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## Efficiently engineering pore-scale processes: The role of force dominance and topology during nonwetting phase trapping in porous media



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#### ABSTRACT

We investigate trapping of a nonwetting (NW) phase, air, within Bentheimer sandstone cores during drainage-imbibition flow experiments, as quantified on a three dimensional (3D) pore-scale basis via x-ray computed microtomography (X-ray CMT). The wetting (W) fluid in these experiments was deionized water doped with potassium iodide (1:6 by weight). We interpret these experiments based on the capillary-viscosity-gravity force dominance exhibited by the Bentheimer-air-brine system and compare to a wide range of previous drainage-imbibition experiments in different media and with different fluids. From this analysis, we conclude that viscous and capillary forces dominate in the Bentheimer-air-brine system as well as in the Bentheimer-supercritical  $CO_2$ -brine system. In addition, we further develop the relationship between initial (post-drainage) NW phase connectivity and residual (post-imbibition) trapped NW phase saturation, while also taking into account initial NW phase saturation and imbibition capillary number. We quantify NW phase connectivity via a topological measure as well as by a statistical percolation metric. These metrics are evaluated for their utility and appropriateness in quantifying NW phase connectivity within porous media. Here, we find that there is a linear relationship between initial NW phase connectivity (as quantified by the normalized Euler number,  $\hat{\chi}$ ) and capillary trapping efficiency; for a given imbibition capillary number, capillary trapping efficiency (residual NW phase saturation normalized by initial NW phase saturation) can decrease by up to 60% as initial NW phase connectivity increases from low connectivity ( $\hat{\chi} \approx 0$ ) to very high connectivity ( $\hat{\chi} \approx 1$ ). We propose that multiphase fluid-porous medium systems can be efficiently engineered to achieve a desired residual state (optimal NW phase saturation) by considering the dominant forces at play in the system along with the impacts of NW phase topology within the porous media, and we illustrate these concepts by considering supercritical CO<sub>2</sub> sequestration scenarios.

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#### 1. Introduction

During immiscible multiphase flow, drainage is the process of nonwetting (NW) fluid invading the pore space and displacing wetting (W) fluid, and imbibition is the process of W fluid invading the pore space and displacing NW fluid. Many engineered processes in the subsurface require fundamental understanding of drainage and imbibition processes and the resulting NW phase capillary trapping; for example, during oil recovery operations, remediation of non-aqueous phase liquid (NAPL) contaminants in the subsurface, and geologic sequestration of carbon dioxide ( $CO_2$ ). In these examples, water is generally assumed to be the W phase; while oil, NAPL, or  $CO_2$  are considered to be the NW phase. Although there are abundant exceptions to this assumption (e.g.

intermediate-wet or oil-wet media), in this work we refer to water as the W phase for simplicity and consistency with previous work. In oil recovery and NAPL remediation processes the system already exists in the initial (post-drainage) state, i.e. both water and NW fluid (oil or NAPL) are already present in the system, and the bulk of previous research has focused on how to alter the imbibition (water flood) process to mobilize the maximum amount of trapped NW phase. Geologic CO<sub>2</sub> sequestration differs from oil recovery and NAPL remediation processes because both the drainage (i.e. CO<sub>2</sub> injection) and imbibition (i.e. water chase or groundwater flow) processes may be engineered, and the overall aim of the process is to trap, rather than mobilize, NW phase in the pore space of the geologic medium. From a CO<sub>2</sub> sequestration perspective, maximizing residual NW phase saturation is optimal; this is in comparison to oil recovery or remediation processes, in which minimal NW phase is the optimal residual state. Comparison



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between different studies and applications is complicated because fluid properties (e.g. density, viscosity, interfacial tension), medium properties (grain and pore size distributions, pore space connectivity, etc.), and flow properties (engineered injection or natural groundwater flow rates) vary widely depending on the application in question.

