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Research papers

Probing the water distribution in porous model sands with two immiscible fluids: A nuclear magnetic resonance micro-imaging study

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ABSTRACT

The effect of the structural heterogeneity of porous networks on the water distribution in porous media, initially saturated with immiscible fluid followed by increasing durations of water injection, remains one of the important problems in hydrology. The relationship among convergence rates (i.e., the rate of fluid saturation with varying injection time) and the macroscopic properties and structural parameters of porous media have been anticipated. Here, we used nuclear magnetic resonance (NMR) micro-imaging to obtain images (down to ~50 μm resolution) of the distribution of water injected for varying durations into porous networks that were initially saturated with silicone oil. We then established the relationships among the convergence rates, structural parameters, and transport properties of porous networks. The volume fraction of the water phase increases as the water injection duration increases. The 3D images of the water distributions for silica gel samples are similar to those of the glass bead samples. The changes in water saturation (and the accompanying removal of silicone oil) and the variations in the volume fraction, specific surface area, and cube-counting fractal dimension of the water phase fit well with the single-exponential recovery function $\{f(t) = a[1 - \exp(-\lambda t)]\}$. The asymptotic values (a, i.e., saturated value) of the properties of the volume fraction, specific surface area, and cube-counting fractal dimension of the glass bead samples were greater than those for the silica gel samples primarily because of the intrinsic differences in the porous networks and local distribution of the pore size and connectivity. The convergence rates of all of the properties are inversely proportional to the entropy length and permeability. Despite limitations of the current study, such as insufficient resolution and uncertainty for the estimated parameters due to sparsely selected short injection times, the observed trends highlight the first analyses of the cube-counting fractal dimension (and other structural properties) and convergence rates in porous networks consisting of two fluid components. These results indicate that the convergence rates correlate with the geometric factor that characterizes the porous networks and transport property of the porous networks.

1. Introduction

Understanding the nature of interstitial fluid flow through porous networks that contain immiscible liquids (e.g., water and oil) is important in the development of methods for enhanced oil recovery and underground disposal of CO_2 (e.g., Taber, 1980). Probing the distribution of two immiscible fluid phases in natural porous networks characterized by structural heterogeneity can help to clarify the physical properties of complex soil systems and melt–fluid–rock composites in Earth's interior (Gonzalez-Sosa et al., 2010, 2001; Takei, 1998, 2002, 2005). Despite its importance, potential implications, and the extensive efforts of earlier studies, the systematic effect of particle size, heterogeneity, and shape on the microscopic nature of water saturation in porous networks that are initially saturated with an immiscible fluid remains to be explored. The degree of water saturation in porous media can be conveniently described by the convergence rate [i.e., the rate of water saturation in porous media that is initially saturated with immiscible fluid, see Eq. (2) below]. The convergence rate has been correlated with parameters (flow rate, effective capillary pressure, permeability, and entropy length of the porous media) that describe the capillary imbibition process (i.e., the displacement of one fluid by another immiscible fluid) (Babadagli, 2002; Kazemi et al., 1992). However, the relationship between the convergence rate and structural parameters of porous media, as well as the

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effect of the rate of fluid saturation (e.g., oil recovery) on properties such as permeability are also insufficiently understood because of the lack of suitable experimental probes of structural heterogeneity in the networks. The objective of the present study is to use nuclear magnetic resonance (NMR) micro-imaging to obtain images of the water distribution in porous networks that are initially saturated with an immiscible fluid (oil) to quantify the water distribution given varying durations of water injection and to explore the relationships among the fluid saturation rate (i.e., the convergence rate), structural parameters, and the transport properties of porous networks.

