Advances in Water Resources 79 (2015) 35–50

Contents lists available at ScienceDirect

Advances in Water Resources

journal homepage: www.elsevier.com/locate/advwatres

Capillary trapping mechanism in strongly water wet systems: Comparison between experiment and percolation theory

Helmut Geistlinger *, Sadjad Mohammadian

Helmholtz-Centre for Environmental Research – UFZ Leipzig-Halle, Department of Soil Physics, Theodor-Lieser-Str. 4, 06120 Halle/Saale, Germany

ARTICLE INFO

Article history:
Received 6 August 2014
Received in revised form 13 February 2015
Accepted 16 February 2015
Available online 25 February 2015

Keywords:
Capillary trapping
Thin-film flow
Trapped gas cluster
X-ray micro computer tomography
Monte-Carlo experiment
Percolation theory

ABSTRACT

To understand capillary trapping mechanism, we conduct a real Monte-Carlo experiment by using packed glass beads with nearly the same pore size distribution, but different stochastic realizations. We study gas phase trapping during imbibition for capillary numbers from 2×10^{-7} to 10^{-6} using X-ray micro tomography and compare the experimental results with predictions from percolation theory. We found excellent agreement. Percolation theory explains (i) that the capillary desaturation curves are not dependent on flow rate, (ii) the linear dependence of the total gas surface on gas saturation that is a direct consequence of the linear relationship between cluster surface area and cluster volume, which is a prediction from percolation theory for large finite clusters, (iii) the power-like cluster size distribution with an exponent $\tau_{exp} = 2.15$ that only deviates by 2% from the theoretical one ($\tau_{theor} = 2.19$), and (iv) that the maximal z-extension of trapped large gas cluster is described by the cut-off correlation length ξ_B (B – bond number).

1. Introduction

Capillary trapping of gas bubbles, residual NAPL, and oil blobs within water-saturated porous media and mobilization of such trapped, isolated phases is of central importance in many processes in oil recovery, hydrogeology and soil physics. For CCS-technology (CCS – carbon capture and storage) capillary trapping is one of the relevant storage processes [1,2]. To achieve homogeneous distribution of trapped oxygen gas bubbles and/or mobilization of trapped residual NAPL-blobs is still a challenge in remediation of contaminated groundwater [3–6]. The effectiveness of oil recovery will depend on how much isolated oil blobs remain in the porous matrix after water flooding [7,8]. Air entrapment during rain infiltration and groundwater level fluctuation is the key process in the unsaturated zone that determines the gas exchange, redox status, and the aerobic microbial activity of the unsaturated soil zone, especially in the highly transient zone – the capillary fringe [9].

Recently, we studied the capillary trapping mechanism of gas bubbles within 1 mm-glass beads by X-ray micro-computer-tomography experiments (μ-CT) and found no systematic dependency of trapping efficiency on capillary number Ca ($=v \cdot \mu_w/\sigma$, v – mean velocity of the injected fluid, μ_w – viscosity of water, σ – interfacial tension) between 2×10^{-7} and 10^{-6} [10]. To understand this behavior, we reviewed the relevant literature on imbibition process for strongly-water wet systems and for small flow rates, i.e. for capillary numbers $<10^{-6}$.

Since the pioneering work by Lenormand and Zarcone [11,12] the pore-level mechanism of capillary trapping for imbibition was studied intensively by Vizika et al. [13], by Blunt and Scher [14], by Hashemi et al. [15,16], by Constantinides and Payatakes [17], and by Joekar-Niasar et al. [18,19], and by Hilpert et al. [20]; for reviews, see [21–23]. According to experimental observations [12,17] capillary trapping can be caused by different displacement mechanisms: (i) pistonlike displacement with cooperative pore-filling and (ii) by precursor thin-film and corner flow. Sometimes the corner flow is also called thin-film flow in the literature [15,16,23] or thin-film flow and corner flow are summarized as flow in crevices [14]. If not explicitly distinguished, we will use the term thin film flow both for flow along the grain surface and along the corners of the pore space. Which displacement type dominates is strongly dependent on wettability, on flow rate, on pore-throat-geometry, and on pore-network topology (connectivity) [11,12,14].

The temporal sequence of displacement types in a strong-water wet system "glass (SiO_2)–water–air" is governed by the order of the corresponding capillary pressures (see Table 1 in [11]). First, a precursor thin-film in front of the mean wetting front (bulk advance) will "spontaneously" invade the porous media and cover some region. The thickness of such thin water films is often estimated by a typical surface roughness of the order of about

* Corresponding author.

http://dx.doi.org/10.1016/j.advwatres.2015.02.010
0309-1708/

Table 1
Different imbibition mechanisms and its relation to percolation theory proposed by Lenormand and Zarcone [11].

Imbibition mechanism	Percolation type	
	$Ca = 10^{-9}$ large pores	$10^{-9} < Ca < 10^{-6}$ medium-sized pores
Thin-film flow/flow by surface roughness	Bond percolation (ordinary percolation)	Bond percolation + cooperative I1 – pore filling
Flow by corners/crevices	Invasion percolation	Invasion percolation + cooperative I1 – pore filling

10 μm [11,14,17]. However, a more detailed study by Kibbey [24] shows that depending on the surface roughness (sand grains and glass beads were compared) and the surrounding pressure of the nonwetting phase the film thickness can vary from nm- to μm-scale. The spreading of this precursor thin film is always faster than the subsequent bulk advance caused by pistonlike displacement process. Under certain unstable conditions this film can swell and snap-off in the smallest throats in front of the mean displacement front without any "visible" continuity to the bulk wetting phase. This results in *randomly filled throats* (bonds) in order of increasing throat diameter and corresponds to classical *bond percolation* (ordinary percolation).

