PORE-NETWORK APPROACH FOR CALCULATING THE INTERFACIAL AREA OF WETTING/NON-WETTING PHASES IN POROUS MEDIA

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ABSTRACT

In this study, we are interested in calculating the interfacial area between wetting and non-wetting phases in porous media. We consider simple pore units made up of pores and throats, of different geometries and sizes, including spherical or cubic geometry for the pores and cylindrical or rectangular geometry for the throats. Our calculations demonstrate that the wetting phase that remains behind inside the pores during immiscible displacement, can contribute significantly to the wetting/non-wetting interfacial area. In order to obtain a more complete picture from our study, we introduce additional calculations using the pore-network approach. In a first step, concepts borrowed from Ordinary Percolation Theory are used. In a further step, and in order to study the effect of the history of the fluid displacement (drainage, imbibition, etc.) on the resulting interfacial areas, principals from Invasion Percolation Theory are utilized.

1. INTRODUCTION

In order to better understand and quantify many flow and transport processes in porous media (e.g. soil remediation strategies, reactive transport, CO_2 sequestration, etc.) it is important to acquire a detailed knowledge of fluid-fluid and fluid-solid interfacial areas at the pore-level scale, since they play a key role in the dynamics of multiphase flow and transport in porous media. Fluid-fluid interfacial areas (e.g., between wetting and non-wetting phases) control many mass transfer processes in porous media such as phase partitioning (adsorption) and volatilization as well as colloidal and microbial transport since it has been shown that such interfaces can serve as sorption sites. Fluid-solid interfacial areas (e.g., in partially saturated media) control mineral reaction rates and adsorption phenomena.

The current work is motivated by our interest in carbon dioxide sequestration in geologic formations (e.g. saline aquifers, depleted oil reservoirs, etc.). Carbon dioxide has been identified as a major contributor to the global warming problem. Current estimates of CO_2 in the atmosphere show an increasing trend when compared to the pre-industrial revolution era. Due to the seriousness of the problem and the magnitude of the possible effects on the future climate (and thus on everyday human life), a number of experimental and theoretical studies have been undertaken to investigate possible solutions including the removal and disposal of produced CO_2. Geologic sequestration is one of the considered alternatives.

During CO_2 sequestration in a saline aquifer, the brine/CO_2 interface controls the rate of CO_2 dissolved in the brine phase and the amount of H_2O dissolved in the CO_2 plume (note that if CO_2 is in equilibrium with the brine, solubility controls the amount of CO_2 in the brine---not the interfacial area). The fluid/solid interface controls the dissolution of the porous media and precipitation of reaction products (thus affecting the so-called ionic- and mineral-trapping of CO_2). Carbon dioxide reaction with the host minerals in the subsurface and subsequent precipitation of the solid reactants, results in the "permanent" removal of carbon dioxide, and is considered as the ideal long-term solution. Note, however, that due to slow kinetics, a significant amount of time is likely required for that process to be completed.

From the discussion, so far, it is evident that detailed knowledge of the fluid/fluid interfacial areas within porous media is required. One could use experimental measurements to calculate wetting/non-wetting fluid interfacial areas. However, due to the complicated nature of experimentally measuring the fluid-fluid interface areas it is useful to have a predictive approach as well. The purpose of this study is to perform calculations of fluid-fluid interface areas using simple pore/pore-network approaches and to provide a complementary analysis to experimental results.

This report is organized as follows: We start by giving a brief description of the available literature. We continue with the problem formulation and we also address a number a simple cases that are of interest.

2. BACKGROUND

The interfacial areas between wetting/non-wetting fluids and the solid/wetting fluid inside porous media are important parameters that enter in the continuum scale system of equations that describe the momentum, heat and mass balance of the system. Therefore, a number of methods have been developed for the calculation of interfacial area. The methods can be grouped under two main categories: experimental and theoretical/numerical. In this section we present briefly the two categories in addition to some representative studies from each category. A large number of published works exist for calculating fluid/fluid interfacial areas in porous media. A complete review of all these works is beyond the scope of this work.

In general, there are two types of experimental approaches: those that use the interfacial tracer technique and those that use some variant of X-ray microtomography. The interfacial tracer technique is based on the steady-state effluent breakthrough curves of a surface-reactive and a non-reactive tracer travel time. Variations of the method have been used by Brusseau et al. (1997), Saripalli et al. (1998), Kim et al. (1999), Schaefer et al. (2000), Constanza-Robinson and Brusseau (2003), and Peng and Brusseau (2005). Constanza and Brusseau (2000) presented a critical review of the methodology. The X-ray microtomography is a non-destructive technique that can produce three-dimensional, high-resolution, images of a porous medium. Variations of the method have been used by Wildenschild et al. (2002), Culligan et al. (2004, 2006), and Schnaar and Brusseau (2005).