NMR micro-imaging is a method to non-destructively probe fluids in complex porous networks (see Lee and Lee, 2013; and references therein). NMR micro-imaging is complementary to conventional X-ray computed tomography (CT), as the latter uses the differences in the fluid electron densities to distinguish distinct phases in porous networks (e.g., Al-Raoush and Willson, 2005; Culligan et al., 2006; Peters and Hardham, 1990; Prodanovic et al., 2006; Schnaar and Brusseau, 2005; Sukop et al., 2008; Wildenscheld et al., 2002). There have been extensive earlier studies that monitored the flow of two or more phases in porous media by means of micro-CT (e.g., Alpak et al., 2016; Khishvand et al., 2016; Song et al., 2016) and synchrotron X-ray tomography (e.g., Pot et al., 2015; Silin et al., 2011). X-ray CT enables shorter experiment times (by several minutes) and higher resolution images (5–10 μm) than NMR micro-imaging, which offers experimental times of a few hours and mm scale resolution. X-ray CT is sensitive to mass density contrast; therefore, it is carried out with large tracer concentrations (Wellington and Vinegar, 1987). In contrast, NMR micro-imaging can image two phases of liquids with similar densities without doping (Chen et al., 2002). A detailed description of these two methods can be found in Blümich (2000) for NMR micro-imaging and Mees (2003) for X-ray imaging. The proton NMR chemical shift (i.e., peak position) depends on the molecular structure around the proton species. Therefore, the NMR micro-imaging technique can be used to distinguish fluid phases by tuning into distinct chemical shifts (Blümich, 2000; Brown et al., 1982; Callaghan, 1991). NMR micro-imaging is sensitive to the chemistry of fluids and has therefore been used to determine the two-dimensional (2D) distribution of diverse fluid phases in porous media that consist of relatively large particles (e.g., Pyrex particles of 420–500 μm) (Chen et al., 1988) and non-aqueous phase liquid (NAPL) in water-saturated columns composed of silica gel (size: 420–500 μm and 850–1000 μm) (Zhang et al., 2002). 3D imaging (resolution: 390 μm × 390 μm × 390 μm and 195 μm × 195 μm × 195 μm) has also been used to show the dissolution of hydrocarbons in packed ballotini (size: 1–5 mm glass beads) (Johns and Gladden, 2000). 3D imaging has been successful in revealing the 3D distribution of NAPLs in a water-saturated dolomite fracture (Becker et al., 2003), the distribution of water and contaminant phases in soil (Simpson et al., 2007), and the distribution of NAPLs during vapor extraction in silica gel columns (Chu et al., 2004), as well as the water and oil phases in glass bead packs (Li et al., 2016), in straight capillaries, bifurcated channels, and monolayered glass bead packs (Liu et al., 2015), and in natural sandstone (Romanenko and Balcom, 2013). Furthermore, the relationships between porous media and two-phase flow were studied to determine the relative permeabilities (i.e., effective permeability divided by absolute permeability) (Bauget et al., 2005; Fourar and Lenormand, 2001). In addition, the effect of distinct surface wettability on the corresponding NMR characteristics including spin-spin relaxation time of fluids in two-phase flow have been theoretically investigated (see Talabi and Blunt, 2010; and references therein).

Earlier advances with NMR micro-imaging for fluids in porous networks hold promise in revealing the effect of structural heterogeneity on the water distribution of porous networks that contain an immiscible fluid with varying durations of water flow (i.e., degree of water saturation in the medium). Despite these advances, these earlier studies focused on the distribution of fluids in relatively large particles (from 400 μm to 5 mm) and with a relatively low resolution (up to ~200 μm). Exploring the distribution of two fluid phases in porous networks using NMR micro-imaging with a resolution of <100 μm allows us to establish the relationships between the convergence rates and macroscopic properties of the porous networks (<100 μm) that make up critical parts of natural aquifers, sediments, and sandstones.

The local structure of amorphous networks on the atomic scale has important implications for its macroscopic properties, such as density, viscosity, diffusivity, configurational entropy, enthalpy, and catalytic ability (Anovitz and Cole, 2015; Lee, 2011; Lee and Ahn, 2014; Lee et al., 2008, 2010a,b, 2016; Navrotsky, 1995). Similarly, progress has been made on the relationships between the local structures of porous networks on the micrometer scale and their corresponding macroscopic properties (e.g., Lee and Lee, 2013). Along with the usual parameters for porous networks, such as porosity and specific surface area, the advances in direct visualization of networks allow us to routinely obtain additional intrinsic parameters for porous networks, such as cube-counting fractal dimension and configurational entropy (Andraud et al., 1994; Mandelbrot, 1982). These parameters have provided insights into the nature and disorder of diverse porous networks (Andraud et al., 1994; Klemm et al., 1997; Müller et al., 1995; Moreau et al., 1999; Perret et al., 2003; Tarquis et al., 2008) (see Section 2.3 and Supplementary information 1 for the detailed definitions of these structural parameters). In particular, previous studies that have used NMR micro-imaging and random packing simulations have shown some relationships among macroscopic properties in porous networks, e.g., the cube-counting fractal dimension of porous networks increases with increasing specific surface area at a constant porosity and with increasing porosity at a constant specific surface area, and the maximum configurational entropy increases with increasing porosity (Lee and Lee, 2013). Additionally, it has been found that the entropy length (i.e., a box size with maximum configurational entropy) provides an accurate measurement of the characteristic size of pores, constituent particles, or both, which constrain the modeled network permeability (Andraud et al., 1997; Lee and Lee, 2013). These newly identified relationships, along with key structural parameters and properties, can be useful in quantifying the fluid network in porous networks that are initially saturated with an immiscible fluid to varying degrees of fluid saturation as the relationships among the convergence rates and microstructural parameters of porous media are not yet currently understood.

In this study, we use 2D and 3D NMR micro-imaging to explore the effects of particle size, shape, and degree of heterogeneity on the distribution of water and the degree of water saturation (convergence rate) in model sands filled with silicone oil. Specifically, we obtain 2D and 3D images of two fluid phases with a resolution below 100 μm, and directly visualize the time-resolved water distribution in various model porous networks to explore the water distribution. We also quantify the degree of water saturation by estimating the network fluid volume, specific surface area, cube-counting fractal dimension, and configurational entropy. Finally, we discuss the relationship between the structural heterogeneity of porous networks, fluid distributions with varying degrees of fluid saturation, and transport properties.

2. Methods

2.1. Materials

We used glass beads (GBs) with spherical grains and crushed silica gel (SG) particles with irregularly shaped grains to