# COMPLEX FLUIDS IN POROUS MEDIA: PORE-SCALE TO FIELD-SCALE COMPUTATIONS

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To my grandma and uncle.

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### ABSTRACT

Soroush Aramideh Ph.D., Purdue University, December 2019. Complex Fluids in Porous Media: Pore-scale to Field-scale Computations. Major Professors: Arezoo M. Ardekani, Pavlos P. Vlachos.

Understanding flow and transport in porous media is critical as it plays a central role in many biological, natural, and industrial processes. Such processes are not limited to one length or time scale; they occur over a wide span of scales from micron to Kilometers and microseconds to years. While field-scale simulation relies on a continuum description of the flow and transport, one must take into account transport processes occurring on much smaller scales. In doing so, pore-scale modeling is a powerful tool for shedding light on processes at small length and time scales.

In this work, we look into the multi-phase flow and transport through porous media at two different scales, namely pore- and Darcy scales. First, using direct numerical simulations, we study pore-scale Eulerian and Lagrangian statistics. We study the evolution of Lagrangian velocities for uniform injection of particles and numerically verify their relationship with the Eulerian velocity field. We show that for three porous media velocity, probability distributions change over a range of porosities from an exponential distribution to a Gaussian distribution. We thus model this behavior by using a power-exponential function and show that it can accurately represent the velocity distributions. Finally, using fully resolved velocity field and pore-geometry, we show that despite the randomness in the flow and pore space distributions, their two-point correlation functions decay extremely similarly.

Next, we extend our previous study to investigate the effect of viscoelastic fluids on particle dispersion, velocity distributions, and flow resistance in porous media. We show that long-term particle dispersion could not be modulated by using viscoelastic fluids in random porous media. However, flow resistance compared to the Newtonian case goes through three distinct regions depending on the strength of fluid elasticity. We also show that when elastic effects are strong, flow thickens and strongly fluctuates even in the absence of inertial forces.

Next, we focused our attention on flow and transport at the Darcy scale. In particular, we study a tertiary improved oil recovery technique called surfactant-polymer flooding. In this work, which has been done in collaboration with Purdue enhanced oil recovery lab, we aim at modeling coreflood experiments using 1D numerical simulations. To do so, we propose a framework in which various experiments need to be done to quantity surfactant phase behavior, polymer rheology, polymer effects on rock permeability, dispersion, and etc. Then, via a sensitivity study, we further reduce the parameter space of the problem to facilitate the model calibration process. Finally, we propose a multi-stage calibration algorithm in which two critically important parameters, namely peak pressure drop, and cumulative oil recovery factor, are matched with experimental data. To show the predictive capabilities of our framework, we numerically simulate two additional coreflood experiments and show good agreement with experimental data for both of our quantities of interest.

Lastly, we study the unstable displacement of non-aqueous phase liquids (e.g., oil) via a finite-size injection of surfactant-polymer slug in a 2-D domain with homogeneous and heterogeneous permeability fields. Unstable displacement could be detrimental to surfactant-polymer flood and thus is critically important to design it in a way that a piston-like displacement is achieved for maximum recovery. We study the effects of mobility ratio, finite-size length of surfactant-polymer slug, and heterogeneity on the effectiveness of such process by looking into recovery rate and breakthrough and removal times.

## 1. INTRODUCTION

Flow and transport of chemicals in porous media is a critical aspect of many biological, natural, and industrial processes. Examples of such processes are subsurface transport of containment and multi-phase flow during enhanced oil recovery or  $CO_2$ sequestration. In this thesis, we study such processes at two different scales, namely pore-scale and continuum-scale.

At pore-scale, flow and transport can be described by Navier-Stokes and advectiondiffusion equations, respectively. Moreover, one needs the exact description of poregeometry and apply proper boundary conditions on the fluid-solid interface. In the case of particle tracking and particle flows, interactions of particles with solid surfaces and Brownian motion needs to be taken into account. Pore-scale modeling is inherently a computationally expensive approach and has recently become a promising tool with the increase in computational power and development of advanced numerical techniques. Thus, our work in this area requires development and usage of high-performance codes and tools to solve all the relevant physics at the smallest scales (high-fidelity).

In the first chapter of this thesis, we report on the effects of different porestructures on Eulerian and Lagrangian statistics of flow and transport through porous media. We first construct two porous media by means of an assemblage of mono and polydisperse hard spheres and overlapping spheres. These two different protocols allow for constructing two very distinct pore-structures and systematic study of the effects of pore-structure on the flow and transport. Then, using pore-scale simulations and particle tracking, we derive different Eulerian and Lagrangian statistics for these two types of porous media.

In chapter 2, we extend our previous studies on flow and transport in Newtonian fluids to non-Newtonian and viscoelastic fluids. As we see later, polymeric fluids and viscoelastic solutions are ubiquitous in flow through porous media applications, especially the one in enhanced oil recovery, which we will study in detail. Viscoelastic fluids could enhance particle dispersion and change flow resistance, either of which could have a huge impact on transport processes. We study the impact of viscoelastic fluids on the particle dispersion and flow resistance using pore-scale simulations.

On continuum level, under certain circumstances, single (or multi-) phase flow is described by Darcy's law. In the case of a single-phase flow, Darcy's law conveniently relates fluid flux to the pressure gradient. In the case of multi-phase flows, other state variables such as capillary pressure and relative permeability arise and are modeled as functions of saturation. In chapters 3 and 4, we have investigated three-phase and two-phase flows arising due to the injection of surfactant-polymers into an oil reservoir at the Darcy-scale.

In the third chapter of this thesis, we focus on modeling of a tertiary oil recovery technique called surfactant-polymer flooding through a series of laboratory experiments and numerical simulations. We lay out a procedure in which, first by a series of experiments, one can understand the phase behavior of water/oil/surfactant systems, dispersion, polymer rheology, etc. Then by means of sensitivity analysis and a multi-stage algorithm, two critically important quantities, namely cumulative oil recovery factor and maximum pressure drop, are matched with those of experiments.

In the fourth chapter of this thesis, we numerically investigate surfactant-polymer flooding applied to a 2-D domain to displace a secondary immiscible phase. Surfactant and polymer are injected as additives to injected water (brine) for a finite time, which corresponds to a finite-size chemical slug. We study the instabilities occurring at leading and trailing edges of this chemical slug during displacing a secondary immiscible fluid (e.g., oil). It is of critical importance to protect such chemical slugs from severe viscous fingering and distortion so that a piston-like displacement is achieved for maximum recovery.

# 2. PORE-SCALE STATISTICS OF FLOW AND TRANSPORT THROUGH POROUS MEDIA

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## 2.1 Abstract

Flow in porous media is known to be largely affected by pore morphology. In this work, we investigate the effects of pore geometry on the transport and spatial correlations of flow through porous media in two distinct pore structures arising from three dimensional assemblies of overlapping and non-overlapping spheres. Using high resolution direct numerical simulations (DNS), we perform Eulerian and Lagrangian analysis of the flow and transport characteristics in porous media. We show that the Eulerian velocity distributions change from nearly exponential to Gaussian distributions as porosity increases. A stretched exponential distribution can be used to represent this behavior for a wide range of porosities. Evolution of Lagrangian velocities is studied for the uniform injection rule. Evaluation of tortuosity and trajectory length distributions of each porous medium shows that the model of overlapping spheres results in higher tortuosity and more skewed trajectory length distributions compared to the model of non-overlapping spheres. Wider velocity distribution and higher tortuosity for overlapping spheres model give rise to non-Fickian transport while transport in non-overlapping spheres model is found to be Fickian. Particularly, for overlapping spheres model our analysis of first passage time distribution shows that the transport is very similar to those observed for sandstone. Finally, using 3-D velocity field obtained by DNS at the pore-scale, we quantitatively show that despite the randomness of pore space, the spatially fluctuating velocity field and the 3-D pore-space distribution are strongly correlated for a range of porous media from relatively homogeneous monodisperse sphere packs to Castlegate sandstone.

### 2.2 Introduction

Understanding the flow through porous media is of great importance in many natural and technological processes such as oil recovery [1], CO<sub>2</sub> sequestration [2], filtration [3], biological flows [4], and reactive transport [5]. In many practical cases, low Reynolds number flows through porous media are modeled at the Darcy scale using Darcy's law which relates the specific discharge, q, to pressure drop,  $\Delta p$ , along the flow direction as:

$$q = \frac{-\kappa}{\mu} \frac{\Delta p}{L} \tag{2.1}$$

where  $\kappa$  is the permeability,  $\mu$  is the viscosity of the fluid, and L is the length of medium along the flow direction. In flow through porous media, intricate pore geometry creates a complex and spatially fluctuating velocity field and it is well-known that such flow variations at pore-level affect the transport processes in porous media and give rise to many anomalous behaviors which macroscopic models fail to predict, such as early breakthrough of the solute [6] or mixing-controlled reactions in heterogeneous media [7]. Naturally, it is of great interest to relate such complex velocity fields and resulting macroscopic transport properties to pore geometry.

Recent advances in porous media imaging techniques such as X-ray micro-tomography and pore-scale simulations have allowed for resolving the flow at the pore-scale for complex and realistic pore geometries. Computationally, fluid flow at pore-scale is investigated either through direct pore-scale or pore-network models. In the latter approach, the pore structure is approximated by a network of pores connected by throats. As a result, pore-network models do not preserve the original pore-space features. A review of pore-scale analysis using pore-network models is given in [8]. Direct pore-scale modeling, on the other hand, is achieved by solving governing equations directly on the pore-structure [9]. Recently, a direct comparison of velocity fields obtained by particle image velocimetry (PIV) and magnetic resonance imaging (MRI) with pore-scale simulations suggests that given an accurate representation of the pore geometry, DNS could predict accurately flow and transport at the pore-scale [10].

Pore-scale modeling has been intensively used to study the effects of pore geometry on transport properties such as hydraulic permeability [11–14], heat transfer [15] and solute dispersion [16, 17]. For example, studies of Daneyko *et al.* [18] and Vidal *et al.* [19] suggest negligible effects of grain size distribution on the hydraulic permeability, whereas Garcia et al. [20] showed that the grain shape is a key parameter and could change permeability by a factor of 2. In a similar study, Pan *et al.* [21] proposed improvements to the existing empirical permeability relationships for sphere packing with various porosities and polydispersities. Contributions of flow inertia to permeability in regular and random sphere arrangements were evaluated in [22]. It was shown that the pore structure plays an important role on the drag force acting on the sphere packing along with the solid volume fraction and the direction of the flow. Anisotropy of the porous medium and its effects on the permeability were studied by Stewart *et al.* [23]. A correlation between the mean tortuosity and the porosity was proposed in stochastically generated porous media in [24].

Many recent observations have shown that displacement of solute could not be accurately predicted by advection-dispersion equation (ADE) [25] and this discrepancy is often attributed to the extreme heterogeneity of the medium and significant changes in the local velocity field [26]. The signature of anomalous transport is the power-law tail of solute concentration long after its breakthrough [27]. Different stochastic models have been developed to address this anomalous behavior such as mobile-immobile concept [28], multiple-rate transfer models [29], fractional ADE [30], and continuous time random walk (CRTW) [31, 32]. In the CTRW framework, the transport of solute is greatly affected by the behavior of transit time distribution,  $\psi(t)$ , where it is generally assumed that  $\psi(t) \sim t^{-1-\beta}$ . For example, truncated power-law (TLP) distribution for  $\psi(t)$  has been particularly successful at describing anomalous transport [33]. However, the key parameter  $\beta$  in  $\psi(t)$ , also a measure of medium heterogeneity, still needs to be determined. In fact, effective parameterization of such models requires knowledge of Eulerian and Lagrangian statistics of flow through porous media. For example, given the physical interpretation of  $\psi(t)$ , it can be determined from pore-scale velocity field [34,35]. Bijeljic and Blunt [36] used pore-network model of a Berea sandstone to determine transient time distribution where they observed a power-law distribution of pore-to-pore travel times.

Importance of detailed knowledge about underlying velocity distributions and correlations in porous media due to their impact on transport behavior and hydraulic properties of the media has led to intensive research focused on measuring velocity probability density functions (PDF) and its correlations through experimental and numerical techniques [37–39]. A combination of log-normal and exponential functions has been suggested [40] to describe the velocity distribution of slow flow in monodisperse bead packing for the low- and high-velocity regions of the velocity distribution, respectively. Velocity distribution for low and moderate Reynolds number flows were reported by Icardi *et al.* [41] in porous media generated by irregular nonconvex polydisperse objects. They obtained almost symmetric distribution for transverse components of velocity with increasing variance with respect to Reynolds number. It was pointed out that the streamwise velocity distribution is highly skewed for low Darcy velocities showing an intensified negative tail as Darcy velocity increases, indicating the existence of recirculation zones. Recently, it was shown that for two-dimensional models of fibrous material, PDFs of all velocity components follow a power-law with some tuning parameters [42]. In an experimental study by Patil and Liburdy [43], they observed in planes near the bed walls where flow is nearly two-dimensional, porespace and velocity autocorrelations feature the same patterns but such patterns vanish in central planes. Using confocal microscopy and PIV, Datta et al. [44] showed that in a channel comprised of glass beads, 2-D velocity components follow an exponential behavior. Moreover, they presented strong relationship between spatial fluctuations of 2-D velocity field with pore-space geometry.

In this work, we study Eulerian and Lagrangian velocity PDFs by means of high resolution direct numerical simulation of Navier-Stokes equations at the pore-scale. Our emphasis is on characterizing statistics of flow and transport and their relationships to pore-space. We study how Eulerian velocity PDFs vary with respect to medium properties (i.e. porosity) and their correlations for two distinct models of porous media namely hard-sphere and overlapping sphere models of porous media. Later on, we study Lagrangian velocity PDFs sampled isochronally and equidistantly along the particle trajectory and their evolutions with time and space. We investigate the Lagrangian velocity PDFs and their relationship with their Eulerian counterpart. Transport properties of porous media are obtained using a streamline approach through analyzing first passage time distributions and mean tortuosity. We show that that wider Eulerian velocity PDFs and higher tortuosity for overlapping sphere system yield in more anomalous transport compared to hard-sphere system where Eulerian velocity PDFs and tortuosity show narrower distributions and smaller values, respectively. Finally, we analyze the correlation functions of pore-space and Eulerian velocity field and show that not only for rather homogeneous porous media comprised of spheres in 3-D, but also for natural rocks there exists a strong relationship between these correlation functions.

### 2.3 Numerical method

The fluid flow is described by the mass and momentum conservation equations:

$$\nabla \cdot \mathbf{u} = 0 \tag{2.2}$$

$$\frac{\partial(\rho \mathbf{u})}{\partial t} + \nabla \cdot (\rho \mathbf{u}\mathbf{u}) = -\nabla p + \nabla \cdot (\rho \nu \nabla \mathbf{u})$$
(2.3)

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Conservation equations are solved using finite volume method (FVM) and projection method [45] by using PArallel Robust Interface Simulator (PARIS) [46]. Eq. (2.3) is discretized on a staggered grid configuration where no-slip and no-penetration boundary conditions are enforced on the fluid-solid interface. Convection and diffusion terms in Eq. (2.3) are discretized using QUICK and central difference schemes [47], respectively and time integration is performed using Euler explicit scheme. The resulting Poisson equation is solved by Biconjugate gradient stabilized method (BiCGSTAB) with the residual tolerance of  $10^{-8}$ . The computational domain is a cube where periodic boundary condition is prescribed on all boundaries. A constant pressure gradient (i.e. body force per unit mass) is applied to induce the flow from left to right in xdirection. In many applications of flow through porous media such as enhanced oil recovery, given the physical properties of water, typical flow rate of 1ft/day (u) and average pore size of 10 microns (l) results in Re << 1 where Re  $= \frac{\rho u l}{\mu}$ . In this paper, we study flow and transport through porous media in the creeping flow regime  $(\text{Re} \ll 1)$ . The results are reported after steady-state condition is achieved based on criterion given by Botella and Peyret [48].

### 2.3.1 Validation

In this section we show the validation of our simulations. For the purpose of validation, we compare our results with experimental data by Suekane *et al.* [49] and numerical results the same experiment by Finn and Apte [50]. In the experiment, pore-scale velocity measurements were carried out for flow packed bed using magnetic resonance imaging (MRI). Figure 2.1 shows a comparison for the normalized velocity. As can be seen, there is an excellent agreement between predicted velocity profile and experimental data. Moreover, Figure 2.1 shows the effect of grid resolution on the predicted velocity profile. Additionally, we obtained  $k/D^2 = 6.82 \times 10^{-4}$  for the permeability of monosized sphere pack for  $\phi = 35.8\%$  which is close to  $k/D^2 =$ 

 $6.8 \times 10^{-4}$  reported by Bryant and Blunt [51] for the same system and for  $\phi = 36.2\%$ .



Fig. 2.1.: (a): comparison of interstitial velocity profiles at Re = 105.57 with experimental data of Suekane *et al.* [49] ( $\bigcirc$ ) and numerical results of the same experiment by Finn and Apte [50] (--). – simulation data with grid spacing of  $\Delta/D = 56$ ,  $\cdot - \cdot$  simulation data with grid spacing of  $\Delta/D = 28$ . (b): schematic of the experimental setup of Suekane *et al.* Simulation is performed with 4 lateral faces as wall and inflow and outflow boundary conditions for bottom and top faces, respectively, according to the experiment of Suekane *et al.* Normalized velocity is measured along the centerline of the plane shown in white (along x axis). Solids are colored gray.

## 2.3.2 Porous media

In this work, we build porous media via arranging spheres in 3-D space. A wide range of real heterogeneous porous media can be represented by considering two distinct categories of packing, (a) impenetrable (hard-) sphere model and (b) fully-penetrable (overlapping) sphere model [52]. In the latter model, also known as boolean model, spheres centers are points of a stationary Poisson process. Boolean model is an extreme case of a penetrable-concentric-shell (cherry-pit) model where impenetrability parameter is zero. In the case of hard-sphere model, the impenetrability constraint makes it difficult to analyze and generate such systems specially at high

solid volume fractions [53]. There are a number of techniques for generating (jammed) packing of hard-spheres. In this work, we adopted the collision-driven molecular dynamics algorithm of [54] which is essentially a generalization of Lubachevski-Stillinger (LS) algorithm [55]. In LS algorithm, spheres are initially added sequentially in a periodic box at a small volume fraction. Spheres then evolve in time while their radii grow at rates proportional to their radii until the desired packing is achieved. Polydispersivity gives more flexibility in representing a wide class of microstructures. Thus, we further generalized this algorithm based on the work of [56] to generate dense polydisperse sphere packings. In the case of polydisperse spheres with M classes of spheres, density function of sphere radii is given by:

$$f(r) = \sum_{j=1}^{M} y_i \delta(r - r_i)$$
(2.4)

where  $y_i = \frac{N_i}{N}$  and harmonic mean particle diameter  $\overline{D}$  is defined as:

$$\overline{D} = \frac{\sum_{j=1}^{M} N_i D_i^3}{\sum_{j=1}^{M} N_i D_i^2}$$
(2.5)

while  $\langle D \rangle = \sum_{j=1}^{M} y_i D_i$ . Since particle size distribution of many granular systems is found to be approximately log-normal [57], spheres radii probability density function takes the form:

$$f(r) = \frac{1}{r\sigma\sqrt{2\pi}} \exp\left[-\frac{(\ln(r) - \mu)^2}{2\sigma^2}\right]$$
(2.6)

where  $\sigma$  and  $\mu$  are variance and average of  $\ln(r)$ , respectively. In LS algorithm, the initial configuration, expansion rate, and initial distribution of particle velocities affect the final packing. It is also worth noting that the expansion rate must be chosen carefully as small values lead to local crystallization while a relatively large ones will not produce random close packing. Finally, we build the overlapping sphere model by placing mono-sized spheres in random locations within the computational domain sequentially until the desired porosity is achieved. In this study, we use three models of porous media including monodisperse hard-spheres, polydisperse hard-spheres, and overlapping spheres with different porosities by changing the number of spheres in the system where all the porous media have the same harmonic mean particle diameter  $(\overline{D})$  (Figure 2.2).



