



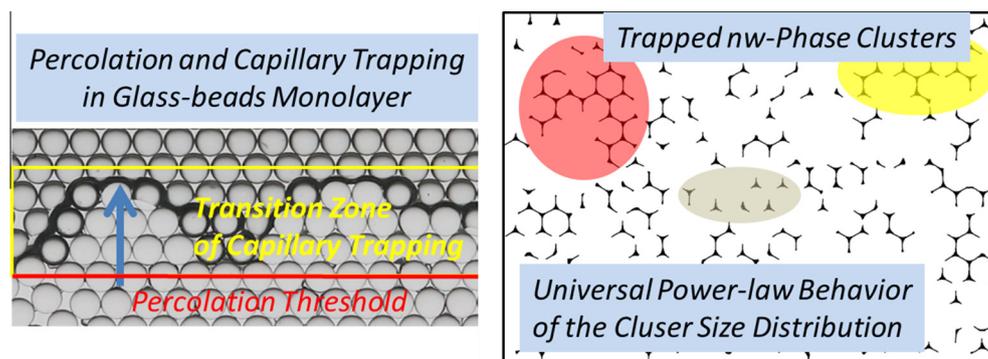
# Influence of the heterogeneous wettability on capillary trapping in glass-beads monolayers: Comparison between experiments and the invasion percolation theory



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## GRAPHICAL ABSTRACT



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## ABSTRACT

We demonstrated that a change in the surface chemistry, i.e., a change from heterogeneous to homogeneous wettability, can dramatically influence capillary trapping, i.e., from significant trapping (~5%) to no trapping. Furthermore, the displacement process (water displaces air) in glass-beads monolayer with heterogeneous wettability shows (i) a heterogeneous morphology and a stochastic advancement of the interface in the highly ordered *triangular* structure, (ii) capillary trapping of a broad variety of gas clusters, notably large ganglia-like and network-like gas clusters, and (iii) a variation in the contact angle between 30° and 100°.

In the second part of this paper, we compared the experimental results of capillary trapping for the monolayer that possesses a heterogeneous wettability with predictions from the invasion percolation theory and found excellent agreement, e.g., that the experimental cluster size distribution can be described by a universal power-law with an averaged exponent  $\tau_{exp} = 2.06$ ; that is a deviation of 5% from the theoretical value. This agreement indicates that capillary trapping within the 2D-monolayer is governed by the 3D critical exponent; therefore, the monolayer shows a trapping behavior similar to a 3D-porous media.

We proposed an analytical approach to calculate the mass transfer rate constant using functional relationships predicted by percolation theory and compare this result with results derived from empirical relationships, which are often used for modelling the dissolution process of trapped non-wetting phases.

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## 1. Introduction

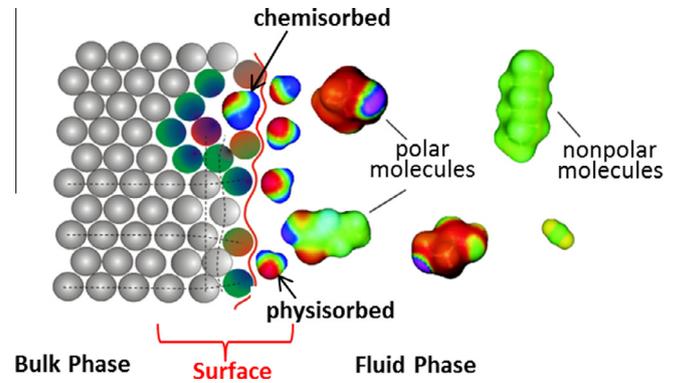
Capillary trapping of gas bubbles, residual NAPL, and oil blobs within water-saturated porous media and the mobilization of such trapped, isolated phases is of central importance to many processes in oil recovery, hydrogeology and soil physics. For CCS-technology (CCS – carbon capture and storage) capillary trapping is a relevant storage process [1,2]. Achieving a homogeneous distribution of trapped oxygen gas bubbles and/or mobilization of the trapped residual NAPL-blobs remains a challenge in the remediation of contaminated groundwater [3–6]. The effectiveness of oil recovery will depend on the quantity of isolated oil blobs remaining in the porous matrix after water flooding [7,8]. Air entrapment during rain infiltration and groundwater level fluctuation are key processes in the unsaturated zone that determines the gas exchange, redox status, and the aerobic microbial activity of the unsaturated soil zone, notably in the highly transient zone – the capillary fringe [9]. In this context of capillary trapping the subsequent dissolution of trapped gas phases is of interest for the bioremediation of contaminated groundwater, for the oxygenation of surface groundwater and for dissolution trapping in CCS-technology. Important open questions addressed in this paper are: (i) how the actual morphology of the trapped gas phase influences the mass transfer rate, and (ii) whether universal scaling laws can describe the distribution of trapped non-wetting phase clusters.

In their pioneering work Chatzis et al. [7] conducted a comprehensive study on residual oil trapping using different porous media (e.g. glass bead packs, 2D-micromodels, sandstones) demonstrating the effect of pore structure, topology and heterogeneity on capillary trapping. This work has initiated numerous studies on capillary trapping using more advanced visualization techniques (X-ray micro-tomography ( $\mu$ -CT) (Wildenschild and Sheppard [10], Blunt et al. [11]; Werth et al. [12]; Schlüter et al. [13]); scanning probe microscopy (Wiesendanger [14]); and optical imaging (Werth et al. [12]; Herman and Lemasters [15]; magnetic resonance imaging (Ronen et al. [16])).

