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Automatic method for estimation of *in situ* effective contact angle from X-ray micro tomography images of two-phase flow in porous media

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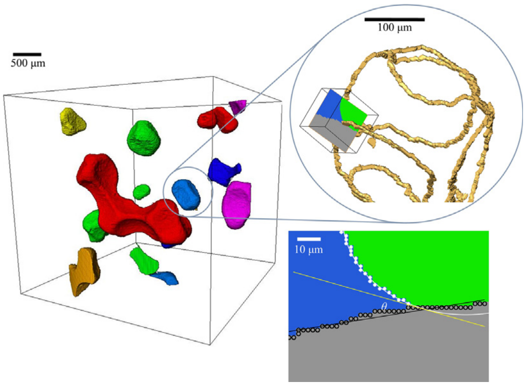
GRAPHICAL ABSTRACT



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ABSTRACT

Multiphase flow in porous media is strongly influenced by the wettability of the system, which affects the arrangement of the interfaces of different phases residing in the pores. We present a method for estimating the effective contact angle, which quantifies the wettability and controls the local capillary pressure within the complex pore space of natural rock samples, based on the physical constraint of constant curvature of the interface between two fluids. This algorithm is able to extract a large number of measurements from a single rock core, resulting in a characteristic distribution of effective *in situ* contact angle for the system, that is modelled as a truncated Gaussian probability density distribution. The method is first validated on synthetic images, where the exact angle is known analytically; then the results obtained from measurements within the pore space of rock samples imaged at a resolution of a few microns are compared to direct manual assessment. Finally the method is applied to X-ray micro computed tomography (micro-CT) scans of two Ketton cores after waterflooding, that display water-wet and mixed-wet behaviour. The resulting distribution of *in situ* contact angles is characterized in terms of a mixture of truncated Gaussian densities.

1. Introduction

Multiphase flow in natural or engineered porous materials is ubiquitous in modern applications involving e.g., chemical transport, geochemical reactions, seawater intrusion, gas diffusion in fuel cells, conventional and unconventional oil recovery, and

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Abbreviations: micro-CT, micro computed tomography; 2D, 3D, two-three dimensional; USBM, United States Bureau of Mines; SEM, ESEM, scanning electron microscopy, environmental SEM; ML, maximum likelihood; RMSE/RMSD, root mean square error/difference; ROI, region of interest.

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carbon dioxide capture and storage [1–8]. The pore scale arrangement of multiple fluid phases in such porous materials is controlled by the topological and geometrical characteristics of the pore space and the wettability of the system. Wettability is represented through the spatial distribution of contact angle at the three-phase contact between two residing fluids and the host solid matrix, and its *in situ* characterization in complex pore spaces is still an open challenge. The arrangement of fluids in a porous medium is governed by the Young-Laplace equation, which stems from energy balance considerations [1] and defines the equilibrium capillary pressure P_c, related to the interfacial tension γ and the principal curvature radii (r_1 and r_2) of the interface:

$$P_c = \gamma\left(\frac{1}{r_1}+\frac{1}{r_2}\right) = \kappa\gamma, \tag{1}$$

where κ is the total curvature of the interface. It can be noted that P_c, and hence the curvature, is constant when the system is at equilibrium, i.e., the fluids are at rest: this will be the key insight used in our approach to characterize the wettability.

The contact angle θ between two-phases (w and n) is related to the interfacial tension between the two fluids through the Young equation [9]:

$$\gamma_{s-n} = \gamma_{s-w} + \gamma_{n-w}\cos\theta, \tag{2}$$

where γ_{s-i}, ($i = n, w$) and γ_{n-w} respectively are the interfacial tension between the solid surface and the two fluid phases, and the interfacial tension between the two fluid phases themselves. For example, in a water–oil system, wettability is classified on the basis of the contact angle, as measured through the water phase. When considering the flow of water and oil in a porous medium, water- and oil-wet systems are characterized by values of θ which are respectively less than or greater than 90° [10].