Fig. 2.2.: Three dimensional periodic assemblies of spheres and cross sectional views of indicator function (solid or pore space) for (a,b) monodisperse hard-spheres, (c,d) polydisperse hard-spheres, and (e,f) (monodisperse) overlapping spheres. Pore space is colored black.

## 2.3.3 Characterization of porous medium

Characterization of the microstructure of porous media plays an important role in determining macroscopic properties such as residual saturation of nonwetting phase, permeability, reaction constant, etc. [58]. Microstructure of porous media can be characterized by n-point probability functions [59], Voronoi statistics [60], fractal geometry [61,62], local-porosity theory [63], Minkowski functional [64], and pore size distribution [65], to name a few. Our aim here is to connect our findings about flow in porous media to such statistical descriptors. Each realization of the porous media is a domain of space  $\mathcal{V} \subseteq \mathbb{R}^3$  consisting of two disjoint phases namely pore and grain. Thus, each phase could be identified through an indicator function  $\mathcal{I}^i(r)$  for phase *i* as:

$$\mathcal{I}^{i}(r) = \begin{cases} 1 & r \in \mathcal{V}_{i} \\ 0 & \text{otherwise} \end{cases}$$
(2.7)

Complete statistical description of a medium is then obtained by means of n-point correlation function  $S_n^i$  given as:

$$S_n^i(r_1, r_2, \dots, r_n) = \langle \prod_{j=1}^n \mathcal{I}^i(r_j) \rangle$$
(2.8)

where  $\langle \cdots \rangle$  denotes the ensemble average over all the possible positions r and  $S_n^i$  defines the probability of finding  $r_1, r_2, \ldots, r_n$  all in the phase i. For statistically isotropic material (i.e. joint probability distributions are invariant of coordinates rotation) n-point correlation function depends only on the relative position of points rather than their absolute positions [66]. Hereafter, for simplicity, we drop the superscript i and let  $\mathcal{I}$  to be indicator function of the pore phase. Perhaps the simplest and most important characteristics of a porous medium is its porosity (i.e. the ratio of pore volume to total volume) and is given by the one-point correlation function as:

$$S_1(r) = \langle \mathcal{I}(r) \rangle = \phi \tag{2.9}$$

Among various statistical descriptors of random media, in this study we focus on twopoint correlation functions as they arise in analytical estimations of many physical properties of random media and reveal their global structures [59]. These functions have been extensively used to characterize the porous media [67,68]. They could be used to estimate upper and lower bounds for fluid permeability, electrical conductivity, and magnetic fields [69]. Recently, Jiao *et al.* [70] used two-point correlation functions to model heterogeneous two-phase media. However, it is worth noting that two-point correlation functions cannot be sufficient alone to completely determine a medium [71] (i.e. for capturing long-range connectivity higher order statistics are required).

For two-phase medium two-point correlation functions have limiting behavior as [66, 68]:

$$\lim_{r \to 0} S_2(r) = \phi \tag{2.10}$$

$$\lim_{r \to \infty} S_2(r) = \phi^2 \tag{2.11}$$

For isotropic two-phase medium, Debye *et al.* [72] showed that derivative of  $S_2(r)$  at origin (r = 0) is in direct relationship with the specific surface area (s) of the medium as:

$$\lim_{r \to 0} S_2'(r) = -s/4 \tag{2.12}$$

This relationship was later generalized for anisotropic media by Berryman [73].

Blair *et al.* [67] showed that two-point correlation functions could also be used to obtain estimations of effective pore size and mean grain diameter. Additionally, the shape of two-point correlation functions reflects an underlying medium morphology. Thus,  $S_2(r)$  plays an important role in this work as we later show that structure of  $S_2(r)$  can determine that of velocity fluctuations in porous media regardless of how the porous media is constructed. Shown in Figure 2.3 is  $S_2$  for our three models of porous media.

## 2.3.4 Determining representative elementary volume (REV)

Modeling of fluid flow at pore-scale or imaging porous media is limited to small samples. Thus, it is important to show that such data sets is representative of larger scale systems. The prerequisite for any analysis of porous medium for calculating macroscopic properties, is evaluating the representative elementary volume of such medium. The concept of REV has different definitions in the literature [74], however, it could be regarded as a length-scale at which flow obeys Darcy's law and macroscopic



Fig. 2.3.: Different models of porous media and their two-point correlation functions. Significant difference in the structure of  $S_2(r)$  is an indication of different morphology of porous media models. In hard-sphere systems (a), oscillations in  $S_2(r)$  is due to exclusion volume effect. These oscillations becomes weaker in the case of polydisperse hard-spheres (b) and completely vanishes for overlapping spheres (c). Different statistics could be obtained simply by calculating  $S_2(r)$ , as shown in the figure.

properties such as porosity and permeability are defined using volume averaging. Many different procedures exist for determining REV [74]. In this work we determine "statistical" REV with slight modifications in the procedure proposed in [75]. It is worth mentioning that in the statistical approach a sample is called REV if the mean and variance of the quantities of interest (e.g. porosity, specific surface) falls below a certain value [21]. Here, instead of generating a reference domain and then dividing it to smaller subdomains for further analysis, we increase the domain size incrementally and generate 20 independent packings for each increment in domain size. We believe this procedure provides a better way of doing such an analysis as (i) every single medium satisfies periodicity in all directions which is consistent with boundary conditions used in numerical simulations, (ii) different packings generated for each domain size allows for variability in packing structure even when porosity is kept constant (i.e. we randomly choose the parameters affecting packing, see §2.3.2), (iii) this procedure eliminates boundary effects (e.g. wall) and the need for adding buffer layers to the outlet and inlet. We studied different REVs for porosity, specific surface area, and permeability and found that permeability gives the largest REV. Here, we define REV where coefficient of variation of permeability falls below 3%. Figure 2.4a shows that domain sizes with  $L/\overline{D} \geq 5.0$ , satisfies this criteria and thus are considered REV. Empirical variograms are useful in determining REV as they show the correlation of data with distance [76]. Thus, to further analyze and quantify how large our domain is with respect to spatial correlations and different length scales of the medium, we evaluated the empirical variograms of porosity and velocity in the direction of flow as:

$$\gamma_p(h) = \frac{\sum_{1}^{N} (I(x_i + h) - I(x_i))^2}{2N},$$
(2.13)

$$\gamma_u(h) = \frac{\sum_{1}^{N} (u(x_i + h) - u(x_i))^2}{2N}$$
(2.14)

where I is the indicator function for porosity  $(I(x_i) = 1 \text{ for pore space and } I(x_i) = 0 \text{ for solid space})$  and N is total number of points with the distance h. Figure 2.4b suggests that beyond one particle diameter  $(\overline{D} \geq 1)$ , variograms of porosity and velocity become uncorrelated and hence our porous medium represents larger samples [77]. Table 2.1 summarizes values of two different characteristic length scales namely, mean pore diameter  $(l_c)$  and integral length scale  $(\lambda)$  and ratios of domain size to these length scales. Mean pore diameters are calculated based on two-point correlations functions (see Figure 2.3) as  $\frac{\phi(1-\phi)}{S'_2(0)}$  (i.e., intersection of  $S_2 = \phi^2$  and a tangent to  $S_2(r)$  at r = 0). Integral length scale of the spatial structure of each medium is calculated as:

$$\lambda = \int_0^\infty S_2(r)dr \tag{2.15}$$



Fig. 2.4.: Representative elementary volume calculation: (a) Variation of permeability with respect to domain size. (b) Variogram of porosity and velocity in the direction of flow normalized by their theoretical asymptotic values  $\gamma_{\infty} = \phi(1 - \phi)$  for porosity and  $\gamma_{\infty} = \langle u^2 \rangle - \langle u \rangle^2$  for velocity variograms. Variograms are plotted as functions of distance from the inlet face. Data is for the monodisperse hard-sphere model at  $\phi = 0.40$ .

Table 2.1.: Mean pore size  $(l_c)$  and integral length scale  $(\lambda)$  of non-overlapping and overlapping systems.

porous medium	$\phi$	$l_c$	λ	$L/l_c$	$L/\lambda$
monodisperse hard-spheres	0.45	0.29	0.14	27.58	57.14
polydisperse hard-spheres	0.45	0.17	0.15	47.00	53.33
overlapping spheres	0.45	0.50	0.30	16.00	26.66

#### 2.4 Lagrangian statistics

In this section we study Lagrangian-based statistics of flow through porous media. Lagrangian description of flow field is obtained by means of particle tracking in the steady-state Eulerian flow field. In each simulation,  $10^5$  tracer particles are introduced on the inlet face uniformly. Particle trajectory is then obtained via integration of Eq. (2.16) as:

$$\frac{d\mathbf{x}(t)}{dt} = \mathbf{v}(\mathbf{x}(t)), \mathbf{x}(t_0) = \mathbf{x}_0$$
(2.16)

where a third order Runge-Kutta scheme is used for time integration and a trilinear interpolation scheme is used to locally interpolate particle velocity from fully resolved Eulerian field. We also performed computations where we used  $10^4$  and  $5 \times 10^4$  particles and confirmed that  $10^5$  particles gives converged statistics. Particle tracking enables us to study useful quantities such as trajectory length, breakthrough curves, and tortuosity. Figure 2.5 shows variations of trajectory length distribution for three



Fig. 2.5.: Variation of trajectory length distribution with respect to porosity for (ac) monodisperse hard-sphere, (d-f) polydisperse hard-sphere, and (g-h) overlapping sphere models of porous media.

models of porous media. For all the three types of porous media, trajectory length distributions are highly conditioned by porosity. Interestingly, trajectory length distributions for the hard-spheres model exhibit less skewness compared to those of overlapping spheres at the same porosity where long-tailed particle trajectory length distribution is observed. Such long tails signify highly tortuous pathways due to closing off of pore throats as spheres overlaps in the overlapping spheres model whereas in porous media comprised of hard-spheres pore space is hydrodynamically well connected. Overall, it is clear that trajectory length distributions are different for each case such that as we move from overlapping sphere to hard-sphere model and from low to high porosities, the mean trajectory length as well as its variation reduces (Table 2.2). From the data in Figure 2.5, we can calculate the tortuosity for different

Table 2.2.: Mean, standard deviation (SD), minimum, maximum, and skewness of trajectory lengths for different monodisperse models of porous media and porosities.

porous medium	$\phi$	mean	SD	min	max	skewness
hard-sphere	0.36	10.09	0.49	8.74	12.60	0.57
	0.45	9.65	0.39	8.57	11.45	0.27
	0.60	9.29	0.36	8.47	10.71	0.45
overlapping	0.36	10.86	1.01	8.97	16.08	1.23
	0.45	10.33	0.79	8.69	13.69	0.91
	0.60	9.57	0.55	8.50	12.18	0.75

models of porous media. Elongation of streamlines in flow through porous media, captured by tortuosity, impacts medium's permeability and its transport properties and thus has been studied intensively in the literature [78–85]. In this work tortuosity is defined as:

$$T = \frac{\langle \lambda \rangle}{L} \tag{2.17}$$

where  $\langle \lambda \rangle$  is the average trajectory length traveled by all the particles while no flux weighting is involved and L represents the straight line connecting injecting and absorbing sites (i.e. inlet and outlet boundaries here). Generally, obtaining fluid trajectories to be used in Eq. (2.17) is notoriously difficult both experimentally and numerically in complex geometries. Alternatively, it has been shown that tortuosity could be obtained by calculating  $\frac{\langle u \rangle}{\langle u_x \rangle}$  [82]. This simple equation, however, has limitations due to existence of recirculation zones in flow through porous media as mentioned by Duda *et al.* [80]. Here we extended the work of Matyka *et al.* [83] by directly calculating tortuosity in our two distinct classes of porous media. Figure 2.6 shows the variation of tortuosity calculated using Eq. (2.17) for porous media comprised of hard-spheres and overlapping spheres. We found that tortuosity scales well with porosity for the range of porosity  $0.36 < \phi < 0.60$  according to two empirical relations [79, 86] as:

$$T = 1 - p\ln\left(\phi\right) \tag{2.18}$$

$$T = 1 - p(1 - \phi) \tag{2.19}$$

where p is a constant parameter equals to 0.26 and 0.55 for hard-sphere and overlapping sphere models, respectively. The values reported here for hard-sphere systems is in excellent agreement with tortuosities reported by Muljadi *et al.* [87] for the same medium using the indirect formula of Koponen *et al.* [82] as well as geometric tortuosity values reported by Sobieski *et al.* [85]. Since streamlines are uniformly initiated at the inlet plane, we expect tortuosity values reported here to be even smaller than when streamlines are initiated in a flux-weighted fashion. It was also found that polydispersivity has negligible effect on tortuosity. Tortuosity is linked to transport phenomena in porous media and its macroscopic properties such as dependence of permeability in Kozeny-Carman theory on tortuosity [41, 80, 88]. It is thus expected that different values of tortuosities result in significance differences in transport properties. To investigate the effect of our two distinct pore-structures on the nature of transport, we have studied the transport of solute particles using streamline approach in purely advective flows (in the absence of diffusion).

Here, we calculated the first passage time of solute particles, which is the travel time required for a particle to percolate through the porous medium. First passage


Fig. 2.6.: Variation of tortuosity with different models of porous media and porosity. Error bars for the case of hard-sphere model are the size of symbols and thus not plotted.

time distribution (FPTD) in fact could be seen as  $\psi(s,t)$  with displacement equal to the distance between injection and absorbing sites. We associate  $\psi(t)$  to FPTD and directly calculate the key parameter  $\beta$  from pore-scale simulations for two models of porous media. It is worth noting that although the models of porous media examined here are idealized ones, but they in fact represent two opposite extremes of porous media classes. For example, overlapping spheres at low porosities can be used as models for consolidated media such as sandstone while on the other hand hard-sphere model could represent unconsolidated media such as packed beds and particulate composites [66]. Using pore-scale simulations on two different types of porous media, we obtained parameter  $\beta$  which could be seen as degree of medium heterogeneity [36]. Shown in Figure 2.7 is FPTD for two models of porous media and two porosities. As can be noted, transport of solute plume in the hard-sphere model is considered Fickian (i.e.,  $\beta > 2$ ). However, overlapping sphere medium shows anomalous behavior (i.e.,  $\beta < 2$ ) with parameter  $\beta = 1.8$ . This is interestingly close to the value reported by Bijeljic and Blunt [36] which was obtained for two-dimensional model of Berea sandstone, corroborating the fact that the model of overlapping spheres could be used to represents consolidated systems. Moreover, the shape of FPTD is consistent with trajectory length distributions where there exists a long tail of particle trajectories for the case of overlapping spheres while in the case of hard-sphere model, trajectory length distributions are essentially symmetric and do not exhibit long-tailed behavior.



Fig. 2.7.: First passage time distribution for (a) monodisperse hard-spheres and (b) overlapping spheres for high  $(\Box)$  and low  $(\bigcirc)$  porosity media.

Now, we study the Lagrangian velocity statistics and their evolution. Lagrangian description of particle velocity consists of two viewpoints which is analyzing Lagrangian velocities sampled isochronally and equidistantly along streamlines. Studies on flow through porous media and observations of intermittency of fluid velocities suggest that particle velocities persist for a characteristic length scale  $l_c$  rather than a characteristic time scale  $\tau_c$  [35]. Thus, stochastic description of particle transport in porous media is based on distribution of Lagrangian velocities sampled equidistantly along streamlines. Naturally, it is important to quantify the velocity statistics in these two frameworks and their relations to the Eulerian velocity field. Recently, Dentz *et al.* [34] provided expressions that relate Lagrangian velocity statistics to those of Eulerian. Here, we briefly review these relations and proceed to our analysis of Lagrangian statistics. We first consider particle velocity along its trajectory as a function of travel distance, s. According to Dentz *et al.* [34], the PDF of s(pace)-Lagrangian and t(ime)-Lagrangian velocities are related through flux-weighting as

$$p_s(v) = \frac{v p_t(v)}{\langle v_t \rangle} \tag{2.20}$$

where  $\langle v_t \rangle = \int v p_t(v) dv$ . Furthermore, due to volume conservation, t-Lagrangian velocity PDF,  $p_t(v)$ , is equal to Eulerian velocity PDF,  $p_e(v)$ . Thus, this means that  $p_s(v)$  is related to  $p_e(v)$  through flux weighting as

$$p_s(v) = \frac{v p_e(v)}{\langle v_e \rangle} \tag{2.21}$$

where  $\langle v_e \rangle = \int v p_e(v) dv$ . It is worth noting that Eq. (2.21) is valid under stationary condition. T- and s-Lagrangian velocity distributions evolve in time and space if their initial distribution is different than their steady-state one. Clearly, at t=0 and s=0both  $p_s(v)$  and  $p_t(v)$  are identical and equal to initial particle velocity distribution,  $p_0(v)$ . Furthermore, under Eulerian ergodicity and uniform injection which is the case here, t-Lagrangian velocity PDF coincides with Eulerian velocity PDF and thus does not evolve [34,89]. However, s-Lagrangian velocity distribution for the case of uniform injection evolves towards its steady-state PDF given in Eq. (2.21). Figure 2.8 shows the evolution of mean s-Lagrangian velocity. As expected, at s = 0 the mean s-Lagrangian velocity (and mean t-Lagrangian velocity) is equal to the mean Eulerian velocity. However, mean s-Lagrangian velocity evolves to its steady-state value while mean t-Lagrangian velocity remains constant and equal to mean Eulerian velocity. It is worth noting that mean s-Lagrangian velocity converges faster to its steadystate value for the case of monodisperse hard-sphere model compared to overlapping sphere model due to relatively simpler pore-structure. Finally, using Eq. (2.21), we can calculate the stationary s-Lagrangian velocity distribution and compare it with the distribution obtained from numerical simulation and particle tracking. Figure 2.9 shows that measured s-Lagrangian velocity PDF agrees well with the flux weighted Eulerian velocity PDF.

# 2.5 Eulerian statistics

A complete picture of velocity field is extremely useful in understanding transport in porous media. For example, Tennekes and Lumley [90] showed that mechanical



Fig. 2.8.: Evolution of the mean velocity sampled equidistantly in space for hardsphere ( $\bigcirc$ ) and overlapping sphere ( $\Box$ ) models. Due to uniform injection of particles, mean velocity sampled equidistantly in space evolves from its initial value which coincides with mean Eulerian velocity ( $\cdot - \cdot$ ) and reaches its steady-state value calculated from Eq. (2.21) (--). Inset: Mean velocity sampled isochronally remains constant until breakthrough and is equal to mean Eulerian velocity as expected.  $n_t$  is the number of time steps.



Fig. 2.9.: Comparison of stationary s-Lagrangian velocity PDF as given by Eq. (2.21) and numerical simulation.

dispersion  $(D_L)$  is directly related to the Lagrangian velocity of a particle  $(V_L)$  and Lagrangian integral time scale  $(T_L)$  as:

$$D_L \cong \overline{\left(V_L - \overline{V_L}\right)^2} T_L \tag{2.22}$$

Patil and Liburdy [43] replaced the Lagrangian velocity variance with Eulerian velocity variance under the assumption of  $V_L$  being stationary and estimated the mechanical component of longitudinal dispersion. Although it has been shown that different pore-space structures have significant effects on the velocity distribution at the porescale as well as changing the nature of transport, a quantitative study showing the interplay of pore structure, velocity, and transport is still lacking. One can imagine that due to incompressibility of fluid, local velocity changes as fluid go through network of throats and pores of different sizes. This simple argument suggests that given the pore and throat size distribution of a medium, the velocity distribution must obey the same distribution, a fact that was studied by Siena *et al.* [39] where they found that the pore size and velocity distributions decay similarly. However, such a relationship is not always very simple as spatial correlation of pore and throat sizes has also found to be an effective parameter [39].