According to recent experimental observations ([8,17,18], see references in [19]), capillary trapping is strongly dependent on the *wettability* and *roughness* of the pore–solid interface. By-pass trapping and snap-off trapping are the primary trapping mechanisms in natural porous media (e.g., sands, sandstones, soils). Precursor thin film flow can cause snap-off in front of the main bulk advancement and lead to an efficient capillary trapping mechanism of residual gas phases during imbibition (Lenormand and Zarcone [20]; Constantinides and Payatakes [21]).

Natural rocks exhibit a *heterogeneous wettability*, because of their heterogeneous mineral composition [22] and their heterogeneous distributed adsorbates [23–26,18]. Such heterogeneous adsorbates are caused by the actual heterogeneous chemical composition (distribution of reactive sites) of a real solid surface containing atomic defects, faults, edges and steps. The adsorbed molecules consist of physisorbed and chemisorbed polar molecules (see Fig. 1) and in the case of a liquid fluid phase also of strongly bounded ions. Kim et al. [27] discussed the main chemical groups on silica surfaces and the corresponding mechanisms that are responsible for altering the wettability.

Following de Gennes [28], the partial wetting on “high-energy” surfaces of “hard” solids (covalent, ionic or metallic binding) is driven by a positive spreading parameter  $S$  ( $S = \gamma_{SV} - (\gamma_{SL} + \gamma_{LV})$ , where  $\gamma$  denotes the free energy per area of the solid/vapor (S/V), solid/liquid (S/L), and liquid/vapor (L/V) interface, also known as the surface tension). When  $S = 0$ , the glass (silica) surface is completely wetted. However, as de Gennes [28] emphasized, solid surfaces under ambient conditions are neither “dry” nor “clean”,



**Fig. 1.** Real solid surface with faults and defects (dashed lines) and physisorbed and chemisorbed polar molecules present in the fluid phase.

i.e. under ambient conditions, an oxidic surface is covered by OH-groups, water molecules, CO<sub>2</sub>-molecules, oxygen and nitrogen molecules. Thus, dipole–dipole and ion–ion interactions determine the interfacial energies, e.g. the non-ideal behavior of CO<sub>2</sub>-gas under high pressures [29] and the pH-dependent surface charge on solid surfaces [30]. de Gennes [28] introduced the term “moist” solid surface, emphasizing the fact that the chemisorbed molecular surface layer will determine the actual (heterogeneous) chemical composition of the surface and can cause a significant change in the adsorbed dipole moment, surface charge, wetting behavior and contact angle. The spreading parameter is determined by a complex interplay of different – sometimes competing – processes; e.g. dissolved ions will linearly increase the liquid–vapor interfacial tension  $\gamma_{LV}$  [31] and decrease the solid–liquid interaction energy,  $\gamma_{SL}$  (increase of the binding energy,  $|\gamma_{SL}|$  [30]). When the decrease of  $\gamma_{SL}$  is larger than the increase of  $\gamma_{LV}$ , the spreading parameter increases, indicating more wetting and a smaller contact angle.

The *first objective* of this paper is to study the capillary trapping dynamics for a 2D-monolayer with both homogenous and heterogeneous wettability. The 2D-glass-beads monolayers show a periodic *triangular* structure similar to that of the 2D-micromodel used by Kim et al. [27].

The *second objective* of this paper is to study the morphology and topology of the trapped gas phase conducting a detailed cluster analysis and comparing the experimental results to theoretical predictions from invasion percolation theory [19]. In particular, we are interested in determining reliable estimates for the cluster size distribution,  $PDF(s)$ , and the surface–volume relationship  $A_g^i = f(V_g^i) = f(s)$  of the  $i$ -th trapped gas cluster that contains  $s$  pores (sites). Neglecting the throat volume the number of pores provides the cluster volume:  $V_g^i \cong s$ . Both functions are crucial for modelling the dissolution process of a trapped non-wetting phase. In general, the effective dissolution rate constant,  $k_{eff}$  (1/s), is given by a multi-rate expression describing the dissolution from trapped NAPL-blobs or trapped gas bubbles of different sizes and shapes [32–35]:

$$k_{eff} = \frac{1}{V_b} \sum_{i=1}^N \beta_i A_g^i = \frac{1}{V_b} \int_{s_{min}}^{s_{max}} ds \cdot \beta(s) f(s) PDF(s) = \bar{\beta} \cdot a_g = \bar{\beta} \cdot 2.6 S_g / d_{50} \quad (1)$$

where  $\beta_i$  (m/s) is the mass transfer coefficient of an individual cluster,  $\beta(s)$  (m/s) is the mass transfer coefficient of a certain class of trapped clusters containing  $s$  pores,  $V_b$  (m<sup>3</sup>) is the bulk volume,  $a_g$  (m<sup>2</sup>/m<sup>3</sup>) is the specific surface of the trapped non-wetting phase,  $S_g$  (–) is the gas saturation, and  $d_{50}$  (m) is the median grain diameter. The *analytical approximation* (second equality in Eq. (1)) replaces the

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