Recent focus on CO<sub>2</sub> sequestration as a global climate change mitigation strategy has prompted renewed study into the phenomena of capillary trapping in porous media (e.g. Juanes et al. [1], Al Mansoori et al. [2], Akbarabadi and Piri [3], Andrew et al. [4], among others). Capillary trapping of CO<sub>2</sub>, wherein CO<sub>2</sub> is trapped by capillary forces in the pore bodies of a porous medium, is a relatively secure form of trapping, as compared to "structural" or "hydrodynamic" trapping, where the buoyant CO<sub>2</sub> is contained by an impermeable caprock; in addition, capillary trapping occurs on relatively short timescales, as compared to dissolution trapping or mineral trapping [5]. Thus, maximization of capillary trapping is important to ensure a safe and stable geologic carbon sequestration operation. Previous work has investigated NW phase capillary trapping over a wide range of fluid pairs, media types, and flow conditions for oil recovery and NAPL remediation applications. The results of these studies could greatly enhance fundamental knowledge that would benefit CO<sub>2</sub> sequestration studies if the results are appropriately compared and characterized. CO<sub>2</sub> is in a supercritical state at sequestration reservoir operations; relatively small variations in pressure, temperature, and salinity of the W phase have significant effects on the CO<sub>2</sub> density, CO<sub>2</sub> viscosity, and interfacial tension exhibited by the supercritical  $CO_2$ -brine system [6–8]. The variability of fluid properties complicates predictions of flow patterns and displacement mechanisms. Further, it has been shown that the initial (post-drainage) state of the reservoir has a substantial impact on the amount of residual capillary trapped NW phase present in the system after imbibition [3,9]. However, experimental work with supercritical fluids is a non-trivial exercise, necessitating methods to approximate supercritical conditions with ambient experiments; Herring et al. [10] demonstrated that ambient condition micromodels were able to accurately predict the flow regime for a supercritical CO<sub>2</sub> drainage process in sandstone cores through application of a dimensional analysis.

We explore capillary trapping of NW phase from two perspectives. First, we investigate how consideration of the dominant forces at play via the pore-scale force balance in a system allows for comparison and better understanding of results from a wide range of experiments. The second study area results from the unique ability to engineer the drainage (CO<sub>2</sub> injection) process, and thus the initial (i.e. post-drainage) state. To this end, we use three dimensional (3D) metrics which can be used to describe NW fluid topology (connectivity) within porous media. In particular, we develop the relationship between initial NW fluid connectivity and the residual state of a system which was first investigated by Herring et al. [9], with new analysis of a high quality data set collected via X-ray computed microtomography (X-ray CMT) experiments performed at the Advanced Photon Source at Argonne National Laboratory. Finally, we provide physical interpretation of these relationships.

Consideration of pore scale forces and fluid topology allow drainage and imbibition processes to be efficiently engineered to provide favorable residual conditions. Geologic  $CO_2$  sequestration scenarios are used as an example to illustrate these concepts.

#### 2. Background

#### 2.1. Pore scale forces

Considerable previous research has been conducted on the topic of residual (i.e. capillary trapped) NW saturation in porous media, with many different W and NW fluids, and in different media. To facilitate characterization of fluid flow over a range of properties, the experiments discussed in this work are characterized by the dimensionless ratios of capillary number (Ca) and Bond number (Bo).

Ca has been presented in several different forms, and has traditionally been defined from an oil extraction standpoint [11–13]. In general, Ca describes the balance between viscous forces and capillary forces with respect to the invading fluid (as opposed to the defending fluid). In this work, Ca is defined as:

$$Ca = \frac{Viscous \ Force}{Capillary \ Force} = \frac{\mu_{INV} \nu_{INV}}{\sigma} = \frac{\mu_{INV} \frac{Q_{INV}}{A \eta}}{\sigma}$$
(1)

where  $\mu_{INV}$  is the invading phase viscosity [mPa·s], [mPa s],  $v_{INV}$  is the invading phase Darcy velocity [m/s], and  $\sigma$  is the interfacial tension [mN/m] between the invading and defending fluid. The invading phase velocity is computed as the volumetric flow rate Q [m<sup>3</sup>/s] divided by the cross-sectional area A [m<sup>2</sup>] of the porous medium and the porosity  $\eta$  [–]. Note that while Ca is defined as a function of the invading fluid, and is thus dependent upon whether the system is undergoing imbibition (brine is the invading phase) or drainage (CO<sub>2</sub> is the invading phase).

Previous work has shown that under certain conditions, the Ca of the imbibition process determines the trapped NW saturation of the post-imbibition, residual state [11,14,15]. When a relationship between imbibition Ca and residual NW saturation is apparent, it is described by a constant residual saturation value for low imbibition Ca values with a sharp decrease in residual saturation as Ca of the imbibition process increases above the threshold or "critical" capillary number [13,14]; this is shown in Fig. 1 (modified from Cense and Berg [13]).

This relationship between Ca and NW saturation post-imbibition has been shown during NW desaturation experiments wherein the initial-state system (NW fluid is present in the medium) is water-flooded at increasing flow rates and the NW phase saturation in the core is measured as a function of Ca [13–16]; it is shown in sets of drainage–imbibition experiments conducted at different imbibition Ca [9,15] and has also been supported by modeling results [17,18]. However, Ca is not a complete descriptor of trapping in a porous medium, because the curves describing the dependence of residual saturation on Ca can shift due to changes in NW fluid properties [19] or medium properties [18]. Additionally, other experimental studies in different systems have found reduced or no dependence on Ca [20–22].



**Fig. 1.** Capillary trapped residual nonwetting (NW) phase saturation ( $S_R$ ) as a function of imbibition capillary number ( $Ca_{imbib}$ ), modified from Cense and Berg [1].

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