Here, we first look at the velocity distribution for our three models of porous media with increasing level of pore space complexity over a range of porosities. Then, we focus on generalizing such velocity PDFs. Figure 2.10 shows that the velocity is broadly distributed and despite the randomness of pore space in different models of porous media, its distribution shows some universality. It is worth noting that in all the cases shown in Figure 2.10, there exist the following common feature as (i) the peak in PDFs locates at zero and it becomes more significant as porosity reduces, (ii) all the PDFs have both negative and positive tails, the latter is due to the tortuous flow path. This effect is the strongest in the case of overlapping spheres, which is supposed to result in the most complex pore space among the models examined here, (iii) in all the cases here, velocity distribution becomes less broad as porosity increases and the positive tail of velocity distribution decays faster, (iv) spanwise velocity components are symmetric with a peak value at zero, similar to the streamwise component of velocity. One can see by looking at Figure 2.10 that flow in overlapping spheres model results in broader range of velocity compared to hard-sphere models. Additionally, Figure 2.11 shows the velocity norm distributions of monodisperse



Fig. 2.10.: Top row: PDF of streamwise velocity. Bottom row: PDF of velocity perpendicular to mean flow direction (spanwise). All velocities are normalized by mean interstitial velocity  $(q/\phi)$  for (a,b) monodisperse hard-sphere, (c,d) polydisperse hard-sphere, and (e,f) overlapping sphere models of porous media.

hard-sphere and overlapping sphere models in log-log scale. It reveals a significant difference in velocity norm PDF of these two models in low velocity ranges. As shown in Figure 2.11, greater portion of pore-space in overlapping sphere model experiences very low velocities, which is consistent with the transport behavior shown in Figure 2.7. Quantitatively speaking, streamwise velocity variance scales as  $0.89\langle u \rangle^2$  compared to  $1.21\langle u \rangle^2$  for hard-sphere and overlapping sphere models, respectively. Since there is no long-range correlation of pore-space in our models of porous media (see Figure 2.3), we can explain this by studying pore-size distribution functions of theses models. Klatt and Torquato [91] numerically estimated complementary cumulative distribution functions (1-CDF) of pore sizes ( $F_{\delta}$ ) for monodisperse hard-sphere and overlapping spheres. Consistent with our velocity PDFs, they showed that  $F_{\delta}$  decays much slower for overlapping spheres compared to hard-spheres, resulting in a



Fig. 2.11.: PDFs of velocity norm for monodisperse hard-sphere ( $\bigcirc$ ) and overlapping sphere ( $\Box$ ) models for  $\phi = 0.36$ .

broader pore size distribution and also larger effective pore-diameter (see Figure 2.3). To generalize the velocity PDFs, we focus on the positive tail of distribution as the contribution of the negative tail to transport is negligible. The velocity distribution could be divided into two regions: first region  $(u < \langle u \rangle)$  is characterized by very small velocities due to existence of extensive fluid-solid interfaces and stagnant zones (see normalized velocities around zero in Figure 2.10) and second region  $(u > \langle u \rangle)$  which is responsible for most of transport and is characterized by the structure of sample-spanning network of pores and throats [92]. Here, we adopted a stretched exponential distribution as [42]:

$$f(u/\langle u \rangle) = a \exp\left[-\left(\frac{u/\langle u \rangle}{\alpha}\right)^{\eta}\right]$$
 (2.23)

where parameter  $\eta$  controls the shape of the positive tail (e.g. exponential or normal) and  $\alpha$  indicates the decay rate. Parameter *a* is a normalizing factor such that Eq. (2.23) is integrated to one and thus is given by  $\frac{\eta}{\alpha\Gamma(1/\eta,1/\alpha^{\eta})}$  where  $\Gamma$  is the incomplete gamma function. Figure 2.12 shows that  $\eta$  heavily depends on the porosity and exhibits a transition from subexponential ( $\eta < 1$ ) to normal ( $\eta \approx 2$ ).

Velocity distributions for models of porous media in this study showed broad dynamic ranges of velocity which can be estimated by Eq. (2.23) accurately for several



Fig. 2.12.: Dependence of model parameters in Eq. (2.23) on porosity for two models of porous media (hard-sphere and overlapping).  $\eta$  strongly depends on the porosity and is consistently larger for hard-spheres (a). On the other hand, the dependence of  $\alpha$  on the porosity is not very strong (b). This figure clearly quantifies the transition in velocity distribution shape as a function of porosity.



Fig. 2.13.: Streamwise velocity distributions for (monodisperse) hard-sphere and overlapping sphere models of porous media with their corresponding fits obtained by Eq. (2.23) (solid line). Porosity is 0.45.

decades in probability (Figure 2.13). Despite the usefulness of velocity distribution, they cannot be used to learn about spatial correlations of velocity field while such correlations are critically informative as the flow pass through tortuous paths and channels of high and low velocities. Thus, to examine the spatial features of velocity, we calculated two-point correlation function of 3-D velocity field which averages the scalar product of all velocity vector pairs separated by distance R by:

$$C_{uu}(R) = \left\langle \frac{\sum_{j} \left[ u'(r_{j}) \cdot u'(r_{j} + R) \right]}{\sum_{j} \| u'(r_{j}) \|^{2}} \right\rangle$$
(2.24)

where  $u' = u - \overline{u}$  and the summation is taken over 10<sup>4</sup> positions  $r_j$  randomly chosen in the simulation domain. Figure 2.14 shows  $C_{uu}$  and  $S_2$  obtained for monodisperse hard-spheres. In particular,  $C_{uu}$  is strongly similar to  $S_2(r)$ , shows a nearly exponential decay. We show that spatial features in velocity field are exactly the same as those of pore space. The oscillations in  $S_2(r)$  due to exclusion volume effects are reflected on pore-scale velocity fluctuations causing velocity fluctuations to be slightly correlated at large distances. To further generalize these findings, we carried



Fig. 2.14.: Spatial correlation in velocity fluctuations for monodisperse hard-sphere model along with their two-point correlation function calculated on pore space. Strong similarities suggests spatial correlations of velocity and pore space are almost identical in the creeping flow regime.

out the same analysis on more complex pore geometries such as those in polydisperse hard-spheres and overlapping spheres. Interestingly, results shown in Figure 2.15 suggest that even in completely different pore structures such as those in overlapping sphere model, spatial correlations of velocity are reflective of those of pore space. Observed strong relationship between pore-structure characteristics and spa-



Fig. 2.15.: Spatial correlation in velocity fluctuations for (a) polydisperse hard-sphere and (b) overlapping sphere models along with their two-point correlation function calculated on pore space.

tially fluctuating velocity field motivated us to see if such a relationship exists for natural rocks. We used cube samples  $(512 \times 512 \times 512)$  of unconsolidated sandpack and Castlegate sandstone with voxel size of  $9.184\mu$ m and  $5.6\mu$ m, respectively (data is publicly available at https://www.digitalrocksportal.org). These samples are originally obtained via x-ray micro-CT and then segmented where each voxel is identified as either void or solid. Porosity is 0.36 and 0.206 for unconsolidated sandpack and Castlegate sandstone, respectively. Figure 2.16, shows two-dimensional cross sections of the segmented image of these samples. To confirm that observed relationship between pore-space characteristics and fluctuating velocity field does not exist due to spatial homogeneity of the pore-space in hard-sphere and overlapping sphere models, similar analysis was performed on samples of unconsolidated sandpack and Castlegate sandstone. Figure 2.17 shows that consistent with what we observed previously for overlapping sphere and mono(poly)disperse hard-sphere models, a similar relationship exists in natural rocks as well where two-point correlation functions of pore-space and fluctuating velocity field decay similarly.



Fig. 2.16.: Cross sectional view of the indicator function (solid or pore space) and normalized velocity magnitude  $(|U|/|U|_{max})$  for (a,b) the unconsolidated sandpack and (c,d) Castlegate sandstone with porosity of 36% and 20.6%, respectively. Pore-space is colored black.

# 2.6 Conclusion

We investigated the flow through porous media through direct numerical simulation for three models of porous media with increasing levels of pore space complexity. We showed that the Eulerian velocity field has a broad dynamic range which reflects the complexity of the pore space. It was shown that streamwise Eulerian velocity distributions in virtually all porous media could be accurately modeled by a stretched exponential function which captures the transition from nearly exponential to Gaussian shapes of Eulerian velocity distribution for several decades in probability as porosity increases. A streamline particle tracking approach was used to calculate the Lagrangian statistics in an advection-dominant transport regime. We showed that



Fig. 2.17.: Spatial correlation in velocity fluctuations for different models of porous media along with their two-point correlation function calculated on pore space. Strong similarities suggest spatial correlations of velocity and pore space are almost identical in the creeping flow regime.

particle trajectory in overlapping sphere model compared to hard-sphere model was more tortuous and its distribution had higher skewness. We then showed by means of FPTD that higher tortuosity and broader Eulerian velocity distributions of nonoverlapping sphere model resulted in non-Fickian transport (with quantitative agreement with transport in sandstone) while transport in the hard-sphere model was found to be Fickian. Evolution of Lagrangian velocity distributions, sampled isochronally and equidistantly along the particle trajectory were studied for the uniform injection rule and it was found that mean s-Lagrangian velocity evolves in space and reaches stationary condition quickly (takes longer for overlapping sphere model compared to hard-sphere model due to more complex pore-structure) while mean t-Lagrangian velocity does not evolve in time. Under stationary condition, s-Lagrangian velocity distribution is related to Eulerian velocity distribution through flux-weighting. Finally, we studied the spatial correlations in 3-D velocity field by means of two-point correlation functions of velocity fluctuations. For the relatively homogeneous porous media examined here (overlapping and non-overlapping spheres in 3-D), we showed that the two-point correlation function of 3-D velocity field decays similarly to that of pore space and even at long distances displays similar oscillations. We tested the generality of this finding on two more heterogeneous porous media (i.e., unconsolidated sandpack and Castlegate sandstone) in which we again observed a similar behavior, where two-point correlation functions of 3-D pore space and velocity fields were extremely similar.

# 2.7 Acknowledgements

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# 3. NANOPARTICLE DISPERSION IN POROUS MEDIA IN VISCOELASTIC POLYMER SOLUTIONS

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#### 3.1 Abstract

Flow of viscoelastic fluids in porous media is ubiquitous in biological and industrial processes, and the choice of viscoelastic fluids is of foremost importance for their effect on macroscopic flow and transport properties. In this paper we study the macroscopic properties of flow and transport of viscoelastic fluids through a model porous media by means of Direct Numerical Simulation (DNS). We show that flow of a viscoelastic fluid modeled via a FENE-P constitutive model features three distinct regimes of flow resistance: a plateau at low Deborah numbers, and a shear-thinning phase followed by an abrupt flow thickening above a critical Deborah number, consistent with experimental observations. Congruous to this shear-thickening, we observe the onset of hydrodynamic instabilities resulting in fluctuations in pressure drop and flow resistance. These fluctuations intensify with increasing the fluid's elasticity. Finally, we investigate Lagrangian attributes of viscoelastic flow and transport through porous media. We find that although the fluid's elasticity broadens the Lagrangian velocity distributions, it does not alter the long-term particle dispersion in disordered porous media.

### 3.2 Introduction

In a wide range of processes, from drug delivery [93, 94] to enhanced oil recovery [95, 96], sub-micron particle transport is central to the efficacy of the process. In many practical cases of flow through porous media, flow is dominated by viscous forces because of very small length-scales and is described by Darcy's law [97, 98]. In the case of Newtonian fluids for creeping flow and advection-dominant transport, pore-geometry profoundly impacts the particle transport through mechanical mixing of fluid stream and thus governs the long-term dispersion of particles [99–101]. In non-Newtonian fluids, fluid rheology could further complicate the transport of particles and other macroscopic properties of flow. In cases where fluid is viscoelastic, there is ample experimental evidence of onset of hydrodynamic instabilities due to elastic nature of flow [102]. Specifically, such instabilities have been experimentally observed in contractions [103], Couette flow between two cylinders [104, 105], curved channels [102], and periodic array of cylinders [106, 107] (see reviews by Larson [108], McKinley et al. [109] and Shaqfeh [110]). These elastic instabilities, in the absence of inertial effects, are so-called elastic turbulence and occur due to extra polymer stress and curved streamlines, even in the absence of inertia. A simple criterion developed by Pakdel and McKinley [111] and later modified by Morozov and van Saarloos [112] characterizes the critical conditions for the onset of elastic instabilities. Furthermore, experimental observations have shown that onset of such instabilities generate secondary flows and at the macroscopic level increase flow resistance [104, 113, 114]. This upturn in flow resistance has been investigated via experimental [115, 116] and numerical [117–119] approaches. Some studies have shown that this upturn in flow resistance coincides with the onset of hydrodynamic instabilities [120–123].

The presence of elastic instabilities can thus have important effects on mixing [124,125], solute dispersion in porous media [126], or displacing trapped secondary immobile phase at the pore-scale [122]. For example, as discussed by Clarke *et al.* [122], increased oil mobilization was attributed to elastic turbulence during polymer flood-

ing in a microchannel. Despite the experimental observation of an increased flow resistance of viscoelastic fluids in porous media, numerical simulations of such problems are scarce and thus it is not clear whether elastic instabilities contribute to the increased flow resistance. Even for simple geometries such as flow over periodic arrays of cylinders, numerical observations are contradictory [120, 124]. Moreover, it is not clear whether and through which mechanism viscoelasticity enhances mixing in flow through porous media. While experimental observations by Scholz et al. [126] show that elastic turbulence significantly enhances transverse dispersion, Babayekhorasani et al. [127] suggest that fluid rheology will not ultimately alter the dispersion of particles. To address this gap in literature, we focus on numerical simulation of flow of viscoelastic fluids through porous media at the pore-scale, to study its characteristics and transport of nanoparticles. We illustrate the effects of fluid viscoelasticity via comparisons with equivalent Newtonian flows. In this study, we also look at the Lagrangian velocity distributions and how the particle dispersion in a random array of cylinders is changed due to fluid elasticity. We study the flow resistance over a range of fluid elasticity. Finally, we show that when elastic effects are strong enough, consistent with experimental observations, flow does not reach a steady-state.

#### 3.3 Governing equations

A finite volume method and body-fitted grid is used to model the flow of viscoelastic fluid through porous media within the open-source framework OpenFOAM [128]. Transient flow of incompressible single-phase isothermal fluids is governed by the conservation of mass and momentum as:

$$\nabla \cdot \mathbf{u} = 0, \tag{3.1}$$

$$\rho\left(\frac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \cdot \nabla \mathbf{u}\right) = -\nabla p + \nabla \cdot \boldsymbol{\tau} + \mathbf{g},\tag{3.2}$$

with velocity vector  $\mathbf{u}$ , density  $\rho$ , pressure p, external body forces  $\mathbf{g}$ , and extra stress tensor  $\boldsymbol{\tau}$ . The extra stress tensor comprises solvent contribution  $\boldsymbol{\tau}_s$  and polymer contribution  $\boldsymbol{\tau}_p$  so that  $\boldsymbol{\tau} = \boldsymbol{\tau}_s + \boldsymbol{\tau}_p$ . In Eq. 3.2, the solvent stress tensor,  $\boldsymbol{\tau}_s$ , is given by  $\eta_s \left( \nabla \mathbf{u} + \nabla \mathbf{u}^T \right)$  where  $\eta_s$  is the solvent viscosity and constant. The polymer stress in this work is modeled via the finite extensible nonlinear elasticity (FENE) model of polymeric fluids. We adopted the shear-thinning FENE-P constitutive equations [129, 130]. The FENE-P constitutive equation reads:

$$\boldsymbol{\tau_p} + \frac{\lambda}{f} \boldsymbol{\tau_p}^{\nabla} = \frac{a\eta_p}{f} \left( \mathbf{u} + \nabla \mathbf{u}^T \right) - \frac{D}{Dt} \left( \frac{1}{f} \right) \left[ \lambda \boldsymbol{\tau}_p + \eta_p \mathbf{I} \right], \qquad (3.3)$$

where  $\eta_p$  is the polymer viscosity at zero shear-rate,  $\lambda$  is the relaxation time of the polymer, **I** is the identity tensor,  $\frac{D}{Dt}$  is the material derivative, and operator  $\nabla$  is the upper-convected time derivative given by:

$$\nabla_{\boldsymbol{\tau}_p} = \frac{D\boldsymbol{\tau}_p}{Dt} - \boldsymbol{\tau}_p \cdot \nabla \mathbf{u} + \nabla \mathbf{u}^T \cdot \boldsymbol{\tau}_p, \qquad (3.4)$$

Finally, the function f is given by:

$$f(\boldsymbol{\tau}) = \frac{L^2 + \frac{\lambda}{a\eta_p} tr(\boldsymbol{\tau})}{L^2 - 3}, \text{ with } a = \frac{L^2}{L^2 - 3},$$
(3.5)

In Eq. 3.5, parameter L characterizes the maximum polymer extensibility. It is worth noting that when  $L \to \infty$ , the Oldroyd-B constitutive equation is recovered. We use L = 1000 for all our simulations. The governing equations are discretized using the finite volume method. Most notably, the log-conformation approach is used for calculating the viscoelastic stress tensor for increased accuracy and robustness. The polymer stress tensor is related to the conformation tensor and for FENE-P constitutive model, natural logarithm of the conformation tensor ( $\Theta$ ) is related to the stress tensor by:

$$\boldsymbol{\tau_p} = \frac{\eta_p}{\lambda} (f e^{\boldsymbol{\Theta}} - a \mathbf{I}) \tag{3.6}$$

Further details of the numerical recipes for pressure-velocity and stress-velocity coupling and overall solution procedure could be found in Pimenta and Alves [131] (RheoTool v2.0). Here, to quantify the elastic nature of the flow, we define the Deborah number (De) as:

$$De = \lambda / t_{flow} \tag{3.7}$$

where  $\lambda$  and  $t_{flow}$  are polymer relaxation time and characteristic flow residence time. We use  $t_{flow} \approx \sqrt{k}/\langle u_x \rangle$  where  $\langle u_x \rangle$  and k are average interstitial velocity in the mean flow direction, and the hydraulic permeability (measured with Newtonian fluid), respectively. The Reynolds number (*Re*) is defined as  $Re = \frac{\rho \langle u_x \rangle \sqrt{k}}{\eta}$  where  $\eta = \eta_s + \eta_p$  is the solution viscosity. We use  $\sqrt{k}$  as an effective length scale as it has been found useful in characterizing other nonlinear effects in flow in porous media [132, 133].

In this work, we are interested in the effects of fluid's rheology on macroscopic flow and transport properties in porous media. We study the transport properties in a Lagrangian framework by tracking a large number of tracer particles. Particle tracking is advantageous over solving the advection-diffusion equation as it does not suffer from numerical dispersion/diffusion. Time evolution of tracer particles is obtained through numerical integration of the equation:

$$\mathbf{r}(t + \Delta t) = \mathbf{r}(t) + \mathbf{v}(\mathbf{r}(t))\Delta t + \zeta \sqrt{2D_m \Delta t}, \qquad (3.8)$$

where  $\mathbf{r}(t)$  and  $\mathbf{v}(\mathbf{r}(t))$  are particle location and particle velocity (i.e., fluid flow velocity at the particle location), respectively. In Eq. 3.8,  $\Delta t$  is the time step,  $D_m$ is the molecular diffusion, and  $\zeta$  is a vector with random components drawn from a standard normal distribution. Particles may hit the solid interface due to Brownian motion. We impose a reflective boundary condition on the fluid-solid interface. The relative importance of advection and diffusion in Eq. 3.8 is characterized by Peclet number (*Pe*) defined as  $Pe = \frac{UL}{D_m}$  where *U*, *L* are characteristic velocity ( $\langle u_x \rangle$ ) and length scale ( $\sqrt{k}$ ).