Computational methods are based on first principles in order to obtain insight into processes controlling interfacial area. Such methods have been reported by Gvirtzman and Roberts (1991) and Dala et al. (2002) as well as Bryant and Johnson (2003, 2004) who used idealized sphere-packings. Reeves and Celia (1996), and Held and Celia (2001), used pore networks. Additional works in this category include those by Cary (1994), and Bradford and Leij (1997).

3. PROBLEM FORMULATION

Single pore and pore-network studies (both experimental and numerical) have been considered over the past years to obtain insight of phenomena occurring at the pore-scale level in porous media (e.g. immiscible or miscible displacements, reactions, phase change, diffusion, phase dissolution, etc.).

Consider the simplest case of a pore unit constructed from a single pore body connected by a number of throats (known as the coordination number, z). Typical examples of spherical and cubic pores are depicted in Fig. 1. Each pore unit can connect to others through the throats and thus form a pore-network as shown in Fig. 2. It is known that when a non-wetting phase displaces a wetting phase inside a restricted medium, most of the wetting phase is removed with the exception of a thin film of wetting phase that remains on the solid phase (Bretherton, 1961). This is demonstrated schematically in Fig. 1a for a spherical pore of radius R_p. A thin, uniform layer of wetting fluid that has thickness equal to $(R_p\text{-}h)$ remains behind. If the pressure in the non-wetting phase keeps increasing, the non-wetting phase eventually can penetrate the narrow throats. This event will occur when the lowest capillary threshold, among all the pore-throat pathways, is exceeded. For the case of the cubic pore, depicted in Fig. 1b, however, the film thickness is not uniform. Most of the wetting phase remains at the corners. Here, we show the case of perfectly wetting liquid phase. When changing the degree of wetting, the amount of liquid at the corners decreases accordingly. The question we try to address at this point is whether the amount of fluid that remains at the corners contributes significantly to the interfacial area of the system. Recall that in a real porous medium randomness and heterogeneity can result in the presence of a large number of corners. Cubic and triangular pores are a simplification to the real problem. Alternatively one could use random packing of spheres, where additional interfacial area is contributed by the formation of pendular rings.

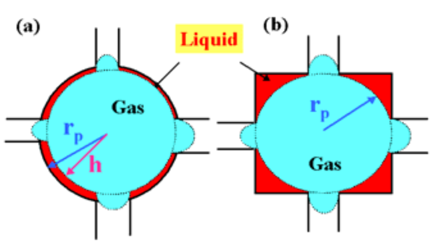

FIGURE 1. Schematic of the 2-D configuration of a wetting (liquid) and non-wetting (gas) phases inside a pore with coordination number $z=4$. **(a)** Spherical pore, and **(b)** cubic pore.

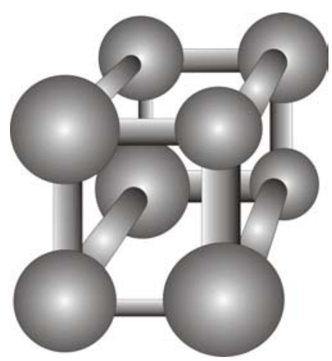

FIGURE 2. Schematic of a 3-D pore network with spherical pores and cylindrical throats with coordination number $z=6$.

In this study we define the two dimensionless numbers, λ and β as follows: $\lambda = h / R_p$, and $\beta = R_t / R_p$, where *R* is the radius and the subscripts *t* and *p* correspond to throat and pore respectively. Fig. 3a shows the dimensionless area S_a as a function of the parameters β and λ for the case of single spherical pore with coordination number z=6. Area S_a was estimated taking into account the remaining wetting films within the pore in addition to the interfacial area inside the throats. Here, we have considered that all throats have the same diameter. That assumption can be easily relaxed. No significant change in the results is expected, however. The surface area was made dimensionless by dividing by the surface area of the pore ($4\pi R_p^2$). The calculation of area S_t takes into account only the interfacial area within the throats (see Fig. 3b). In order to make clear the effect of the interfacial area contributed by the remaining liquid we subtract the calculated surface areas ($S_a - S_t$) and that result is shown in Fig. 4. As can be seen for the case of $\lambda \sim 0.7$-0.8 (which represents a realistic wetting film thickness/pore case), a 70-80% difference in the estimated values of the interfacial areas are observed. We repeated the calculation for the case of a cubic pore (considering a perfectly wetting liquid and λ=1) with results shown in Fig. 5. For the case of β=0.1 we estimate that $S_a \sim 40S_t$, which again is an important difference.