In case where corner flow is the dominant mechanism, the interface moves by a succession of jumps. All the menisci in pores and along corners are at the same pressure which progressively decreases as the wetting fluid is injected (the volume of fluid increases in the corners, therefore the radius of curvature increases). The jump occurs when the pressure reaches the critical snap-off threshold in the smallest duct at the front. The radius of the swelling corner film starts to decrease once the critical capillary pressure for snap-off (Eq. (3d)) is reached and the gas–water interface becomes unstable. If the interface moves by such snap-off-filling events, this mechanism is similar to an *invasion percolation* mechanism. As the flow rate increases, but Ca is still smaller than 10^{-6}, cooperative $I1$-pore filling mechanism (I1: cooperative pore filling with wetting fluid by $Z-1$ throats, Z – coordination number) also occurs, but the snap-off trapping mechanism remains the dominant mechanism. Possible imbibition mechanisms for the relevant range of capillary numbers are shown in Table 1 that is a section of Table 3 from [11].

Based on the work by Lenormand and Zarcone [11], Blunt and Scher [14] have developed a 3D-pore-level model of wetting that represents these effects (thin-film- and corner flow, cooperative pore filling mechanisms) and discussed possible trapping mechanisms and their dependence on flow rate. We will compare our experimental results with their theoretical results.

With the noninvasive technique of μ-CT and new sophisticated algorithms of image processing [25] the data sets are becoming highly reliable and amenable to cluster analysis at the pore scale. In order to make clear what are the new contributions of our paper, we give a short review of μ-CT studies on non-wetting phase trapping during imbibition, which focus on cluster analysis.

Iglauer et al. [26] analyzed the residual oil cluster size distribution and find consistency with percolation theory that predicts a power-law cluster size distribution, $n(s) \sim s^{-\tau}$, with $\tau = 2.189$ [27], where s denotes the number of sites (=pores) of a trapped cluster. The authors obtained a power-law exponent for the oil-wet core $\tau = 2.12$ (oil–brine–sandstone system, $Ca = 10^{-7}$) which is higher than τ for the water-wet system ($\tau = 2.05$, oil–brine–sandstone system, $Ca = 2 \times 10^{-6}$ [28]) or a CO_2/brine system ($\tau = 2.01$ $scCO_2$–brine–sandstone system, $Ca = 10^{-6}$ [2]). The derived τ-values are in reasonable agreement with the prediction from percolation theory. However, fitting the power law to the data, the voxel number was used instead of the site number, i.e. no renormalization of the $n(s)$-distribution from voxels to pores was carried out. Furthermore, Iglauer et al. [8] analyzed the relationship between the surface area A_i and the volume V_i of individual clusters. They found a power law relationship, $A_i = \text{const}\,(V_i)^p$ with $p \approx 0.8$ based on a best fit using a log–log-plot. An overview of derived τ- and p-values for different multiphase systems is given in [8].

Landry et al. [29] ($Ca \sim 10^{-6}$) studied the microscopic A_i–V_i-relationship and found a similar p-value of about 0.8 based on a log–log plot. Furthermore, they studied the macroscopic relationship between the total surface area (A_{nw}) and the total volume (V_{nw}) of non-wetting phase and found a strong linear relationship (regression coefficient = 0.98).

Georgiadis et al. [30] conducted imbibition and drainage experiments with brine–oil fluid pairs varying interface tension from 9 to 51 mN/m. The capillary numbers were in the range between 10^{-5} and 10^{-4}. Assuming that percolation theory is still valid for these high capillary numbers (compare with Table 1), they fitted their experimental data to the universal power law and could not achieve reasonable agreement, especially in the range of large cluster sizes. Since the universal power law is valid in the limit of large cluster sizes [31], the strong deviations indicate that percolation theory is not applicable and that the data set may be incomplete in this range, because of the too small sample dimensions (10 mm × 20 mm).

Al-Raoush and Willson [32] studied the imbibition process in 0.4–0.6 mm oil-wet glass beads for a brine–oil fluid pair for $Ca = 2 \times 10^{-6}$. The focus of this study was to derive a pore-throat network model with realistic pore- and throat distributions. They obtained a mean pore radius of 58 μm and mean throat radius of 39 μm and a mean coordination number of about 4. They conducted a cluster analysis for the residual brine clusters and found similar results as Geistlinger et al. [10], i.e. the majority of trapped clusters (75%) had a volume less than the average grain volume. About 8% of the blobs are branched, complex in shape, and extend over more than 10 pores. Interestingly is that the cumulative cluster size distribution shows a natural cutoff-length at about five mean grain diameters (=d_{50}). For the individual clusters the authors derived also a shape factor (deviation from spherical shape) and the cluster orientation along the major axis.

The motivation for this paper was initiated from a recently conducted experimental μ-CT-study on capillary trapping mechanisms of gas bubbles within 1 mm-glass beads. We found the following interesting results [10]: (i) no systematic dependency of trapping efficiency on capillary numbers between 2×10^{-7} and 10^{-6}, (ii) the majority of trapped gas bubbles (about 85%) are multipore trapped gas clusters, and (iii) a significant ($R^2 = 0.98$) linear relationship between the total gas surface area and gas saturation. The objective of this paper is to understand these results based on percolation theory, because this is "*...the natural language for describing connectivity effects...*" of fluid phases during displacement of one fluid by another fluid [33]. We focus on the imbibition displacement process for strongly-water wet systems.

Our paper is organized as follows: In Section 2 we give a short overview of the μ-CT experiments. In Section 3 an analytical study of the fluid–fluid displacement process is carried out, and in Section 4 we compare the experimental results with predictions from percolation theory.