#### 3.4 Problem description

We study the unsteady and steady-state flow of viscoelastic fluids and transport through a random array of monodispersed cylinders. Our computational domain is  $20D \times 20D$  where D is the cylinder diameter with porosity of 0.4 as shown in Figure 5.5. The random array of cylinders is generated by a collision-driven molecular dynamics algorithm used in our previous work [99] and is periodic in both directions. Due to the randomness of the pore-space and periodicity of our computational domain, it is worth noting that the domain should be large enough to be representative of larger samples and prevent the flow from interacting with its periodic image [134]. This condition is satisfied for all simulations presented here. Flow is driven by a pressure gradient in the x-direction so that the average interstitial velocity  $(\langle u_x \rangle)$ of unity is achieved. Although the mean interstitial velocity of unity is achieved in a short time for a range of *De* examined here, as we will show later in this paper, pressure gradient could not reach a steady-state for high De. For such unsteady simulations, we run our simulations for  $20\lambda$  to obtain meaningful statistics and report time-averaged quantities. In this work, we examine flows with a wide range of Defrom 0 (i.e., Newtonian) to 48 by varying the polymer relaxation time. In all the simulations, the Reynolds number is kept small  $(Re \ll 1)$  which allows us to only study the interaction of fluid rheology and viscoelastic effects with random pore space geometry.

To obtain Lagrangian statistics and transport properties via particle tracking, we place  $2 \times 10^5$  tracer particles randomly in the pore space. We have verified that this number of particles gives converged statistics. Since our computational domain is periodic in both directions, particles that exit from each boundary will naturally reenter the computation domain. This will help us study the particle transport over a long time. Finally, in all simulations we use  $Pe = 10^3$  and thus focus on the mechanically-mixed regime where advection is dominant.



Fig. 3.1.: Computational domain: random array of cylinders with solid volume fraction of 0.6 (i.e., porosity of 0.4). The zoomed-in view illustrates refined and layered-mesh around solid surfaces to accurately resolve the flow.

#### 3.5 Results

#### 3.5.1 Flow type and normal stresses

We seek to answer whether the fluid rheology significantly alters the flow topology. We quantify the flow topology with the parameter  $\xi$  defined as:

$$\xi = \frac{|\mathbf{D}| - |\Omega|}{|\mathbf{D}| + |\Omega|},\tag{3.9}$$

where  $|\mathbf{D}|$  and  $|\mathbf{\Omega}|$  are magnitudes of rate-of-deformation and vorticity tensors, respectively:

$$\mathbf{D} = \frac{1}{2} \begin{bmatrix} \nabla \mathbf{u} + (\nabla \mathbf{u})^{\mathrm{T}} \end{bmatrix} \quad \mathbf{\Omega} = \frac{1}{2} \begin{bmatrix} \nabla \mathbf{u} - (\nabla \mathbf{u})^{\mathrm{T}} \end{bmatrix}, \quad (3.10)$$

Flow type parameter,  $\xi$ , thus varies from -1 (pure solid-body rotation) to +1 (pure extensional flow) and pure shear-flow yields  $\xi = 0$ . Shown in Figure 3.2 is flow type for flow of Newtonian and viscoelastic fluids in porous media. It is clear that the fluid is undergoing mostly shear and elongation for both cases and the fluid elasticity does not result in a significant change in the flow structure and type. Flow topology is largely affected by pore-structure and inherent randomness in pore-space distribution rather than fluid rheology. Figure 3.3 signifies the importance of fluid rheology on the distribution of flow topology and strain rate throughout the porous media. Although the changes between Newtonian and viscoelastic fluids are not significant, fluid elasticity slightly enhances regions of elongational flow and shifts the strain rate towards higher values. We expect such effects to increase with De, however, random pore-space is the dominant factor determining the flow topology and strain rate of fluid in random porous media. Flow in porous media features preferential pathways [99, 135]. Figure 3.4 shows normalized velocity magnitude for Newtonian and viscoelastic flows at the pore-scale. While in both cases the flow structure and preferential pathways remain similar, subtle differences are observed due to presence of normal stress. As mentioned above, extra-stress due to polymer makes subtle changes to velocity profile around solid surfaces. Figure 3.5 reveals



Fig. 3.2.: Contour plot of flow type  $(\xi)$  for Newtonian fluid (left) and viscoelastic fluid (right). Flow with De = 12 results in qualitatively similar flow topology throughout the porous media at the pore-scale. The flow is shear-dominated ( $\xi = 0$ ) in both cases.



Fig. 3.3.: Probability density function (PDF) of flow type parameter ( $\xi$ ) and strain rate for a viscoelastic fluid compared to a Newtonian fluid.

the first normal stress profile at the pore-scale. The contour plot of log-conformation tensor reveals how polymer compresses and stretches around solid obstacles, resulting in filament-like regions of high first normal stress which interact with the underlying velocity field, changing the local velocity profile from that of a Newtonian fluid where normal stresses do not exist. Although not shown here, magnitude of (normalized) normal stresses increase with the increase of De.



Fig. 3.4.: Normalized velocity magnitude  $(|\mathbf{u}| / \langle u_x \rangle)$  for the Newtonian and viscoelastic case with De = 12.



Fig. 3.5.:  $\Theta_{xx}$  where  $\Theta$  is the log-conformation tensor (left) and normalized normal stress ( $\tau_{xx}/\eta U/D$ ) for De = 12 (right).

# 3.5.2 Shear thinning and thickening

Experiments of flow of viscoelastic fluids in porous media have reported Newtonian behavior at low-shear rates, followed by a shear-thinning and then onset of flow thickening after a critical De [123]. This, interestingly, is in contrast to data obtained from purely shear flows (e.g., in a rheometer) where shear-thickening behavior is not observed, while in fact, such flow thickening has important implications in many

biological and industrial applications such as enhanced oil recovery [136]. Up to now, most of studies of viscoelastic flows have focused on simple flow configurations such as those in parallel-plate [113,137], Taylor-Couette [138], serpentine [102] geometries. In this section, we look at the onset of elastic instabilities and evolution of flow resistance in flow of viscoelastic fluids through porous media. We characterize the elastic instabilities in terms of fluctuations in pressure gradient [114], dp/dx, required to drive the flow at the mean interstitial velocity of unity. To study the flow resistance, we define a resistance factor parameter as:

Resistance factor = 
$$\frac{\left(\frac{dp/dx}{\langle u_x \rangle}\right)_V}{\left(\frac{dp/dx}{\langle u_x \rangle}\right)_N}$$
, (3.11)

where V and N denote viscoelastic and Newtonian, respectively. Shown in Figure 3.6 is the variation of resistance factor as a function of *De*. Consistent with experimental observation, the resistance factor features three distinct regions: a plateau at small De, shear thinning at intermediate De, and flow thickening and increased resistance above a critical De. In flow through porous media, polymer molecules experience shear near walls and constantly undergo elongation and contraction due to the converging-diverging nature of the flow. When flow time scale is small and polymer relaxation time is large (i.e., large De), flow thickening appears as polymer molecules could not adjust to the flow rapidly. Furthermore, we analyzed the contributions from  $\frac{d\tau_{xx}}{dx}$  and  $\frac{d\tau_{xy}}{dy}$  to the pressure gradient,  $\frac{dp}{dx}$ . We found out that the contribution from the first term to pressure gradient follows the same behavior as the pressure gradient itself. Meaning that initially, it decreases with De until around the critical De. Above the critical De contribution from  $\frac{d\tau_{xx}}{dx}$  to the pressure gradient increases with De and becomes the dominant factor at large De. An important observation here is that for flows with De above a critical value and due to elastic instabilities, steady-state cannot be achieved even when  $Re \ll 1$ . Although such instabilities are quite small around the critical De, they grow quickly as De increases and cause strong fluctuations in pore-scale velocity and pressure drop across the porous medium. Fig 3.6 also features error bars showing variation in the resistance factor because of variations in pressure drop across the porous medium (see Eq. 3.11). Figure 3.7 shows the time history of pressure gradient fluctuation for three flows with increasing levels of fluid elasticity. For the case with De = 6 and where elastic effects are relatively small, the pressure gradient fluctuates with a relatively small amplitude and a distinct period. As we make fluid elasticity stronger, pressure drop fluctuations show enhanced magnitude and become more irregular and the time period no longer simply corresponds to the polymer relaxation time.



Fig. 3.6.: Three regions of resistance factor parameter: initial plateau, shear thinning followed by a shear thickening period. The onset of shear thickening behavior is congruous with elastic instabilities and thus the variability in the resistance factor for high De. The magnitude of this variability grows rapidly with De. Note that fluctuations of resistance factor (dp/dx) is absent before the onset of flow thickening. Although we observe small but growing fluctuations with De, small fluctuations could not be seen on this plot as they are smaller than the marker size used here.



Fig. 3.7.: Pressure gradient fluctuations as a result of onset of elastic stress instabilities for high values of *De*. Magnitude of pressure fluctuations grows substantially as *De* increases.

### 3.5.3 Lagrangian statistics

In this section, we investigate whether fluid elasticity changes particle dispersion in porous media. To answer this question, as described earlier, we place  $10^5$  particles uniformly in the pore-space and record their position and velocity isochronally (as opposed to equidistantly) along the particle trajectory. Particle trajectories are shown in Figure 3.8.

Next, we study the particle velocity distributions for the case of Newtonian and viscoelastic fluids. Particle velocity distributions are important as they reveal the velocity distribution from which particles are sampling and thus determining the degree of media heterogeneity and particle dispersion. Figure 3.9 shows the effects of fluid elasticity on the particle velocity distribution.

Evidently, flow of viscoelastic fluids creates a broader underlying velocity field, consistent with experimental findings of [127]. Broader, more heterogeneous velocity



Fig. 3.8.: Sample of particle trajectories. Each (colored) line represents one particle. Only few trajectories are shown for better visualization.



Fig. 3.9.: Probability density function of longitudinal  $(u_x/\langle u_x \rangle)$  and transverse  $(u_y/\langle u_x \rangle)$  velocities for Newtonian and viscoelastic (De = 12) fluids. In both directions, fluid elasticity broadens the velocity distribution. This implies that particles undergo stronger dispersion as they sample from a broader velocity distribution in the case of viscoelastic fluid compared to that of a Newtonian fluid.

distributions imply a more dispersive regime. However, it is important to understand if polymer solutions change the long-time dispersion coefficient. To answer this question, we next look at the mean-squared displacement (MSD) of particles defined as  $\sigma_x^2 = \langle (x(t) - \langle x(t) \rangle)^2 \rangle$  where  $\langle \cdot \rangle$  denotes averaging over all particles. Plotted in Figure 3.10 is the MSD for tracer particles in Newtonian and viscoelastic fluids. In short times, MSD in both directions and for both fluids show ballistic scaling (MSD  $\sim t^2$ ) as reported elsewhere [126, 139]. This is followed by a period of non-Fickian scaling before a linear scaling (i.e., Fickian regime) regime is achieved. It is worth noting that while MSD of tracer particles in the longitudinal direction in both fluids scales similarly in early, intermediate, and long times, MSD in the transverse direction grows faster in intermediate times in viscoelastic fluids compared to that of a Newtonian fluid. Of course after reaching the Fickian regime MSD scales similarly, regardless of fluid rheology. This is important as it shows that in long times, in random porous media, dispersion of tracer particles occurs regardless of fluid rheology. In fact, randomness of pore-space, in long times, averages out any effects of fluids elasticity on the underlying velocity field and thus resulting in a similar scaling of MSD and thus dispersion coefficient  $(D(t) = \frac{1}{2} \frac{d\sigma^2}{dt})$ . These results are consistent with experimental observations of Babayekhorasani *et al.* [127], where they showed that fluid elasticity only alters the dispersion coefficient in intermediate times and nanoparticles have similar long-time dispersion behavior in both Newtonian and viscoelastic fluids in disordered media (packed bed of monosized beads). Our finding that fluid rheology



Fig. 3.10.: Mean-squared displacement (MSD) of tracer particles in longitudinal (L) and transverse (T) directions. After an early-time ballistic scaling followed by a pre-asymptotic regime, MSD for both Newtonian and viscoelastic cases reaches its asymptotic Fickian regime and scales linearly with time.

(viscoelastic effects) cannot be used to modulate the particle dispersion in discorded media in long times is in contrary to findings of Scholz *et al.* [126] in periodic and ordered porous media. They showed experimentally that velocity fluctuations arising from high *De* flows in ordered porous media could significantly enhance the transverse dispersion of particles. However, experimental observation of Babayekhorasani *et al.* [127] and our numerical results of the viscoelastic flows through disordered media show that randomness of pore-space is the dominant factor determining the long-term dispersion of particles in disordered media and mechanically-mixed regime ( $Pe \ge 72$ ).

#### 3.6 Conclusions

In this study, we investigated the macroscopic properties of flow of viscoelastic fluids represented via a FENE-P model and compared it to those of Newtonian fluids. We showed that in both cases, the flow type parameter remains the same and flow is shear-dominated. In addition, we showed that in the flow of viscoelastic fluids filament-like regions of high normal stresses develop in the wake of cylinders (stagnation points) and become stronger with increasing De. By showing a contour plot of first component of conformation tensor we illustrated how polymer molecules are compressed or stretched at the pore-scale. We then studied the flow resistance of viscoelastic fluids in a model porous media and showed that flow goes through phases of shear-thinning followed by intense shear-thickening, above a critical De. Additionally, we demonstrated the onset of fluctuations in the flow by studying temporal evolution of pressure gradient. Finally, we investigated the particle dispersion in disordered media and the role of fluid rheology (viscoelasticity) by means of particle tracking. We found that particle dispersion in the transverse direction is slightly enhanced due to broadening of velocity distribution and changes in the underlying velocity field in intermediate times. However, consistent with experimental observations of the same problem, we observed that fluid elasticity does not alter the particle dispersion in disordered porous media in long times where we observed Fickian scaling of MSD in both cases of Newtonian and viscoelastic flows. This result has great implications when choosing viscoelastic polymers in various processes such as enhanced oil recovery where particle/solute dispersion is crucial.

# 3.7 Acknowledgments

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# 4. MULTI-OBJECTIVE HISTORY MATCHING OF SURFACTANT-POLYMER FLOODING

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#### 4.1 Abstract

Surfactant-polymer flooding is an effective process in extracting most of the original oil in place remained after conventional water flooding process. However, this technique is complicated and involves extensive screening and numerous experiments to find the optimum chemical composition, salinity, etc. Surfactant-polymer flood modeling can facilitate the optimization of the process, however the inherently large parameter space results in great uncertainty and poor predictive capability. Here, by means of a novel approach using global sensitivity analysis, we reduce the parameter space of a typical surfactant-polymer flood model to facilitate model calibration and history matching process.

To inform our analysis, we performed three Berea coreflood experiments with different slug designs resulting in different salinity profiles. The results from our coreflood experiments revealed and quantified the high sensitivity to salinity, underlying the importance of accurate phase behavior modeling.

In addition to coreflood experimental data, we used an extensive set of laboratory data including polymer rheology, surfactant phase behavior, polymer permeability reduction, and capillary desaturation along with results from sensitivity analysis to build a mechanistic surfactant-polymer flood model. After modeling of sub-processes such as polymer flood model or phase behavior of our surfactant/oil/water system, through a multi-stage calibration algorithm, coreflood experimental data was used to build a thorough surfactant-polymer flood model where cumulative oil production and pressure profile were history matched simultaneously. Finally, we showed that our surfactant-polymer flood model has predictive capabilities with no need for ad-hoc tuning of the model parameters by modeling two additional coreflood experiments where cumulative oil production and pressure profile matched those of experiments.

#### 4.2 Introduction

Enhanced oil recovery is of great importance as two-third of the original oil in place (OOIP) remains intact after waterflooding of many mature reservoirs [140,141]. Waterflooding becomes ineffective as oil is dispersed and trapped in small pores by strong capillary forces. Surfactant-polymer (SP) flooding is a tertiary oil recovery technique targeting the oil trapped in small pores through reducing the interfacial tension between water and oil, improving mobility control as a result of polymer injection, and avoiding early breakthrough. Despite the elegant mechanism of oil recovery in SP flooding and its high efficacy in controlled laboratory experiments, it has showed poor performances in field-scale experiments [142, 143] due to significant uncertainties [144].

Numerical simulations of subsurface flows are subjected to various sources of epistemic uncertainty due to lack of data. This issue is more severe in the case of chemical enhanced oil recovery as such processes are very complicated. A typical SP coreflood model requires about 170 input parameters [145] and extensive screening processes and various experiments. A successful SP process necessitates an optimal design of parameters such as slug sizes, chemical concentration in each slug as well as taking into account uncertain variables such as residual oil saturation to chemical flooding, chemical adsorption rates, etc. Consequently, the design of a successful SP flood is highly dependent on uncertain parameters.

There exists a large body of literature on SP flooding models [141, 146–148]. History matching with coreflood experiments is the first step in developing these models [149], where some model parameters such as relative permeability curves or capillary desaturation curves (CDC) are tuned until a satisfactory match between experiment and simulation results is achieved. AlSofi *et al.* [147] used data from several SP coreflood experiments on carbonates and history matched cumulative oil recovery. In their work, they tuned parameters such as dispersivity, critical micelles concentration, interfacial tension, and capillary desaturation to predict the incremental oil recoveries correctly. Hosseini-Nasab et al. [150] studied the performance of Alkalinesurfactant-polymer (ASP) flooding at sub-optimum conditions in Bentheimer cores and history matched their model for pressure drop profile, cumulative oil recovery, and effluent profile. However, most previous works did not provide a quantitative analysis of performance of their history matched model or did not further validate their models by predicting new coreflood experiments.

After successful history matching, one can use the SP flood model for various purposes such as production forecast or optimization. In literature, much effort has been devoted to sensitivity analysis of design parameters (i.e., slug sizes and concentrations, reservoir characteristics such as porosity and permeability, fluids properties such as viscosity, etc.) [151–156]. One of the most comprehensive studies has been performed in [157] where optimum phase type, effects of salinity profile, oil viscosity, salinity window, and solubilization ratios among other parameters were separately studied on the overall oil recovery. To quantify uncertainty in SP flooding models, a probabilistic collocation method was used to propagate uncertainty in polymer viscosity multiplier, chemical adsorption rates and  $S_{orc}$  [158]. In the work of Hou *et al.* [159], a quasi-Monte Carlo sampling approach was adopted for efficient sampling of uncertain variables and then the effects of medium heterogeneity on CO<sub>2</sub> migration was studied. Dourache *et al.* [160] carried out sensitivity analysis of SP flooding at

the reservoir scale using a response surface methodology (RSM) and Gaussian regression to approximate the reservoir output as a function of time. To avoid large computational costs, Mollaei et al. [156] used Winding Stairs (WS) as a sensitivity analysis method in conjunction with an analytical chemical flood predictive model (CFPM). Although quite important, sensitivity analysis of design parameters facilitates optimization of SP flooding and not history matching. Thus, it is important to perform a separate sensitivity analysis on model parameters to facilitate the history matching process. Sensitivity of SP flood experiments to model parameters in SP floods, however, are well-known to be the main difficulty in scaling up a SP coreflood experiment to field scale [142, 161]. Thus, sensitivity of SP floods to model parameters and intrinsic uncertainty associated with them are critically important and must be quantified. Despite the numerous studies on modeling SP flooding, only a few studies have examined sensitivity of important quantities of interest (i.e. cumulative oil production and maximum pressure drop) to key parameters in SP flood model. In the early work of Brown et al. [142], a simple SP flooding model based on a fractional flow theory was employed and the effects of adsorption, relative permeability curves at high capillary numbers, residual water saturation  $S_{orw}$  and residual oil saturation  $S_{orc}$  were examined. Similar analysis is presented in studies such as [160, 162, 163] using rather simple models for SP flooding. In those studies oil/water/surfactant phase behavior were disregarded and thus formation of the third phase (i.e. middle phase microemulsion) or solubilization of oil/water in the surfactant-rich phase cannot be captured. Consequently, [160, 162] did not focus on successful history matching and further validating it. Inability of those simple models is further clear by relatively poor history matching presented in [163]. In an experimental study, Walker et al. [164] showed that microemulsion viscosity alone has major effects on the pressure gradient and overall recovery efficiency. Recently, AlSofi et al. [147] studied SP flooding in carbonates by means of a 1D coreflood model. They later quantified the sensitivity of their 1D model to some detailed model parameters such as those used in polymer viscosity calculation and surfactant phase behavior. Finally, although in mechanistic modeling studies of SP flooding such as [150, 165] good matches between experimental and modeling data have been observed, no systematic history matching algorithm were presented. Furthermore, predictive capability of such history matched models were not further tested.