So far, we have discussed the simplest case of an abstract pore unit. Tsimpanogiannis et al. (2005) have discussed the concept of the Elemental Building Block (EBB) which is a regular solid block that has embedded at the centre a pore and a number of throats emanating from it (see Fig. 6 for a schematic). A porous medium consists of an ensemble of EBB's. Pores and throats can be of various geometries and their sizes can follow some given distribution. Consider an EBB that has a unit length (equal in the three directions) that is $L_x = L_y = L_z = 2R_p^{\max}$ and consists of a cubic pore and six equal sized rectangular throats. In Fig. 7 we plot the area ratio (area including contribution of liquid at corners over the area that ignores the liquid at corners) as a function of the dimensionless radius R_p/R_p^{max} and for three different values of β. Once again the importance of liquid at corners becomes obvious.

4. WORK IN PROGRESS

While most of the discussion in the current report evolved around simple systems of pores, the ultimate goal of this effort is to extend the work to pore networks with regular geometry. To this purpose a series of numerical simulations are currently in progress utilizing 3-D pore-networks to calculate the interfacial area between wetting and non-wetting phases in porous media. We are considering regular, cubic lattices of pores and throats that have different geometries and sizes. The pores can have either spherical or cubic geometry and the throats can have cylindrical, rectangular, or triangular geometries. Other geometries are also considered for possible inclusion in our study. The constructed networks can contain one or multiple types of pores/throats and can have variable saturation of wetting and/or non-wetting phases, resulting in different interfacial areas.

Two possible scenarios are examined. First, we examine the simpler case in which different network saturations are obtained by randomly distributing wetting/non-wetting phases in the network based on concepts borrowed from Ordinary Percolation Theory. Next, we consider the more complicated case where we study the effect of the history of the fluid

(a) **(b)**

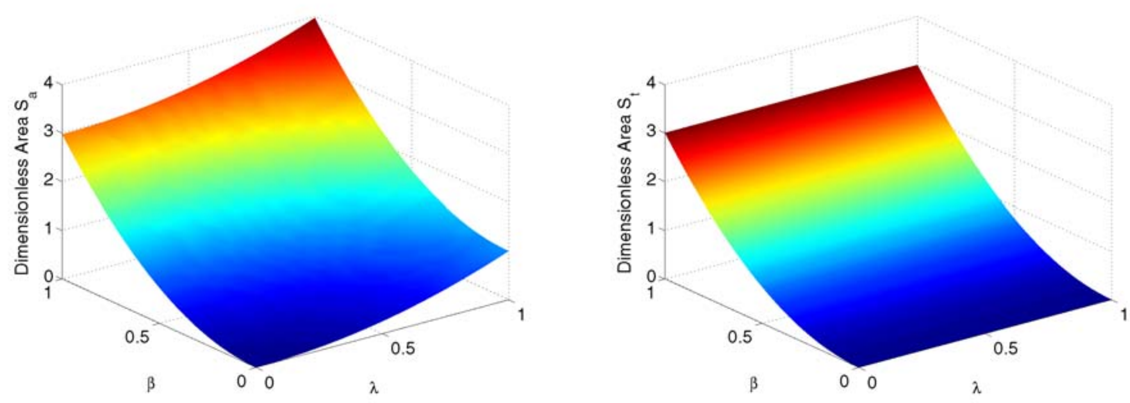

FIGURE 3. Dimensionless Area as a function of the parameters β and λ for the case of single spherical pore with coordination number z=6. **(a)** Remaining wetting films within the pore contribute to the interfacial area in addition to the interfacial area inside the throats (S_a). **(b)** Interfacial area within the throats (S_t).

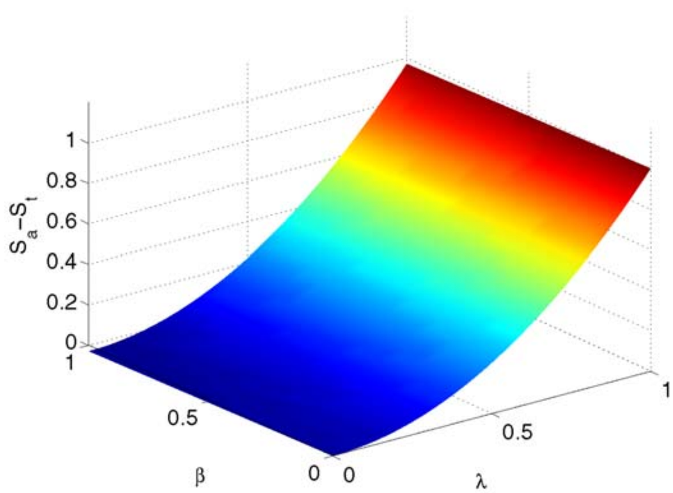

FIGURE 4. Difference between interfacial areas S_a- S_b as a function of the parameters β and λ for the case of single spherical pore with coordination number z=6.

