foam is required).

- The initial water saturation of the medium plays a significant role because it is directly related to water-bank size ahead of foams injected. The implication of water-bank size in actual field applications should be carefully investigated in multi-dimensional experimental and/or simulation studies.

- Overall, this study demonstrates the complexity of foam delivery mechanisms for immobilization and stabilization. Because most field and foam parameters are interconnected in a complicated manner, discussions on the optimum field applications will be very field-specific. The influencing parameters will be, not limited to, surfactant formulations and concentrations, soil types and mineralogy, pore size and size distribution, wettability of the medium, level of heterogeneity, surfactant adsorption,
interaction between foams and medium, ground water chemistry, size and depth of the area of interest, and foam injection quality and strategy.

Chapter 4 demonstrates how the foam model developed in chapter 3 can be applied to make a fit to a set of existing laboratory flow tests (Zhong et al., 2009) such as history of inlet pressure, wet front and foam front locations, and average water saturation. The major outcome is summarized as follows:

- This study shows the procedures to follow, in a step-by-step manner, how MoC-based foam fractional flow solutions can be constructed to make a fit to typical laboratory flow experiments. Although the solution algorithm may change depending on the input and output parameters, the mathematical framework still remains the same.
- Qualitatively fitting the foam model to flow data such as wave propagation, saturation, and pressure history is not a difficult task; exactly which combination of basic input parameters should be used is a challenging task, however. It is because different combinations of input parameters can lead to the same flow response. This implies that conducting experiments to determine basic parameters prior to flow tests is an essential step. Such parameters include, not limited to, surfactant adsorption ($D_{sf}$), initial and residual saturations of water and gas phases ($S_{w}^{I}$, $S_{w}^{R}$, $S_{g}^{I}$, $S_{g}^{R}$), and coefficients (A, B) and exponents (m, n) of Corey-type relative permeability functions. It is extremely important to capture the flow behavior at low water saturation ($S_{w}$), because the fluid/fluid and fluid/rock interactions at very dry condition (near the initial and injection conditions) have a paramount influence on the overall foam displacement mechanism.
- The compressibility of gas phase in laboratory flow tests can have a huge impact on the outcome of foam fractional flow solutions, especially wave velocities and breakthrough
times, as demonstrated by three different cases (Cases 1 through 3). Such a complexity caused by gas compressibility is believed to be overcome by conducting laboratory flow experiments at evaluated back pressures. Foam kinetics, foam texture building up slowly with time to reach its steady-state value, obviously plays an important role, making the interpretation more complicated.

5.2 Recommendations

Based on the outcomes obtained from this study, the following recommendations for future study can be made:

- Different surfactant formulations and sediments result in different levels of adsorption. Hence, a thorough experiment measuring the adsorption for the surfactant concentration and formulation of interest and Hanford sediment is required for accurate modeling. Measuring relative permeability is another essential step.

- Gas compressibility plays a very important role in this process. As a result, construction of a numerical simulator which could take care of gas compressibility together with slow foam kinetics is advised. Mechanistic foam simulation can be a good solution to this problem.

- Conducting foam flow modeling and simulation in a large scale and multidimensional space, as an extension of this study, is recommended prior to field-scale treatment.

- Although the mathematical model introduced in this study can give great insights on the process of foam-assisted remediation in deep vadose zone, this model has some limitations originated from fractional flow analysis. They include, not limited to 1D flow, Newtonian rheology, instantaneous reach of steady state, homogenous porous media, no
chemical or biological reaction, negligible dissipative effects of capillary pressure, fluid compressibility, and dispersive effects.

- The developed model can be improved and expanded by including the chemical and biological reactions and dispersion. The model could also be further expanded to investigate how dispersion can affect this process. Furthermore, the model can be taken into 3D space and utilized to probe the multidimensional phenomena such as fingering and channeling.

- It is important to understand parameters which can, or cannot, be manipulated in the design of field applications. Example parameters that can be manipulated include surfactant formulation and concentration to come up with optimal MRF and surfactant adsorption, injection conditions, well patterns and spacing. On the other hand, parameters such as initial water saturation, residual water and gas saturations, and the relative permeability are dictated by the formation and there is not much of room to manipulate these parameters during the design process.
REFERENCES


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VITA

Alireza Roostapour was born in June 1980 Shiraz, Iran. He studied chemical engineering as an undergraduate student at Amirkabir University of Technology in Iran and received his bachelor’s degree in 2002. Alireza got an admission from the department of petroleum engineering at Colorado School of Mines in 2003. He received his master degree in petroleum engineering in 2005. After that he returned to his home country Iran and worked as a reservoir engineer for 3 years. In spring 2009 he travelled back to the US to obtain his PhD in petroleum engineering at the Louisiana State University. His research focuses on foam flow in porous media and its application in petroleum and remediation industry.