One can easily notice that in most previous SP flooding studies, (i) there exists an arbitrariness in history matching methodology and thus a systematic approach to history matching of SP flooding is needed, (ii) history matched SP models have not been further tested to assess their predictive capabilities and rather blindly applied at larger scales, (iii) history matching has been performed by matching the cumulative oil production only and other important quantities such as oil-cut, pressure and effluent profiles were ignored, (iv) sensitivity analysis and uncertainty quantification are carried out using simplistic SP models which are far from real processes occurring during a SP flood experiment.

In this work, we aim at addressing two issues we mentioned above namely lack of robust history matching with multiple objectives and determining sensitivity of typical SP flood models to some key physical parameters using a comprehensive SP flood model rather than a simplified one. To do so, we build a mechanistic SP flood model where most of the model parameters are determined prior to coreflood simulation using laboratory measured data and only few parameters are left for the history matching process. Model calibration is greatly assisted by means of global sensitivity analysis to quantify the response of the model to uncertain input parameters taking into account all major physical processes occurring during a SP flood experiment. First, each sub-model in a SP flood model is examined to identify the most important parameters. Next, a sensitivity analysis is done on the entire SP flood model determining the most important processes. Then, we history match the cumulative oil production and pressure profile using a multi-stage calibration algorithm. Finally, we validate the model by predicting new independent experimental results without further tuning the model parameters and we quantify the accuracy of the numerical results.

In the next section, we describe the coreflood experiments, which we used for history matching and calibrating our models. In §4.3, we discuss the modeling approach and how we divide a typical SP flooding process into smaller sub-processes, where model calibration can be done robustly. In §4.4, we perform a thorough sensitivity analysis of SP flood model to further reduce the parameter space and facilitate the history matching process. Finally, we present the calibrated SP flood model and its validation in §4.4.5 obtained via a multi-stage algorithm.

#### 4.3 Coreflood experiments

In this section, we briefly discuss the coreflood and other experiments used in this study for model calibration and validation. Berea sandstone samples (Length: 12", diameter: 2") of similar permeability ( $\approx 400 \text{ mD}$ ) and porosity (0.2) to the reservoir of interest were used for all the flooding experiments under reservoir temperature (24 °C) and pressure (400 psi). Throughout the experiments, a synthetic field brine with total dissolved solid (TDS) of 9400 ppm and reservoir oil (dead oil) were used. A combination of PETROSTEP<sup>®</sup> S-13D HA (Alcohol Alkoxy Sulfate) and A6 (Alkyl Benzene Sulfonate) and Huntsman SURFONIC<sup>®</sup> L series co-solvent was used at total chemical concentration of 8000 ppm for the SP slug. Partially hydrolyzed polyacrylamide polymer (SNF Flopam 3330) was used for mobility control. An injection rate of 1 ft/D was used for all the oil recovery experiments. A summary of fluid properties is shown in Table 4.1. Experiments were performed in secondary and tertiary

Table 4.1.: Fluid properties used in experiments

Property	oil	brine
Viscosity (cP)	13.8	1.0
Interfacial tension (dyn/cm)	22	
pH		8.0
Anions $(meq/ml)$		0.145
Divalent cations (meq/ml)		0.0097

modes of recovery following the same injection sequence: initial waterflooding (IWF)
at reservoir salinity, high total dissolved salt (HTDS) preflush, surfactant-polymer flooding (SP), polymer flooding (P), and finally extended waterflooding (EWF). Experimental data and models used to describe the experiments are provided in the next section.

#### 4.4 Coreflood modeling

Berea coreflood experiments (BCF) are modeled using UTCHEM [166,167]. UTCHEM is a multicomponenet multiphase simulator with robust phase behavior modeling and is considered the most comprehensive tool for SP flood modeling [157,168]. Generally, chemical flooding requires a large set of model parameters relating to polymer behavior, surfactant phase behavior and interfacial tension, and multiphase displacement models. A common problem with such a high-dimensional problem is the existence of a non-unique solution, meaning that many different sets of model parameters yield the same results [169]. Our approach here is to avoid this condition through finding and calibrating most of the model parameters separately via additional experiments. Figure 4.1 shows a summary of different processes during a typical SP coreflood and highlights the experimental data used for model calibration.



Fig. 4.1.: A summary of different processes in SP flooding and corresponding model calibration approach. Colored boxes are those left for the history matching process due to lack of experimental data (i.g. microemulsion viscosity) or large uncertainty involved in measuring them such as surfactant/oil/water phase behavior.

# 4.4.1 Modeling polymer flooding

#### Polymer viscosity

In order to quantify and model the polymer behavior, a series of rheology experiments were performed on HPAM 3330. The goal is to model polymer behavior as a function of salinity, shear rate, and its concentration [170]. Eq. 4.1 accounts for the polymer concentration and the salinity effects

$$\mu_p^0 = \mu_w (1 + (A_1 + A_2 C_p^2 + A_3 C_p^3) C_{SEP}^{S_p}), \qquad (4.1)$$

where  $\mu_p^0$  is the polymer viscosity at the zero shear rate,  $C_{SEP}$  is the effective salinity to polymer, and  $S_p$  is the exponent used in Eq. 4.2 (slope of the line in Figure 4.2b).

$$\frac{\mu_p^0 - \mu_w}{\mu_w} \propto C_{SEP}^{S_p}.$$
(4.2)

Finally, shear rate effect is modeled via Eq. 4.3

$$\mu_p = \mu_w + \frac{\mu_p^0 - \mu_w}{1 + (\frac{\dot{\gamma}}{\dot{\gamma}_{1/2}})^{P_\alpha - 1}},\tag{4.3}$$

where  $\dot{\gamma}_{1/2}$  and  $P_{\alpha}$  are fitting parameters.

The parameter  $\dot{\gamma}_{1/2}$  used in Eq. 4.3 is not constant and rheological data at different polymer concentrations and salinities shows significant variations in this parameter. In order to account for these effects,  $\dot{\gamma}_{1/2}$  is assumed to be of the from:

$$\dot{\gamma}_{1/2}(c_{SEP}, c_p) = (a_1 c_{SEP} + a_0) \exp(a_2 c_p),$$
(4.4)

where  $c_{SEP}$  and  $c_P$  are effective salinity to polymer and polymer concentration, respectively. Figure 4.3 shows variation of  $\dot{\gamma}_{1/2}$  with salinity and concentration for HPAM 3330 and the corresponding fit of Eq. 4.4 to the experimental data.



Fig. 4.2.: Polymer rheology data and model calibration. Left: dependency of polymer viscosity on its concentration and the corresponding model, Eq.4.1, at three different salinities. Middle: Polymer viscosity  $\left(\frac{\mu_p^0 - \mu_w}{\mu_w}\right)$  as a function of C<sub>SEP</sub> described by Eq. 4.2. C<sub>SEP</sub> is the same as the anion concentration here and is in meq/ml. Right: Effects of shear rate on the polymer viscosity captured by Eq. 4.3. Salinity is in ppm.



Fig. 4.3.: Variation of  $\dot{\gamma}_{1/2}$  with salinity and concentration for HPAM 3330 and its modeling using Eq. 4.4. Salinity is in ppm.

# Residual resistance factor (RRF) experiments

One of the most important effects of polymer flooding is its permeability reduction. In order to obtain correct model parameters to describe this process, we performed a single phase polymer flooding experiment with the following steps:

- 1. Waterflooding and measuring the absolute permeability.
- 2. Polymer flooding until pressure profile reaches steady state.
- 3. Waterflooding and measuring the absolute permeability.

The flooding experiments were carried out on a 0.5 ft long Berea sandstone (Length: 6", Diameter: 2") at 24 °C and flow rate of 0.25 cc/min. One can easily estimate polymer residual resistance factor by taking a ratio of mobility before and after polymer flooding. It is worth noting that in UTCHEM permeability reduction is not reversible, meaning that the polymer reduction factor is equal to its residual resistance factor. In addition to finding the permeability reduction factor for our polymer, we have used the pressure data in this experiment to approximate the inaccessible pore volume as well. Figure 4.4 shows the evolution of pressure drop along the core and simulation results. Choosing value of 0.068 for  $C_{rk}$  in Eq. 4.14 to model permeability reduction



Fig. 4.4.: Evolution of pressure drop in the residual resistance factor experiment.

factor and 0.85 for inaccessible pore volume gives the best match to our polymer flooding experiment.

# 4.4.2 Modeling surfactant flooding

Surfactant-oil-water systems at surfactant concentrations above critical micelles concentration (CMC) are complex and modeling their phase behavior requires a series of phase behavior experiments. The phase environment of such systems is strongly affected by the salinity with type II(-) emulsions forming at low salinities, type III at intermediate salinities, and type II(+) at high salinities. Phase behavior experiments were performed at room temperature and water to oil ratio (WOR) of 1. Based on the experimental data, solubilization ratios for oil and water were calculated and used for phase behavior modeling. After solubilization data is obtained, phase behavior can be represented through binodal curve and tie lines and Hand's rule [171–174]. Phase behavior modeling requires three input parameters related to solubilization ratios and two parameters defining salinities where transition from type II(-) to type III and from type III to type II(+) occurs (salinity window). Phase behavior experiments are quite complex and the solubilizations ratios and the salinity values at which change in microemulsion type occurs are subjected to uncertainty. Solubilization data and the resulting phase behavior model are shown in Figure 4.5.



Fig. 4.5.: Solubilization data as a function of effective salinity for phase behavior (CSE) for WOR of 1.

# 4.4.3 Capillary desaturation experiments

A high-permeability (air permeability  $\approx 700 \text{ mD}$ ) Berea core (Length: 6", Diameter: 2") was used to determine the capillary desaturation process through a flooding experiment. A commercial co-solvent in the reservoir synthetic brine was used to lower the interfacial tension (IFT) of the waterflood. The core was initially saturated with the reservoir synthetic brine + co-solvent (20% Vol/Vol) solution and aged with the reservoir oil for two weeks. It was assumed that the IFT of the system remained

Value
0.09
0.23
2.14, 14.2
2.80
2.5E-06

Table 4.2.: Initial parameters for the capillary desaturation experiment.

ually increasing the flow rate (0.09 to 100 cc/min) of the injectant (reservoir brine + co-solvent solution). Shown in Figure 5.1 is the capillary desaturation experimental results and its corresponding model. The residual oil saturation of  $\approx 10\%$  can be



Fig. 4.6.: Experimental results showing the reduction in the residual oil ratio vs. capillary number and the corresponding model, Eq. 4.8

achieved with a capillary number of 2.75E-03. So, for this particular Berea under the experimental conditions, an in-situ IFT reduction of 0.002-0.003 mN/m is required to achieve  $S_{orc} \approx 10\%$ . The experimental results agree well with the literature [175]. At intermediate capillary numbers, relative permeabilities and residual saturations are obtained by the interpolation given in Eq. 4.8,4.9.

#### 4.4.4 Conductivity experiments

Since our models does not account for heterogeneous permeability at the sub-core scale, adopting reasonable values for dispersivity is essential to account for additional mixing arising from inherent heterogeneity of cores. In order to estimate correct values of dispersivity, we used electrical conductivity (EC) as a direct indicator of the tracer concentration (i.e., ions). This is a common practice due to the simplicity of EC measurements and the simple relationship between EC and TDS, which is described as  $TDS = A \times EC$  [176,177]. In each coreflood experiment, EC of effluent is measured during the entire process, however, we only use the EC values corresponding to IWF and HTDS flooding, where oil production is at steady state and negligible. Figure 4.7 shows that adopting dispersivity value of 0.01 ft gives an excellent match to measured EC profile of effluent. This is also consistent with findings of other researchers for Berea sandstone [178]. Finally, we assume the same dispersivity for aqueous, oleic, and microemulsion phases.



Fig. 4.7.: Measured conductivity for known TDS solutions (outside the core) and its linear relationship with TDS in the range of 9400-20000 ppm (left). Salinity of effluent fluid in a coreflood experiment measured via its conductivity, resulting in dispersivity value of 0.01 ft (right).

#### 4.4.5 History matching and model validation

As described in previous sections, many of the model parameters can be obtained through different experiments than corefloods. However, some model parameters such as relative permeabilities at high capillary number, microemulsion viscosity, and phase behavior parameters need to be tuned through modeling the entire SP flooding process. In doing so, we honor the already estimated model parameters in the previous sections.

# Sensitivity analysis

We often do not have enough experimental data to find all the model parameters needed for SP flooding. Thus, it is crucial to assess the impacts of lack of knowledge about some model parameters on various quantities of interests (i.e., overall recovery). To do so, we adopted the Sobol method to carry out global sensitivity analysis (SA) [179, 180]. Global SA explores the entire range of model inputs (global rather than local) and ranks model inputs according to their contributions to the output variance [181]. Here, we emphasize on SA as it can potentially reduce the number of calibration parameters within each model drastically, thus making the history matching process tractable.

In this SA framework,  $X = (x_1, x_2, \dots, x_n)$  denotes the model input vector and Y = f(X) is the quantity of interest, where f is our deterministic model. It is worth mentioning that each variable in vector X is a random number with a probability distribution function (PDF). Such PDFs reflect our knowledge about each variable. In global SA, importance of each variable is quantified through sensitivity indices. First order effects of a variable i is calculated as:

$$S_{i} = \frac{V_{X_{i}}(E_{X \sim i}(Y|X_{i}))}{V(Y)},$$
(4.5)

where V denotes variance and the expectation is taken over the n-1 dimensional parameter space  $X_{\sim i}$ . An additional and more popular sensitivity index is the total-order sensitivity index [181], ST<sub>i</sub>, which quantifies the contribution of variable *i* including first order effects and its higher order effects to the output variance as:

$$ST_i = \sum_{k \neq i} S_k, \tag{4.6}$$

where  $k \neq i$  includes all the indices where variable *i* is involved. Total-order index is calculated as:

$$ST_{i} = \frac{E_{\sim i}(V_{X_{i}}(Y|X_{\sim i}))}{V(Y)}.$$
(4.7)

We perform SA on two levels. First by analyzing each model separately to identify the dominant model parameter within each model such as microemulsion viscosity with five input parameters (see Eq. 4.17). Since we history match both the cumulative oil production and pressure drop, the SA is carried out with respect to these two quantities of interest (i.e. final cumulative oil recovery and peak pressure drop). Similar to [147], we assume %20 uncertainty via uniform distribution in the experimentally measured data and consequently model parameters obtained via experimental data. For the parameters that were not experimentally measured namely those in IFT model (Chun Huh model, Eq. 4.11,4.12), ME viscosity model (Eq. 4.13), and Corey's exponent for relative permeability at high capillary number, we adopt the range from [170], [157, 182–185], and [186], respectively. We then generate sufficient number of model inputs (i.e., to ensure convergence of Sobol indices) using method of [179] and evaluate our two quantity of interests namely cumulative oil recovery (%OOIP) and maximum of dP via UTCHEM simulations. Finally, we compute the Sobol indecies based on the Sobol's method which is briefly described here (Eq. 4.5-4.7). Figure 4.8 summarizes a series of sensitivity simulations for the five models with more than one calibration parameter including IFT, residual resistance factor, phase behavior, ME viscosity, and dispersion models that are separately examined. The purpose of this analysis is to reduce the parameter space of each model rather than



comparing different models with each other. Results shown in Figure 4.8 suggest that,

Fig. 4.8.: Sensitivity analysis on different models in SP flooding using the Sobol technique.

for our parameter ranges and experiment designs, only a few parameters in the five model examined here are contributing to the variance of final outputs (cumulative oil recovery and maximum pressure drop). For example, in microemulsion viscosity model, the parameter describing the dependency of microemulsion viscosity on oil is the most significant one and the other four parameters including the one determining the contribution of brine have negligible effects. The same is true for other models such as Chun Huh model for IFT and residual resistant factor model for polymers both with only one significantly important parameter. Furthermore, this results show negligible effects of dispersivity of oil and microemulsion on overall oil recovery and maximum pressure drop along the core. Our next level of SA is done on the entire SP flood model, meaning that the dominant model parameters found in the previous step are used to rank the sensitivity of cumulative oil production and peak pressure drop to different models. This gives us an idea of how different models are contributing to the total variance of our outputs. Figure 4.9 shows the sensitivity of our two quantities of interests to different sub-models in a SP flood model. Here, we include two additional parameters namely residual saturation and relative permeability exponents in Corey model at high capillary number. We assume the same values of residual saturation and Corey's exponents for high capillary number flows and dispersivity for all the phases (i.e., aqueous, oleic, and microemulsion). Results from Figure 4.9 gives insight about



Fig. 4.9.: Sensitivity analysis on an entire SP flood model using the Sobol technique.

which model parameters should be targeted in the next section for history matching and which ones should be left out as they do not contribute to the output variance. For example, the constant, a, in the Chun Huh IFT model (Eq. 4.11) has almost no effects on either maximum pressure drop or cumulative oil production and thus must not be included into history matching process. Our SA also shows how critically important inaccessible pore volume (EPHI4) for polymer is with regards to maximum pressure drop, justifying its measurement it in a separate experiment (our polymer flooding experiment). Moreover, with regards to cumulative oil production, the most important parameter is dispersivity. Again, it justifies the need for measuring this model parameter via a separate experiment due to its high impact on the overall oil recovery as it can greatly improve the accuracy of the final SP flood model.

## Final model calibration and validation

After the first stage of model calibration using the experimental data and finding the most sensitive parameters in the remaining models, we aim at history matching the entire SP model including the remaining models that needed to be calibrated. Based on the SA presented in Figure 4.9, we aim at tuning microemulsion viscosity model ( $\alpha_2$ ), phase behavior model ( $h_1$ ), and Corey's exponent at high capillary numbers. As opposed to similar studies, where only the overall oil recovery is history matched [147], in this study we aim at history matching two key outputs namely, overall oil recovery and maximum pressure drop. This ensures a more realistic solution to the coreflooding problem. To carry out the history matching, we adopted a multistage history matching algorithm as shown in figure 4.10. In this approach, we first history match the waterflooding part by matching the oil production and pressure profile history. Next, we history match the performance of SP flood by matching the incremental oil recovery and ignoring the pressure profile developed during SP flooding. Once a match is achieved for overall oil recovery, we move to the final stage of the history matching by matching the maximum pressure drop. It is worth noting that history matching the pressure might impact the overall oil recovery in step two and thus it is necessary to correct for mismatched overall oil recovery after the pressure is history matched in an iterative fashion. Here, we use one coreflood



Fig. 4.10.: Flowchart of the multi-objective history matching process. The process includes three stages and in each stage only one objective function is minimized (i.e., difference between numerical and experimental values) using different calibration parameters for each stage.

experiment (BCF 1) for history matching and validate the obtained SP flood model by predicting two different coreflood experiments (BCF 2 and BCF 3). A summary of the injection schemes and chemical slug designs for our three coreflood experiments are shown in Table 4.3. Figure 4.11 summarizes the history matching process for BCF 1 from the 1<sup>st</sup> to 5<sup>th</sup> iteration. At the end of this process, a perfect match is obtained for cumulative oil recovery and maximum pressure drop. Figure 4.11 also presents how history matching BCF 1 is improving the model predictions for two other BCFs (also refer to Table 4.4). Figure 4.12 shows the history matched SP flood model with experimental results for BCF 1 as a function of pore volume (PV) of injected fluids. As can be seen, the bimodal experimental oil cut, corresponding to neat oil and oil+microemulsion productions in the experiment were accurately captured. Additionally, a good match is achieved for the overall oil recovery and pressure drop history. Next, we apply the same parameter set to model BCF 2 and

Slug	Property	BCF 1	BCF 2	BCF 3
IWE	PV	1.60	1.60	1.45
TAAT	TDS	9400	9400	9400
UTDO	PV	1.0	0.25	
ппра	TDS	17500	17500	
	PV	0.25	0.25	0.50
SP	TDS	17500	17500	9400
	Surf. conc.	6780	6780	6780
	Poly. conc.	2500	2500	3300
	PV	0.50	0.51	1.01
Р	TDS	9400	9400	9400
	Poly. conc.	2500	2500	3300
FWE	PV	1.59	1.49	1.27
EVVE	TDS	9400	9400	9400

Table 4.3.: Injection sequence and data of BCF experiments. Concentrations are in ppm.