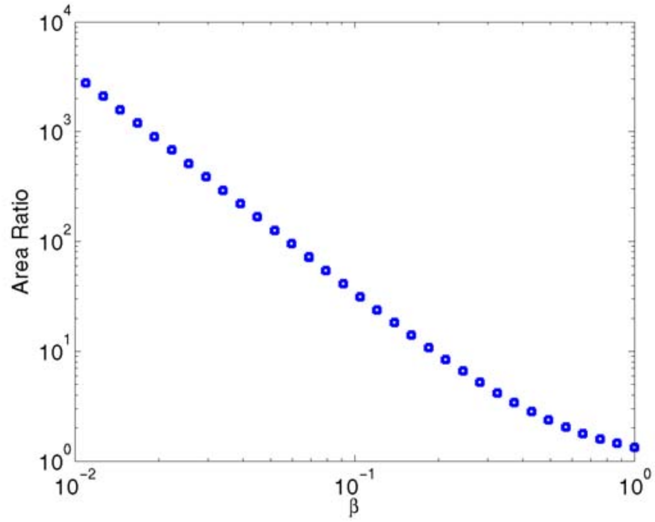

FIGURE 5. Area ratio for a cubic pore with coordination number *z=6*, defined as: The total interfacial area that includes contributions from liquid remaining at the corners of the pore, over the interfacial area only within the throats.

displacement (drainage, imbibition, etc.) on the resulting interfacial areas. These simulations are based on principals from Invasion Percolation Theory.

In addition to obtaining expressions of the interfacial area between wetting/non-wetting phases we are also interested in how the calculated interfacial areas are affected by parameters such as the range and distribution of pore/throat characteristic lengths, the wetting saturation, the pore-size distribution, and the extent of overlapping of pore/throat distributions.

5. SUMMARY

In this work we presented some simple calculations of the interfacial area developed between wetting/non-wetting phases during displacement in porous media. The results presented here are of simple single-pore geometries. We also put forward the foundation of the more elaborate pore-network calculations, which are currently under development. It was demonstrated that an important part of the interfacial area is contributed by the wetting phase that remains after displacement is completed inside the corners of the porous medium. Ignoring that contribution can lead to significant errors in calculating the interfacial area.

We close by pointing out once again that pore/pore-network studies can offer additional guidance to experimental and theoretical work.

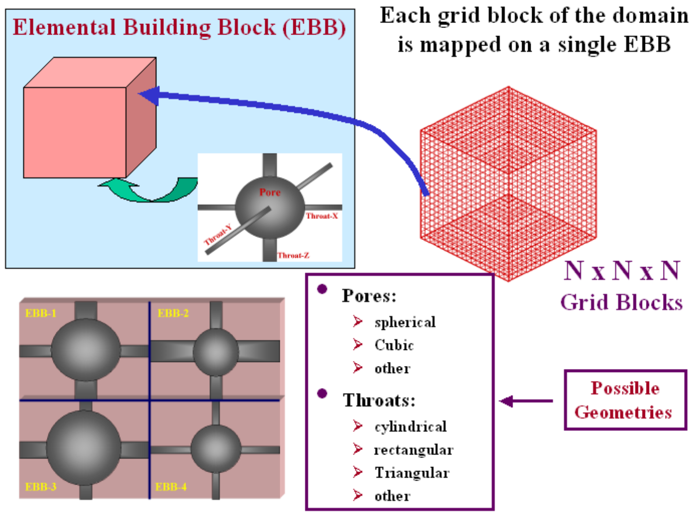

FIGURE 6. Schematic representation of the Elemental Building Block concept. (From Tsimpanogiannis et al., 2005).

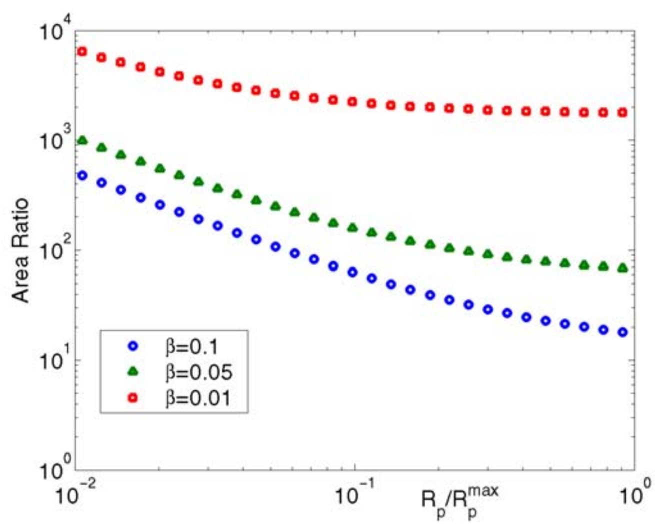

FIGURE 7. Area ratio calculated from a single Elemental Building Block as a function of the dimensionless radius R_p/R_p^{max} for three different values of β.

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