Fig. 4.11.: History matching results with BCF 1. Predictions by history matched model in each iteration for BCF 2 and 3 are also shown.

Table 4.4.: History matching the SP flood model for BCF 1

Physical parameter	Variable	1 <sup>st</sup> iteration	$2^{nd}$ iteration	3 <sup>rd</sup> iteration	4 <sup>th</sup> iteration	5 <sup>th</sup> iteration
Surfactant phase behavior	$h_1$	0.009	0.0155	0.0155	0.0155	0.0155
Corey's exponent at high Ca	e1c, e2c, e3c	1	1	2	2	2
Microemulsion viscosity	$\alpha_{ m V2}$	1	1	1	1.5	2
Cumulative oil production	OOIP%	92	89	81.3	80	79.4
Cumulative oil production (Exp.)	OOIP%	79.63	79.63	79.63	79.63	79.63
Max. pressure drop	dP(psi)	2.76	2.77	4.26	4.96	5.66
Max. pressure drop (Exp.)	dP(psi)	5.6	5.6	5.6	5.6	5.6

BCF 3 for model validation. Our experimental data shows extreme sensitivity for oil production to HTDS pre-flush. In BCF 1, 1 PV of HTDS pre-flush was used to



Fig. 4.12.: History matching results with BCF 1. From left to right: oil cut, cumulative oil production, and pressure drop.

elevate the salinity level of the core to the optimum salinity where we observed the maximum recovery. In BCF 2 and BCF 3, we reduced the slug size of HTDS pre-flush to 0.25 PV and zero, respectively, resulting in lower overall oil recoveries compared to that in BCF 1. Shown in Figure 4.13 is the validation study with BCF 2 and BCF 3. It is worth noting that there is a drop in the overall oil recovery consistent with the experimental results. Again, oil cut, cumulative oil recovery, and pressure drop profile were successfully predicted. Table 4.5 summarizes all the parameters used in



Fig. 4.13.: Validation study with BCF 2 (top row) and BCF 3 (bottom row). From left to right: oil cut, cumulative oil production, and pressure drop.

our SP flood model.

UTCHEM parameter(s)	Description	Value(s)
Polymer models		
AP1, AP2, AP3	Zero shear viscosity multipliers	15, 0, 155
BETAP	Effective divalent salinity parameter	1
CSE1	Salinity above which viscosity is affected	0.0001
SSLOPE	Log-log slope of viscosity-effective salinity	-0.76
GAMMAC, POWN	Polymer shear rate dependency parameters	40, 1.66
A0, A1, A2	Constants in Eq. 4.4	-588,7683,-12
Surfactant models		
C2PLC, C2PRC	Oil concentrations at the plait point of type II $(-)$ , and type II $(+)$	0, 1
EPSME	Critical micelle concentration	0.0001
CSEL,CSEU	Salinity window for type III	0.255, 0.305
HBNC70, 71, 72	Heights of binodal curve at zero, optimal, and twice optimal salinities	0.027,  0.0155,  0.02
chuh, ahuh	Constants in Chun Huh IFT model	0.3, 10
Xiftw	Log of oil-water interfacial tension	1.32
ALPHAVi; i: 1–5	ME viscosity parameters	2, 2, 0, 0.9, 0.7
Chemical-rock interactions		
t11, t22, t33	CDC parameters for the aqueous, oleic, and ME phases	1500, 1100, 346
e1c, e2c, e3c	Corey exponents at high Ca numbers for the aqueous, oleic, and ME phases	2.0, 2.0, 2.0
AD31, AD32, B3D	Surfactant adsorption parameters	0.31, 0, 100
AD41, AD42, B4D	Polymer adsorption parameters	8.4, 0, 100
EPHI4,BRK,CRK	Polymer permeability reduction parameters	0.85, 100, 0.068

Table 4.5.: Final SP flood model parameter set

#### 4.5 Conclusion

In this study, we showed that model calibration and history matching can be greatly facilitated by means of sensitivity analysis, and we presented a comprehensive methodology integrating extensive experimental data required for a successful SP flood modeling. Some of the key model parameters such as those in polymer permeability reduction factors can be determined prior to SP coreflood modeling and thus reduce the uncertainty of final SP flood model. Using the rheological data for polymer, phase behavior tests, capillary desaturation and polymer RRF experiments we were able to build a SP flood model via a multi-stage calibration algorithm where the cumulative oil production and pressure profile were successfully history matched. Improving the polymer shear-thinning model in UTCHEM led to better agreement between experimental and simulation pressure profile among three different BCF experiments. We also provided insight into the relative importance of some of key model parameters in SP flooding. Results presented in Figures 4.8-4.9 can be readily used for other SP flooding experiments to build accurate history matched models. However, one limitation would be regarding phase behavior modeling, as in our experiments we have explored the optimum and under-optimum salinity regimes. It is likely that results presented in Figures 4.8-4.9 slightly change for over-optimum salinity regime, however, it must be noted that the main insight from our sensitivity analysis is relative importance of model parameters rather than their exact sensitivity indices. Finally, we showed that our model can accurately predict the performance of two additional independent coreflood experiments conducted with different slug design and thus salinity profiles.

#### 4.6 Acknowledgement

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# 4.7 Appendix

# 4.7.1 Capillary Number and Relative Permeability

The residual saturation is modeled based on the trapping number as shown below:

$$S_{lr} = min\left(S_l, S_{lr}^{high} + \frac{S_{lr}^{low} - S_{lr}^{high}}{1 + T_l(N_{T_l})^{\tau l}}\right)$$
(4.8)

where  $\tau l$  and  $T_l$  are fitting parameters for trapped saturation data for phase l (l=1: aqueous, l=2: oleic, l=3: microemulsion).  $S_{lr}^{high}$  and  $S_{lr}^{low}$  are trapped saturation at high and low capillary number respectively. We use a value of one for  $\tau_l$ . The endpoint relative permeability for each phase increases in a very predictable way as the trapping number increases. The following equation is used to modify the endpoint relative permeability based on residual saturation of conjugate phase:

$$k_{rl}^{0} = k_{rl}^{0,low} + \frac{S_{l'r}^{low} - S_{l'r}}{S_{l'r}^{low} - S_{l'r}^{high}} \left(k_{rl}^{0,high} - k_{rl}^{0,low}\right)$$
(4.9)

where  $S_{l'r}$  is the residual saturation of the conjugate phase.  $k_{rl}^{0,high}$  and  $k_{rl}^{0,low}$  are endpoint relative permeability at high and low capillary number respectively. Now, using a simple function such as Corey-type relative permeability equation, we can calculate relative permeability.

$$k_{rl} = k_{rl}^0 (\overline{S_l})^{n_l} \tag{4.10}$$

where  $\overline{S_l} = \frac{S_l - S_{lr}}{1 - S_{wr} - S_{or}}$  is the normalized saturation and  $n_l$  is the Corey exponent for  $l^{th}$  phase.

# 4.7.2 Interfacial Tension Model

We use Chuh-Huh equation to model interfacial tension which correlates the interfacial tension to the solubilization ratio as:

$$\sigma_{l3} = \sigma \exp(-aR_{l3}) + \frac{cF_l}{R_{l3}^2} (1 - \exp(-aR_{l3}^3)), \ l = 1, 2$$
(4.11)

where  $\sigma_{ow}$  is oil-water interfacial tension,  $R_{l3}$  is the solubilization ration for  $l^{th}$  phase, a and c are fitting parameters. The Hirasaki's correction factor,  $F_2$ , is defined as :

$$F_{l} = \frac{1 - \exp(-\sqrt{con_{l}})}{1 - \exp(-\sqrt{2})}$$
(4.12)

where  $con_l = \sum_{k=1}^{3} (C_{kl} - C_{k3})^2$ 

#### 4.7.3 Permeability Reduction Model

The model used for permeability reduction accounts for both the reduction in mobility of the displacing fluid as well as reduction in effective permeability of the porous medium. This model assumes that the permeability reduction is irreversible i.e. permeability reduction does not decrease with the decrease in polymer concentration. The equation for permeability reduction is given by:

$$R_k = 1 + \frac{(R_{k,max} - 1)b_{rk}C_{4l}}{1 + b_{rk}C_{4l}}$$
(4.13)

where  $R_{k,max}$  is modeled as:

$$R_{k,max} = \max\left[\left[1 - \frac{c_{rk}(A_{p1}C_{SEP}^{S_p})^{1/3}}{(\sqrt{\frac{k_x k_y}{\phi}})^{1/2}}\right], 10\right]$$
(4.14)

where  $c_{rk}$  and  $b_{rk}$  are input fitting parameters, maximum  $R_k$  is taken as 10.

# 4.7.4 Adsorption Model for Surfactant and Polymer

The effect of adsorption is assumed to be irreversible with concentration and reversible with salinity. We use Langmuir type isotherm to model the adsorption of both the surfactant and polymer given by:

$$\hat{C}_m^* = \frac{a_m(\tilde{C}_m - \hat{C}_m^*)}{1 + b_m(\tilde{C}_m - \hat{C}_m^*)}, \ m = 1, 2$$
(4.15)

where

$$a_m = (a_{m1} + a_{m2}C_{SEP})(\frac{k_{ref}}{k})^{1/2}$$
(4.16)

where m = 1 is surfactant and m = 2 is polymer,  $a_3$  and  $b_3$  are fitting parameters,  $C_{SEP}$  is effective salinity,  $\overline{C}_m^*$  is volume of adsorbed surfactant or polymer/volume of water, and  $\tilde{C}_m$  is adsorbed surfactant or polymer concentration/pore volume.

# 4.7.5 Microemulsion viscosity

The micro-emulsion viscosity is modeled using the following model:

$$\mu_{3} = C_{13}\mu_{w} \exp[\alpha_{1}(C_{23} + C_{33})] + C_{23}\mu_{o} \exp[\alpha_{2}(C_{13} + C_{33})] + C_{33}\alpha_{3} \exp[\alpha_{4}C_{13} + \alpha_{5}C_{33}]$$
(4.17)

where  $\alpha_1, \alpha_2, \alpha_3, \alpha_4, \alpha_5$  are fitting parameters.  $\mu_w$  and  $\mu_o$  are water and oil viscosity and  $C_{kl}$  is concentration of  $k^{th}$  component in  $l^{th}$  phase where k = 1: water, k = 2: oil, k = 3: surfactant, and l = 1: aqueous, l = 2: oleic, and l = 3: microemulsion.

# 5. UNSTABLE DISPLACEMENT OF NON-AQUEOUS PHASE LIQUIDS WITH SURFACTANT AND POLYMER

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#### 5.1 Abstract

In this paper we study two-phase multicomponent displacement of two immiscible fluids in both homogeneous and heterogeneous porous media. In many applications such as enhanced oil recovery, fluid mixing and spreading can be detrimental to the efficacy of the process. Here, we show that when an initially immobile phase is being displaced by a finite-size slug of solvents (surfactant and polymer), viscous fingering significantly enhances mixing and spreading of solvents. This effects are similar to those caused by medium heterogeneity and lead to poor displacement efficiency. We first quantify the efficacy of our displacement subject to different mobility ratios, Peclet numbers, and levels of medium heterogeneity. We observe a non-monotonic behavior in displacement efficiency as a function of mobility ratio, indicating that although stable frontal interface is desirable, miscible viscous fingering on the rear interface will eventually disintegrate the solvents slugs and reduce the displacement efficiency. Then, we show that miscible viscous fingering developing on the rear interface of the chemical slug could be greatly suppressed when viscosity contrast is gradually decreased using exponential or linear functions, leading to 10% increase in displacement efficiency while using the same amount of chemicals. To elucidate this low displacement efficacy, we study the evolution of mixing, spreading, and interfacial length and show that while higher viscosity ratios are quite effective in mobilizing the initially immobile phase in 1D displacements, they are in fact detrimental in 2D unstable displacements since they enhance mixing and spreading of solvents.

#### 5.2 Introduction

Spatially fluctuating velocity field in porous media causes concentration fields to spread [99]. Spreading creates new fluid-fluid interface and thus enhances mixing through diffusion. Mixing and spreading of passive tracers in porous media have been intensively studied due to their fundamental role in many applications such as CO<sub>2</sub> sequestration [187–189], enhanced oil recovery [190,191], groundwater flows [192,193], and reactive transport [194–196].

Recently many studies have focused on quantifying mixing in porous media due to their importance primarily in reactive transport [197]. Besides variation in rock properties that is a primary cause of spreading and mixing [198, 199], hydrodynamic instabilities can also further enhance mixing in flow through porous media [200–203]. Such hydrodynamic instabilities are ubiquitous in natural and industrial process due to viscosity (or mobility in the case of multi-phase flow) or density contrast.

Displacement of a more viscous fluid by a less viscous fluid leads to a hydrodynamic instability called viscous fingering [204–206]. In the realm of two-phase flows, viscous fingering has been studied for decades through experiments [207–210], numerical simulations [211–213], and linear stability analysis [214,215]. Different aspects of viscous fingering for immiscible displacements such as effects of relative permeability [216], permeability heterogeneity [217,218], viscosity ratio and gravity [219], and flow rate [220] have been studied. A series of numerical simulations and experiments for drainage process were performed in [221] where the region of stable displacement was identified as a function of viscosity ratio and capillary number (Ca).

Most previous studies have focused on drainage and imbibition processes in the absence of solvents. Solvents such as polymers and surfactants play a central role in many technological processes such as enhanced oil recovery (EOR) [140] and surfactant enhanced aquifer remediation [167, 222–224]. For example, in mature oil reservoirs more than two-third of oil remains trapped in small pores as a disperse phase due to capillary forces where injecting surfactants and polymers have been proven very effective in mobilizing the remaining oil in place [225–228]. In this paper, we focus on a tertiary enhanced oil recovery process called surfactant-polymer (SP) flooding [168]. SP flooding efficacy in mobilization of non-aqueous phase liquids (NAPL) is due to its synergy between polymer and surfactant. The key mobilization mechanism in SP flooding is lowering the interfacial tension (IFT) between the two immiscible fluids by injection of surfactant in concert with enhanced sweeping efficiency due to polymer injection.

In this study, we investigate multi-phase transport at continuum (Darcy) scale in a rectilinear domain with line source injection [229]. Efficacy of SP flooding is directly related to stable displacement of displacing fluids often referred to as piston-like displacement [230–232]. In piston-like displacement there is a sharp interface between displacing and displaced fluid. However, this condition virtually never occurs due to variations in rock properties (i.e., permeability and porosity) and hydrodynamic instabilities. In practical applications, due to economical and technical considerations, surfactant (s) and polymer (p) are only injected for a small pore volume (PV). This creates a leading and trailing interfaces. These interfaces are both prone to become unstable and will interact with each other due to finite width of injected solvent slug [233]. We aim to answer following questions: how does mobility ratio change spreading and mixing of solvents? what are the effects of finite-size solvent slug, and finally how does recovery rate depend on hydrodynamic instabilities. We perform high-resolution numerical simulations and analyzing degree of solvent mixing, its spreading, breakthrough curves, and evolution of interfacial length.

#### 5.3 Mathematical method

#### 5.3.1 Governing equations

We study a multicomponent (wetting, non-wetting, surfactant, and polymer) twophase (wetting and non-wetting) imbibition process in which a wetting phase is injected into a porous media to displace a non-wetting phase. Transport equation for each component is given by:

$$\phi \frac{\partial \left(S_w C_{iw}\right)}{\partial t} + \nabla \cdot \left(\mathbf{u}_w C_{iw} - \phi S_w \mathbf{D} \nabla C_{iw}\right) = R_i, \ i = w, s, p \tag{5.1}$$

with  $S_w$  the wetting phase saturation,  $\mathbf{u}_w$  Darcy flux of wetting phase,  $\phi$  the porosity, and  $C_{iw}$  the concentration of wetting phase (w), surfactant (s), and polymer (p) in the wetting phase and  $R_i$  is the source/sink term accounting for injection/production of each component. In this study, solvents (surfactant and polymer) are only soluble in the wetting phase and non-wetting phase saturation,  $S_n$ , is calculated as  $1 - S_w$ . In Eq. 5.1, **D** is the dispersion tensor and is given as [199]:

$$D_{ij} = \left(D_m + \left(\alpha_T |\mathbf{u}_w|\right)\right) \delta_{ij} + \left(\alpha_L - \alpha_T\right) \frac{u_{wi} u_{wj}}{|\mathbf{u}_w|}, \ i = x, y$$
(5.2)

where  $D_m$  is the molecular diffusion coefficient and  $\alpha_L$  and  $\alpha_T$  are longitudinal and transverse dispersivities of solvents in the wetting phase, respectively. The Darcy phase velocities for each phase are given by:

$$\mathbf{u}_{i} = -\frac{k_{ri}}{\mu_{i}} \mathbf{K} \left( \nabla p_{i} + \rho_{i} g \nabla z \right), \ i = w, n$$
(5.3)

where **K** is the absolute permeability tensor of medium, g and z are the gravity and depth, respectively. Moreover,  $\mu_i$ ,  $\rho_i$ ,  $k_{ri}$  are the viscosity, density, and relative permeability of phase i where w and n denote wetting and non-wetting phases, respectively. Here, relative permeabilities are modeled based on Corey-type functions [234, 235]. These functions for the wetting and non-wetting phases have the general form of:

$$k_{ri} = k_i^e (S_{ni})^{ei}, \ i = w, n \tag{5.4}$$

where  $k_i^e$  is the endpoint relative permeability for each phase and ei is the exponent of the each phase Corey function.  $S_{ni}$  is the normalized saturation of phase *i* defined as:

$$S_{ni} = \frac{S_i - S_{ir}}{1 - S_{wr} - S_{nr}}, \ i = w, n \tag{5.5}$$

where  $S_{ir}$  is the residual saturation of phase *i* during drainage (i = w) and imbibition (i = n) processes, respectively. End-point relative permeabilities along with Corey's exponents and residual saturations fully describe a set of relative permeability curves for wetting and non-wetting phases. Displacement of trapped non-wetting phase is governed by the interplay of viscous and capillary forces and thus is a function of  $Ca = \frac{|K\nabla p|}{\sigma}$  where  $\sigma$  is IFT [236]. Injection of surfactant increases the Ca number by orders of magnitude which results in mobilization of trapped fluid, a process called capillary desaturation [175, 237]. Shown in Figure 5.1 is the capillary desaturation curve used in this study which is obtained from experiments on sandstone (see [228]). In addition to reducing the residual saturation, an elevated Ca number further modifies the shape of relative permeability curves, shifts them toward linear functions of  $S_{nw}$  ( $ei \rightarrow 1$ ) [239–242]. In this study, we use relative permeability data at low Ca number (during waterflooding) obtained via experiments on sandstone. At high Ca numbers (ultra-low IFT,  $Ca \approx 10^{-1}$ ) it is assumed that relative permeability curves are linear with zero residual saturation [243, 244] (see Figure 5.2). We do not consider gravitational effects and only focus on viscous fingering. Moreover, the capillary pressure is neglected due to an ultra-low IFT during surfactant flooding. We use the Implicit Pressure Explicit Saturation (IMPES) algorithm [245–247]. Pressure field is obtained by solving a linear system of equations arising from imposing an



Fig. 5.1.: Effects of increasing Ca number on residual saturation of non-wetting phase.  $S_{nr}^*$  is normalized with residual saturation after waterflooding (i.e. low Ca number). The normalized residual saturation is modeled as  $\frac{1}{1+TCa}$  where T is a fitting parameter [167,238].



Fig. 5.2.: Impact of increasing Ca number on relative permeability curves.

overall mass balance on volume-occupying phases (i.e. here wetting and non-wetting phases). Once the pressure field is known the phase velocities are calculated via Eq. 5.3 and then components concentration (wetting phase, surfactant, and polymer) are obtained by explicitly solving Eq. 5.1.

A surfactant concentration above its critical micelle concentration (CMC) creates a complex surfactant/oil/water phase behavior [171, 248, 249]. Here, surfactant is present at concentrations below its CMC where there exist only two phases namely wetting and non-wetting phases with former one containing all the surfactant. We use experimental data for a commercial surfactant to model IFT as a function of surfactant concentration as shown in Figure 5.3.



Fig. 5.3.: Variation of IFT between wetting and non-wetting phases with respect to surfactant concentration in the wetting phase. Note that IFT remains constant for concentrations above CMC.

Polymer injection is an effective technique for mobility control and better sweeping efficiency [140, 250]. Here, we use the experimental data obtained for a Hydrolyzed polyacylamide (HPAM), one of the most widely used and successful polymer types [251, 252]. In general, the viscosity of polymer solution depends on the shear rate, salinity, and concentration of polymer. In this study, we neglect the other two effects and only consider dependency of polymer viscosity on its concentration given by the Flory-Huggins equation [253]:

$$\mu_p = \mu_w \left( 1 + A_{p1}C_p + A_{p2}C_p^2 + A_{p3}C_p^3 \right) \tag{5.6}$$

where  $\mu_w$  is the viscosity of the wetting phase without polymer and  $C_p$  is the polymer concentration in the wetting phase. Fitting parameters  $A_{p1}$ ,  $A_{p2}$ , and  $A_{p3}$  are obtained from rheology experiments on HPAM 3230 as shown in Figure 5.4 (see [228] for detailed rheological data).



Fig. 5.4.: Variation of viscosity of polymer solution with respect to polymer concentration.

#### 5.3.2 Problem setup

We are interested in investigating hydrodynamics instabilities occurring on leading and trailing edges of a finite-size solvent slug and their impacts on solvents' mixing, spreading, and efficiency in mobilizing an immobile non-wetting phase. As shown in Figure 5.5, our computational domain is a rectangular with aspect ratio of L/W = 10. It is assumed that solvents have small diffusivity within the wetting phase which results in  $Pe = \frac{UL}{D} = 10^6$  where U is the Darcy velocity, L is the length-scale (either length or width of the domain), and D is the solvent diffusivity in the wetting phase. Here, the computational domain is discretized by a fine  $200 \times 500$  mesh. We have performed additional simulations with coarser grids and observed nearly identical overall recovery and breakthrough curves [202]. Furthermore, we use a total variation diminishing (TVD) flux-limited higher-order scheme for convection terms which minimizes numerical dispersion. This is necessary to accurately capture the smallscale viscous fingering. Solvents such as surfactant and polymer are often used after waterflooding process. Thus, to obtain a realistic condition, we performed a separate simulation to obtain the saturation profile of the wetting phase for the entire domain. After waterflooding, average of wetting phase and non-wetting phase saturations are 0.55 and 0.45, respectively  $(S_w + S_n = 1)$ . In all simulations, solvents are injected through vertical wells covering the entire depth of the domain where it takes two days to inject one pore volume (PV) of fluids. It is worth noting that we do not place the solvent slug inside the domain and instead inject it similar to real applications. After the desired amount of solvents are injected we keep injecting pure wetting phase (with no solvent) for two PVs. The ratio of viscosity of non-wetting phase to that of pure wetting phase (without polymer) is  $\mu_n/\mu_w = 10$ . In all simulations surfactant is injected at its CMC concentration and for our baseline case polymer concentration is chosen such that the wetting phase viscosity be equal to that of non-wetting phase. A no-flow boundary condition is imposed in the transverse direction. To trigger hydrodynamic instabilities, the injected concentration of polymer is randomly perturbed with a small amplitude of 1% of the injected concentration and duration of 0.001 PV in all cases. In all simulations, the time step is selected during run-time so that maximum CFL=0.01 is achieved.



Fig. 5.5.: Schematic of problem setup with initial and boundary conditions.

# 5.4 Results

We expect two types of displacement and viscous fingering instability. On the frontal interface, the displacement and consequently the viscous fingering are close to those of immiscible condition whereas on the rear interface they are close to those of miscible condition as there is little to no mobile non-wetting phase. Miscible displacement becomes unstable to transverse perturbations when viscosity ratio of displacing fluid is less than that of displaced fluid. This criterion is always satisfied in our study and thus displacement is unstable on the rear interface. This is also the case for real applications as injecting an infinitely long solvent slug to prevent viscous fingering on the rear interface is not feasible and economical. One remedy to this situation is to gradually decrease (graded slug) the solvent concentration (i.e., only polymer) rather than a sharp decrease to zero. We will discuss the effects of graded polymer injection later. In immiscible displacement flow becomes unstable when mobility ratio calculated as:

$$M = \frac{\lambda_w}{\lambda_n} = \frac{k_{rw}/\mu_w}{k_{rn}/\mu_n} \tag{5.7}$$

becomes larger than one. Mobility ratio is thus the key parameter governing the stability of the displacement. In the works of [215,254], end-point relative permeabilities  $(k_{n,w}^{e})$  were used to evaluate Eq. 5.7. In this approach, mobility ratio does not depend on the shape of relative permeability curves (i.e., only depends on end-point relative permeabilities). This is in contrast to finding of [216] where they found that shape of relative permeability curves greatly influences the onset of instability. Solution of Eq. 5.1 in 1D is a well-known Buckley-Leverett profile which features a saturation shock and instabilities in immiscible displacement are localized around the shock region [214]. It has been shown in the works of [255,256] that mobility ratio measured across the shock  $(M_s)$  should be used to assess the onset of instability. Moreover, imbibition experiments in [257] shows that shock mobility ratio correctly predicts the onset of flow instability (as opposed to end-point mobility ratio). Properties of saturation shock (i.e., shock velocity, saturation, etc.) could be obtained from fractional flow curves. However, as opposed to the case of constant relative permeabilities, we cannot construct fractional flow curves due to spatio-temporal change of relative permeability curves as surfactant and polymer flow through the medium. Nevertheless, displacement on the frontal interface is still an immiscible one and a saturation shock forms at a specific saturation and travels at a constant speed and displacement is expected to become unstable when  $M_s > 1$  similar to the case of constant relative permeabilities. Thus, to confirm this, we perform a series of unperturbed displacement simulations with three values of polymer concentrations and numerically obtain shock mobility ratio. In our work, shock mobility ratio is defined by the ratio of total mobilities upstream and downstream of shock front [258]. As shown in Figure 5.6, for three cases with different viscosity ratios (i.e., different polymer concentration) there forms a saturation shock and mobility ratio across the shock is obtained accordingly. Performing the same simulations but this time with perturbed interfaces shows that, consistent with the theory, displacement remains stable for shock mobility ratio smaller than unity whereas it is unstable for shock mobility ratio greater than unity. In the present study, we also investigate the effects of medium heterogeneity



Fig. 5.6.: Top row: The formation of saturation shock at specific saturations for three cases of polymer concentration which results in  $\mu_n/\mu_w = 4.39, 1.05, 0.36$  (unperturbed cases). Arrow shows the saturation shock location. Bottom row: snapshots of wetting phase saturation at PV = 0.075 (perturbed cases). Long fingers develops around the saturation shock when  $M_s > 1$  whereas displacement is stable for  $M_s < 1$ .

on the displacement efficiency. In addition to a homogeneous permeability field, three heterogeneous permeability fields with isotropic, multi-lognormal, and stationary distributions are considered. An exponential covariance function with correlation lengths of 0.1 of domain size (i.e., mean flow parallel to bedding [259]) and log-k variances of 0.01,0.1, and 1 is used to build heterogeneous permeability fields (Figure 5.7). All permeability fields have a constant porosity of 0.2 and geometric mean permeability (K) of 420 md.



Fig. 5.7.: Three heterogeneous permeability fields used in this study with increasing level of heterogeneity ( $\sigma_{\rm lnK}^2 = 0.01, 0.1, 1$ ) from left to right.

# 5.4.1 Displacement efficiency

In surfactant enhanced aquifer remediation or enhanced oil recovery applications displacement efficiency is the foremost parameter. Displacement efficiency (recovery) is quantified by calculating the ratio of the total amount of recovered (produced) immobile phase to its initial in-place amount. Here, the wetting phase displaces the non-wetting phase and we report displacement efficiency as the percentage of the nonwetting phase produced after injecting 2 PV of wetting phase. To properly quantify how viscous fingering and heterogeneity affects the displacement efficiency, we first carried out a series of simulations in a homogeneous permeability field with no perturbation of interfaces (referred to ideal displacement hereafter) thus eliminating the two main sources that negatively impact the displacement efficiency. Such a condition yields to a displacement type often called piston-like displacement where the vertical sweeping efficiency is maximum and displacement efficiency is the highest. In real applications, however, a piston-like displacement cannot be achieved due to hydrodynamic instabilities and medium heterogeneity. In figure 5.8, we present the effects of viscosity ratio, Pe number, and medium heterogeneity on the displacement efficiency of our problem of interest. Intuitively, more of immobile phase could be displaced by more favorable mobility ratios (i.e., through adding more polymer to the wetting phase). This is however true only for an ideal (piston-like) displacement. As shown in Figure 5.8a, the displacement efficiency increases monotonically as the viscosity ratio  $(\mu_n/\mu_w)$  and consequently the mobility ratio reduces. In a realistic displacement process driven by a finite-size slug of solvents (i.e. surfactant and/or polymer) the displacement efficiency is not solely governed by the favorable mobility ratio across the saturation shock but also hydrodynamics of the rear interface. Figure 5.8a shows that in a 2D displacement a more favorable mobility ratio (on the frontal interface) is not as effective as it is in an ideal displacement and in fact could negatively impact the displacement efficiency by creating unfavorable viscosity ratio (on the rear interface). This effect is attributed to excessive viscous fingering occurring on the rear interface as the viscosity ratio becomes more favorable on the frontal interface. Theoretically, lower values of Pe number implies stronger diffusive forces and consequently more dilution of solvents within the wetting phase; one thus expects lower displacement efficiency as a result of stronger solvent dilution. Figure 5.8b shows that for ideal displacements this effect is not important until around a very low Pe number (Pe  $\approx 10$ ) and above this value displacement efficiency is not negatively affected by the Pe number. Interestingly, for 2D cases, this effect is completely reversed and by reducing the Pe number the displacement efficiency dramatically increases such that for Pe number of 10, an unstable displacement (2D) recovers almost the same amount of non-wetting phase as in its corresponding ideal displacement. Although stronger diffusive forces severely smear out the solvent concentration and it could be detrimental to the displacement efficiency, such a smearing effect heavily suppresses and delays the growth of fingers on the rear interface, resulting in almost the same displacement efficiency as in the ideal case. In addition to hydrodynamic instabilities caused by unfavorable viscosity (mobility) ratio, medium heterogeneity could be detrimental to displacement efficiency through enhanced dilution of solvents and channeling through high-permeability pathways. Figure 5.8c quantifies such negative effects on displacement efficiency. It is clear that compared to the case of homogeneous permeability field, even a very small value of heterogeneity ( $\sigma_{lnK}^2 = 0.01$ ) drastically reduces recovery and this effect is stronger for more heterogeneous media. Figure 5.9 shows



Fig. 5.8.: Effects of Pe number, viscosity ratio, and medium heterogeneity on the displacement efficiency and comparisons with corresponding piston-like (ideal) displacement.

surfactant concentration field for different viscosity ratios, Pe numbers, and levels of medium heterogeneity.

Based on our results presented in Figures 5.8a,b, it is clear that viscous fingering occurring on the rear interface due to inevitable unfavorable viscosity ratio is the main reason for deviating the displacement regime from the piston-like or the ideal one. Ideally, one aims to attenuate the viscosity contrast on the rear interface while making sure enough polymer exists throughout the solvent slug to efficiently displace the immobile non-wetting phase. To suppress the viscous fingering, various solutions have been proposed such as addition of a low dose of nanoparticle to base fluids [260], varying injection ratio of the electric current to flow rate [261], or by gradual reduction of pore sizes along the flow direction [262]. While those ideas are exciting and show great promise in reducing the extent of viscous fingering and flow instabilities, they often are not applicable to large scale problems of our interest (e.g. chemical enhanced oil recovery). A possible solution has been sought through grading (tapering) the polymer solution such that the polymer concentration is gradually reduced and thus decreases the negative effects of viscous fingering in two ways: (i) reducing the viscosity contrast on the rear interface and thus suppresses the growth of viscous fingers, (ii) enlarging the polymer slug (note that injected polymer mass remains



Fig. 5.9.: Top row: snapshots of surfactant concentration  $(c/c_0)$  for three different injected polymer concentrations results in  $\mu_n/\mu_w = 4.39$ , 1.05, and 0.36 from left to right at PVI = 0.2. It is clear that smaller viscosity ratio suppresses instabilities on the frontal interface while promoting viscous fingering on the rear interface. Middle row: snapshots of surfactant concentration  $(c/c_0)$  for three different Pe numbers of  $10^6$ ,  $10^3$ , and  $10^2$  from left to right at PVI = 0.5. Stronger diffusive forces suppress viscous fingering and thus integrity of the solvent slug is preserved for longer times, resulting in a higher overall displacement efficiency even though higher diffusion of solvents within the wetting phase and hence dilution of the solvent concentration field is not desired, but here the gains through delayed hydrodynamic instabilities are much higher. Bottom row: snapshots of surfactant concentration  $(c/c_0)$  for three different heterogeneity permeability fields with an increasing level of heterogeneity  $(\sigma_{lnK}^2 = 0.01, 0.1, 1)$  from left to right at PVI = 0.2.

constant or could be even smaller than the conventional scenario) and thus it takes more time for the pure wetting phase to develop fingers through the solvent slug and disintegrates it [157,263,264]. Here, we are interested in studying the efficacy of such approach in which we keep the total amount of injected polymer constant and continuously decrease its concentration as a function of time. We used three exponential (with different decay rates) and linear functions to obtain the polymer concentration profile at the injection well as a function of time. Again, we have performed two series of simulations corresponding to 2D unstable displacements and piston-like or ideal displacements. It is worth noting that grading the polymer reduces the maximum achievable displacement efficiency compared to the case of pulse injection. Figure 5.10 shows a comparison of various grading functions and how they perform compared to their corresponding ideal case. It can be seen that the graded viscosity profile re-



Fig. 5.10.: The effects of graded polymer injection and comparison with the corresponding piston-like (ideal) displacement. It is found that grading the polymer slug improves the displacement efficiency for all types of grading (i.e. exponential and linear functions) due to suppressing the viscous fingering through creating smaller viscosity ratios on the rear interface. Note that the maximum efficiency indicated by the ideal recovery is slightly reduced by the grading of the polymer slug. This reduction in displacement efficiency is due to smaller polymer concentration around the saturation shock where it is needed the most. Linear grading achieves the highest efficiency.  $H(t_0 - t)$  is the Heaviside function.

gardless of its shape (exponential with different decay rates or linear) improves the displacement efficiency even though the maximum achievable displacement slightly drops compared to the case of pulse injection. Effects of graded polymer injection on the development of viscous fingering on rear and frontal interfaces are illustrated in Figure 5.11.

# 5.4.2 Mixing, spreading, and interfacial length

As mentioned and showed before, in applications such as enhanced oil recovery or surfactant remediation mixing and spreading of the solvent slug is detrimental to displacement efficiency. It is thus important to quantify to what degree our solvent



Fig. 5.11.: Snapshots of surfactant concentration  $(c/c_0)$  at PVI = 0.3 for three polymer injection schemes from left to right:  $c_0H(0.15 - t)$ ,  $c_0e^{-6.3t}H(0.45 - t)$ , and  $c_0e^{-3.0t}H(0.2 - t)$ . The graded polymer injection effectively suppresses the development of viscous fingering.

slug mixes and spreads as it travels from the injection well to production well. In the case of homogeneous medium, hydrodynamic instabilities (i.e. viscous fingering) are responsible for mixing and spreading of solvents and in the case of heterogeneous porous media heterogeneous conductivity field further enhances mixing and spreading. We express the degree of segregation (or mixing) through the variance of solvent (i.e. surfactant or polymer) concentration field normalized by its maximum value which corresponds to the complete segregated state (i.e. piston-like displacement) as:

$$\sigma_c^2 = \frac{\langle c^2 \rangle - \langle c \rangle^2}{\sigma_{max}^2} \tag{5.8}$$

where  $\sigma_{max}^2 = \langle c \rangle (c_0 - \langle c \rangle)$  and  $c_0$  is the injected concentration of the solvent.  $\sigma_c^2$ varies between 0 and 1. The former indicates the perfectly mixed state where  $c = \langle c \rangle$ and the latter corresponds to when PDF of concentration field is bimodal (either 0 or 1) indicating a perfectly segregated state [200, 265, 266]. Shown in Figure 5.12 is evolution of segregation intensity for three different shock mobility (M<sub>s</sub>) ratios. For the case of M<sub>s</sub> = 2.8, mixing occurs earlier than the other two cases with more stable frontal interfaces. This initial mixing is thus due to viscous fingering on the frontal interface when M<sub>s</sub> = 2.8. After solvent slug is completely injected (square symbol, PV = 0.15) however, viscous fingering on the rear interfaces of cases with M<sub>s</sub> = 1.7 and 0.8 enhances mixing of the solvents rapidly (see the rapid decline of segregation intensity,  $\sigma_{\phi}^2$ ). Thus, despite of more stable frontal interfaces, rear viscous fingering results in more mixing and deterioration of solvent slug at later times.


Fig. 5.12.: Evolution of segregation intensity for three different shock mobility ratios. Different times are marked by symbols including end of solvent injection at PV = 0.15 (square), breakthrough time (circle), and removal time (hexagon).

We now direct our attention to cases with  $M_s = 1.7$  and 0.8. According to results presented in Figure 5.8 discussing the effects of mobility (viscosity ratio) on the recovery for ideal (piston-like or perfectly segregated state) and 2D displacements, the case with  $M_s = 0.8$  results in higher recovery than the case with  $M_s = 1.7$  for ideal displacement. However, this improvement in 2D displacement where hydrodynamic instabilities are taken into account is not observed. This is in fact that due to consistently higher mixing of solvents before breakthrough (marked by circle) for the case of  $M_s = 0.8$  compared to  $M_s = 1.7$ . Thus, although injecting higher concentration of polymer results in more stable frontal interface and results in higher Ca numbers, excessive viscous fingering on the real interface counteracts those favorable effects.

Next, we quantify the spreading of solvents along the mean flow direction caused by viscous fingering in homogeneous media by [267]:

$$\sigma_{xx}^2 = \frac{\langle cx^2 \rangle}{\langle c \rangle} - \left(\frac{\langle cx \rangle}{\langle c \rangle}\right)^2 \tag{5.9}$$

Shown in Figure 5.13 is spreading dynamics of solvents. Before focusing on 2D displacements, it is worth noting that spreading behavior for these three mobility ratios are not quite the same for ideal displacement (i.e. in the absence of viscous fingering). This is due to different wetting phase saturation profiles (note that solvents only exist in the wetting phase) as shown in Figure 5.6. For the case with  $M_s = 2.8$ , varying (maximum at the trailing edge and minimum at the leading edge of the solvent slug) wetting phase saturation coupled with relative permeability causes the mixing zone to expand as solvents move with different velocities [268]. This effect is minimal for the case with  $M_s = 1.7$  and does not exist when  $M_s = 0.8$  as wetting phase saturation remains uniform across the solvent slug and thus solvents throughout the slug move with the same velocity, spreading only due to diffusion. As can be seen, for two



Fig. 5.13.: Spreading of solvents along the mean flow direction for three different shock mobility ratios (open symbols). Different times are marked by vertical lines including end of solvent injection at PV = 0.15 (black dashed) and breakthrough time (colored dashed dot lines). Black solid lines show approximate scaling behavior. Colored solid lines show spreading behavior during the corresponding ideal displacements in the absence of viscous fingering.

types of displacement and all mobility ratios, during the injection period, longitudinal spreading exhibits ballistic scaling,  $\sigma_{xx}^2 \sim t^2$  [269]. However, after complete injection of solvents slug, PV = 0.15, and by start of viscous fingering on the rear interface,

we observe different scaling behaviors. While for the cases with more stable frontal interface we observe superdiffusive scaling ( $\sigma_{xx}^2 \sim t^2$ ), the case with  $M_s = 2.8$  shows Fickian scaling ( $\sigma_{xx}^2 \sim t$ ) which persists until breakthrough. Overall, when compared to their corresponding ideal spreadings, one can see that miscible viscous fingering on the rear interface for  $M_s = 1.7$ , and 0.8 significantly increases spreading of solvents whereas this enhancement of spreading for  $M_s = 2.8$  is moderate.

Due to finite width of our solvent slug and in the presence of instabilities, rear and frontal interfaces may meet each other and start interacting. This could be accurately measured by calculating the interfacial length given by [270, 271]:

$$I(t) = \int_0^{l_x} \int_0^{l_y} \left[ \left( \frac{\partial c}{\partial x} \right)^2 + \left( \frac{\partial c}{\partial y} \right)^2 \right]^{1/2} dx dy$$
(5.10)

Interfacial length in fact measures distortion of the interface due to instabilities and its evolution provides insight about the rate at which small fingers disappear and large fingers grow [216]. Shown in Figure 5.14 is the evolution of interfacial length for three different shock mobility ratios normalized by the injected solvent concentration  $(c_0)$ . Here, viscous fingering is responsible for distortion of the interface and enhancement of mixing of solvents. As can be seen from the Figure 5.14, the interfacial length is consistently higher for  $M_s = 2.8$  while it is the lowest for  $M_s = 0.8$  until breakthrough, indicating poorer mixing in the former compared to the latter. This is also consistent with segregation intensity  $(\sigma_c^2)$  results presented in Figure 5.12. Interfacial length for the least stable frontal interface  $(M_s = 2.8)$  increase the earliest due to viscous fingering on the frontal interface, this happens at a later time for  $M_s = 1.7$  and does not happen for  $M_s = 0.8$  until after solvent injection completes. Interfacial length displays the t<sup>0.75</sup> growth after a period of sharp growth (see point g). Interestingly, this growth rate does not change after point a (end of solvent injection and start of instabilities on the rear interface) as majority of fingers are developing on the frontal interface and thus the growth rate is controlled by those fingers. The same observation can be made for the case with  $M_s = 1.7$  where after a short period of rapid increase



Fig. 5.14.: Evolution of interfacial length for three simulations with  $M_s = 2.8$ , 1.7, and 0.8. End of solvents injection (PV = 0.15) and breakthrough time for each case are marked by square and circle symbols, respectively.

in interfacial length after point b, interfacial length grows with a rate  $\propto t^{0.25}$  until breakthrough, point e. In the case with  $M_s = 0.8$ , interfacial length departs from its almost constant value (note that in the absence of viscous fingering interfacial length increases by time due to diffusion which is negligible here with  $Pe = 10^6$ ) and increases rapidly by the onset of fingering on the rear interface. After this sharp increase in interfacial length (until point h), it shows a complete different scaling behavior than the other two cases by growing at a rate  $\propto t^{-0.25}$ . Point h is the time that rear and frontal interfaces meet and start interacting with each other [233]. This does not happen for the other two cases due to the constant expansion of the mixing zone as fingers are developing on both interfaces. Another important difference between the three cases with different shock mobility ratio is the structure of the fingers on the frontal and rear interfaces. For the case with the least stable frontal interface,  $M_s = 2.8$ , after coalescence of relatively large numbers of fingers at the earlier stages (see Figure 5.14 point a), a few main fingers dominates the flow at later times (see Figure 5.14 point d). However, this is not the case for the other two cases with more stable frontal interfaces. In those cases, solvents seem to be well mixed around the frontal interface and form one dominant finger as wide as the domain height (see Figure 5.14e,f). On the rear interface, for the cases with the higher viscosity contrast (i.e.,  $M_s = 1.7$ , and 0.8) we observe tip splitting and detachments of blobs of solvents whereas in the case with less viscosity contrast ( $M_s = 2.8$ ) these effects are not observed.

#### 5.4.3 Breakthrough and removal

In applications such as enhanced oil recovery or surfactant enhanced aquifer remediation it is critical to have an estimate of breakthrough curves (BTC). BTCs also carry information regarding the nature of the transport in porous media where heavy-tailed curves imply anomalous transport. Here, we look into BTCs (mass flux of solvents at the production well in time) for three shock mobility ratios in homogeneous media to emphasize on the role of viscous fingering alone. As can be seen in Figure 5.15, lower shock mobility ratio (i.e., more unfavorable viscosity ratio on the rear interface) reduces the peak value of mass flux, the same effect caused by medium heterogeneity [272]; in both cases it is due to enhanced mixing and spreading which in our study is significant for  $M_s = 1.7$ , and 0.8. Another effect of enhanced mixing and spreading due to viscous fingering is heavy tailing in the BTCs; as shown in the Figure 5.15, the case with  $M_s = 0.8$  exhibits anomalous behavior with its BTC scaling as  $t^{-1-1.1}$  whereas the transport for the case with  $M_s = 2.8$  is Fickian. It is worth noting that BTCs are highly fluctuating for the cases with  $M_s = 1.7$  and 0.8, indicating that packets of solvents with different concentrations and velocities arrive at the production well (see Figure 5.14), due to the severe viscous fingering. Interestingly, although we observe early break through for the case with  $\rm M_s = 2.8~due$  to unstable shock front and development of long viscous fingers on the frontal interface, BTC for this case does not show fluctuations in later times as in the other two cases,



Fig. 5.15.: Mass flux of the solvent (surfactant) exiting the production well ( $F_{out}$ ) as a function of time for three shock mobility ratios. Inset: BTCs plotted in log-log scale. Solid lines show scaling of the tails of BTCs at late time obtained using maximum-likelihood method (MLE) assuming scaling of form  $t^{-1-\beta}$  where for  $\beta > 2$  transport is Fickian and is called anomalous for  $\beta < 2$ . Our results give  $t^{-1-2.1}$  and  $t^{-1-1.1}$  for  $M_s = 1.7$  and  $M_s = 0.8$ , respectively.

indicating a rather homogeneous concentration field. In Fig. 5.16, we plotted the viscosity profiles at the end of solvents injection at PVI=0.15 and at their corresponding breakthrough. Similar to what is shown before in Fig. 6, at PVI=0.15, as a result of viscous fingering on the leading front we observe smearing of the viscosity profile. The most smearing occurs for the case with  $M_s = 2.8$  and the least for the case with  $M_s = 0.8$ . At breakthrough, however, viscosity profile is smeared on both leading and trailing fronts in all cases with  $M_s = 2.8$  showing the most symmetric profile and the least drop from its nominal viscosity (that is one). For the other two shock mobility ratios the viscous fingering severely decreases the polymer efficacy. In the case with  $M_s = 0.8$  viscosity profile is highly skewed with a long tail exhibiting much lower viscosity than its corresponding nominal value (<< 1). Finally, it is clear that the mixing length increases as the shock mobility ratio decreases from values above one to under one. Next, we look at the effects of heterogeneity on the shape of BTCs for



Fig. 5.16.: Viscosity profiles in a homogeneous permeability field and for three shock mobility ratios  $M_s = 2.8$  and 1.7, and 0.8 from left to right. Viscosity is normalized with maximum achievable viscosity for each case where  $(\mu_w - 1)_{norm} = (\mu_w - 1)/\max(\mu_w - 1)_{ideal}$ .

a fixed shock mobility ratio  $M_s = 1.7$ . As shown in Figure 5.17, even a small amount of heterogeneity reduces the peak mass flux compared to that of homogeneous case. However, at small amount of heterogeneity, late-time tail in BTC is quite similar to the homogeneous case as it is governed by viscous fingering. This changes drastically as strength of heterogeneity increases. Here, the interplay of viscous fingering and conductivity heterogeneity, as shown in Figure 5.17, results in long residence time of solvents in the medium which will be discussed quantitatively next. We now look into



Fig. 5.17.: Mass flux of the solvent (surfactant) exiting the production well as a function of time for homogeneous ( $\sigma_{\ln K}^2 = 0$ ) and three heterogeneous media. Inset: BTCs plotted in log-log scale.

the breakthrough and removal times of solvents as functions of shock mobility ratio and  $\sigma_{\ln K}^2$  in homogeneous and heterogeneous conductivity fields, respectively. We define the breakthrough and removal times when 0.2% and 99.8% of the surfactant mass has exited the production well, respectively. As shown in Figure 5.18a, viscous fingering in homogeneous media affects the breakthrough and removal times of solvents. However, this effect is not significant for breakthrough time and does not vary much with respect to shock mobility ratio (or equivalently viscosity ratio on the rear interface). This is apparently due to the formation of well mixed region near the saturation shock in the cases with  $M_s = 1.7$  and 0.8 where there dose not exist a well defined finger. Nevertheless, removal time is found to be monotonically increasing as shock mobility ratio decreases (i.e., viscosity ratio on the rear interface increases). This is due the significant viscous fingering on the rear interface and lobes of solvent detaching from the solvent slug and thus increasing the removal time. Note that these lobes travel at much slower velocity than their ambient fluid. In the presence of heterogeneity, normalized breakthrough time reduces as  $\sigma_{\ln K}^2$  increases. Additionally, removal time drastically increases due to presence of both viscous fingering and heterogeneity. We previously observed that in the presence of viscous fingering and due to lobe detachments, packets of solvent slowly arrive at the production well. Here, in addition to that mechanism, some solvents with quite lower mobility than their ambient fluid could also flow in regions of low velocity due to heterogeneity in the conductivity field. Combination of these two effects thus yields in very long removal times.

#### 5.5 Conclusion

In this study, we performed high-resolution numerical simulation of two-phase multicomponent flow in porous media and investigated the role of hydrodynamics instabilities and medium heterogeneity on displacement efficiency and fate of solvents. We showed that a finite-size slug of solvents (i.e., surfactant and polymer) often used



Fig. 5.18.: Breakthrough (t<sub>b</sub>) and removal (t<sub>r</sub>) times of solvents. Left: homogeneous media at three different shock mobility ratios normalized by their corresponding breakthrough and removal times in ideal displacements (i.e., no disturbance). Right: heterogeneous media at three different values of  $\sigma_{\rm lnK}^2$  but the same shock mobility ratio  $M_{\rm s} = 1.7$ . Breakthrough and removal times are normalized with those in the homogeneous case ( $\sigma_{\rm lnK}^2 = 0$ ) with the same shock mobility ratio  $M_{\rm s} = 1.7$  to isolate the effects of heterogeneity.

in enhanced oil recovery and groundwater cleanup processes could develop viscous fingering on both frontal and rear interfaces where the latter is controlled by the viscosity ratio whereas the former is controlled by the shock mobility ratio. Although in the absence of any flow disturbances and thus a piston-like displacement more viscous slugs results in better displacement efficiency due to increased Ca number and better sweep, we showed that there exists an optimum viscosity ratio that gives the highest recovery in unstable displacements and increasing viscosity above that point leads to severe viscous fingering on the rear interface which in turn could even reduce the recovery.

We tested the idea of gradually decreasing the viscosity contrast between the ambient fluid (wetting phase) and the finite-size slug of solvents (graded slug) as it is being injected and showed that in fact it is an effective strategy for suppressing development of miscible viscous fingers on the rear interface and could increase the displacement efficiency by around 10%.

To elucidate the low recovery in unstable compared to idea or piston-like (i.e., in the absence of any disturbance) displacements, we studied the evolution of mixing, spreading, and interfacial length and showed that viscous fingering significantly enhances mixing and spreading of solvents and serves as the key mechanism in decreasing the displacement efficiency.

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## 6. CONCLUSION

In this work, we studied flow and transport in porous media from a fundamental and applied point of view. Fundamentally, flow and transport is affected by porestructure at the pore-scale. Theses pore-scale effects then manifest themselves at different macroscopic properties of flow and transport. Using DNS and particle tracking, we studied pore-scale statistics of flow and transport in detail for two cases: (i) Newtonian fluids and (ii) viscoelastic fluids. We showed that the Eulerian velocity field has a broad dynamic range, which reflects the complexity of the pore space. Using particle tracking on three distinct pore-space models, we quantified differences in FPTS, tortuosity, and transport regime in those porous media. Finally, we studied the spatial correlations in the 3-D velocity field by means of two-point correlation functions of velocity fluctuations. We showed that for the relatively homogeneous porous media (overlapping and non-overlapping spheres in 3-D), the two-point correlation function of 3-D velocity field decays similarly to that of pore space and even at long distances displays similar oscillations. We tested the generality of this finding on two more heterogeneous porous media (i.e., unconsolidated sandpack and Castlegate sandstone) in which we again observed a similar behavior, where two-point correlation functions of 3-D pore space and velocity fields were extremely similar. We then moved into the second study, where the background fluid is considered viscoelastic. Fluid viscoelasticity could have drastic effects on the general flow and transport in porous media. We looked into the effect of this change in fluid rheology on long-term particle dispersion and flow resistance compared to the equivalent Newtonian flows in the same porous medium. We found out that for the flow of a FENE-P fluid through porous media, the flow goes through phases of shear-thinning followed by intense shear-thickening, above a critical *De*. Additionally, we demonstrated the onset of fluctuations in the flow by studying the temporal evolution of the pressure gradient.

With regards to particle dispersion, consistent with experimental observations of the same problem, we observed that fluid elasticity does not alter the particle dispersion in disordered porous media in long times where we observed Fickian scaling of MSD in both cases of Newtonian and viscoelastic flows.

In the last two chapters, we looked into multi-phase and multi-component flows and transport at the Darcy scale. This allowed us to look at practical cases where various techniques are used at large scale in processes related to enhanced oil recovery (EOR). In particular, we looked into chemical enhanced oil recovery where chemicals like surfactant and polymer are used to reduce the oil saturation in an already waterflooded reservoir. First, in collaboration with our experimental teams, we successfully designed experiments to model various aspects of processes related to the flow of multi-phase flow in porous as well as those related to the flow of surfactant and polymer. We then presented a comprehensive methodology integrating extensive experimental data required for a successful SP flood modeling. In particular, using the rheological data for polymer, phase behavior tests, capillary desaturation, and polymer RRF experiments, we were able to build an SP flood model via a multi-stage calibration algorithm where the cumulative oil production and pressure profile were successfully history matched. In the end, we showed that our history matched model could successfully predict the new and unseen experiments with the model.

In the last chapter, we drew our attention to the important problem of unstable displacement in porous media. As we saw in our previous work on surfactant-polymer flooding in porous media, mobility control, and the onset of flow instability could drastically change the overall recovery efficiency. Thus, we focused on modeling such instabilities and their role in displacement efficiency and the fate of solvents. We used a finite-size slug of solvents (i.e., surfactant and polymer) to displace a secondary immiscible phase. We showed that depending on the viscosity ratio and shock mobility ratio, viscous fingering could develop on rear and frontal interfaces, respectively. We found out that due to the presence of competing mechanisms, there exists an optimum flow property set where the recovery is maximum. Finally, we quantified the evolution of mixing, spreading, and interfacial length and showed that viscous fingering significantly enhances mixing and spreading of solvents and serves as the key mechanism in decreasing the displacement efficiency. REFERENCES

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# Education

•	Purdue University,	Ph.D. in Mechanical Engineering	2019

- Iowa State University, M.Sc. in Mechanical Engineering 2014
- Sharif University of Technology, B.Sc. in Mechanical Engineering 2012

# **Journal Publications**

- S. Aramideh, P. P. Vlachos, and A. M. Ardekani. "Nanoparticle dispersion in porous media in viscoelastic polymer solutions." Journal of Non-Newtonian Fluid Mechanics, 2019
- P. Naik, P. Pandita, S. Aramideh, I. Bilionis, A. M. Ardekani. "Bayesian model calibration and optimization of surfactant- polymer flooding." Computational Geosciences, 2019
- P. Naik, S. Aramideh, and A. M. Ardekani, "History matching of surfactantpolymer flooding using Polynomial Chaos Expansion." Journal of Petroleum Science and Engineering", 2018
- S. Aramideh, P. P. Vlachos, and A. M. Ardekani, "Unstable displacement of Non-Aqueous Phase Liquids with surfactant and polymer." Transport in Porous Media, 2018
- 5. S. Aramideh, P. P. Vlachos, and A. M. Ardekani, "Pore-scale statistics of flow and transport through porous media." Phys Rev E, 2018
- S. Aramideh, R. Borgohain, P. Naik, C. T. Johnston, P. P. Vlachos, and A. M. Ardekani "Multi-objective history matching of surfactant-polymer flooding." Fuel, 2018
- 7. S. Aramideh, Q. Xiong, S. C. Kong and R. C. Brown, "Numerical simulation of biomass fast pyrolysis in an auger reactor." Fuel 2015
- Q. Xiong, S. Aramideh, A. Passalacqua, S. C. Kong, "Characterizing effects of the shape of screw conveyors in gas-solid fluidized beds using advanced numerical models." J. Heat Transfer, 2014
- Q. Xiong, S. Aramideh, A. Passalacqua, S. C. Kong, "BIOTC: An open-source CFD code for simulating biomass fast pyrolysis." Computer Physics Communications, 2014
- Q. Xiong, S. Aramideh, and S. C. Kong, "Assessment of devolatilization schemes in predicting product yields of biomass fast pyrolysis." Environmental Progress & Sustainable Energy, 2014
- Q. Xiong, S. Aramideh, and S. C. Kong, "Modeling effects of operating conditions on biomass fast pyrolysis in bubbling fluidized bed reactors." Energy & Fuels, 2013

## **Conference** Presentations

- S. Aramideh, A. M. Ardekani, and P. P. Vlachos, "Pore-scale simulation of complex fluids through porous media." APS 2018
- 2. S. Aramideh, T. Guo, A. M. Ardekani, and P. P. Vlachos, "Spatial velocity fluctuations in flow through porous media." APS 2016
- 3. T. Guo, S. Aramideh, A. M. Ardekani, and P. P. Vlachos, "Volumetric microscale particle tracking velocimetry (PTV) in porous media." APS 2016

- 4. S. Aramideh, A. M. Ardekani, I. Bilionis, and P. P. Vlachos, "Stochastic reservoir modeling: Uncertainty analysis of the chemical injection in enhanced oil recovery." Interpore 2016
- Q. Xiong, S. Aramideh, and S. C. Kong, "Modeling Effects of Interphase Transport Coefficients on Biomass Fast Pyrolysis in Bubbling Fluidized Beds." tcbiomass 2013
- S. Aramideh, Q. Xiong, and S. C. Kong, "Modeling Effects of Interphase Transport Coefficients on Biomass Fast Pyrolysis in Bubbling Fluidized Beds." Iowa EPSCoR, 2013