A series of studies have documented that the spatial distribution of wettability - the pore-scale distribution of contact angle - strongly affects fluid displacement and recovery in rocks [10–19] as well as the performance of fuel cells [20,21]. For example, fluid snap-off is enhanced in water-wet systems and causes high residual saturation of the non-wetting phase [1,2,22–24]. As another example, the wettability influences the relationship between capillary pressure and liquid saturation which in turn governs gas diffusion in the porous electrodes of fuel cells [7,25].

Measurements of contact angles have been typically performed on flat surfaces at ambient conditions through sessile drop and/or captive bubble approaches [11,13,26–28], for different pressure and temperature conditions [29]. In this context, micro-CT imaging has been used to observe drops of fluid [30]. Bachmann et al. [31] modified the sessile drop method for the assessment of the contact angle of powered or granular material. While these methods yield valuable information, they do not yield indication about the *in situ* contact angle within a porous medium, as roughness of the rock surface and the irregular shape of pores are not taken into account. Another key drawback of these approaches is that they usually consider pure mineral surfaces. The ensuing contact angle estimates are then seldom transferable to settings typical of engineering applications, such as hydrocarbon reservoirs, where the porous media have a mix of mineralogy and small-scale pore texture and morphology.

In current petroleum engineering practice the contact angle, or wettability, is indirectly inferred from capillary pressure curves [14], through the Amott or USBM (United States Bureau of Mines) indices [32,33]. These are also used for the characterization of gas diffusion of fuel cells [7]. Other indirect approaches are documented in the literature, such as the Wilhelmy plate method [34], the water drop penetration test [35] and the capillary rise method [36]. These have been applied on different granular soils to obtain estimates of receding and advancing contact angles, without the direct visualization of the microscopic interfaces [37]. All the indirect methods provide an appraisal of the average behaviour of macroscopic samples without providing insights on the distribution of local contact angles.

Scanning electron microscopy (SEM) methods can also be applied to study contact angles between fluids and a surface at high resolution [38]. Cryo-SEM methods establish a representative *in situ* distribution of fluids, and then freeze and cleave the sample followed by imaging to determine contact angle [38,39]. However, the freezing process might alter the contact line between the phases: the method does not therefore directly yield contact angle *in situ* at the conditions under which the fluids flow in reservoir settings. Environmental SEM (ESEM) methods can study contact angles under a range of temperature and pressure conditions [40], but again do not directly observe these angles during displacement [40,41]. Instead what is needed is a way to observe the contact line inside a rock at representative conditions during, or at the end of, a multiphase displacement.

Knowledge of effective *in situ* contact angle is needed for pore-scale models of multiphase fluid flow to quantify the fluid configurations and threshold capillary pressures. Averaged quantities, such as relative permeability and capillary pressure can than be predicted using pore-scale modelling - see, for instance [42–44]. However, at present - in the absence of direct measurements - a distribution of contact angle is simply guessed, possibly to match measured capillary pressure or residual saturation, which adds a significant uncertainty to the predictions of these pore-scale models.

The advent of pore-scale imaging has allowed the determination of contact angle *in situ* from imaged rocks with a micron-scale resolution. This information can constitute a critical input to advanced direct pore-scale numerical modelling of multiphase flow because it can account for the effect on rock wettability of solid matrix roughness and mineralogical composition. Andrew et al. [45] were the first to illustrate a procedure to perform such direct measurements of rock wettability for a supercritical CO_2/brine system. This approach has been applied to different fluid systems in porous media by several authors [42,46–49] and here it will be used for comparison with the new proposed algorithm.

The approach of Andrew et al. [45] is based on a manual measure of contact angles. As such, it might be prone to bias/subjectivity and is time-consuming, thus hampering the possibility of acquiring extensive data sets to capture spatial distributions of contact angles. Khishvand et al. [46] applied this method on segmented images to estimate advancing and receding contact angles under two- and three-phase flow conditions. Klise et al. [50] proposed an automatic method, which was tested only on simple settings: bead packs comprised of beads of one or two uniform wettabilities. Their method relies on the availability of accurate imaging of the three-phase (e.g., water–oil–solid) contact line, a feature which is very rarely available due to the difficulty of obtaining a clear and accurate discrimination of the three phases in realistic rocks where there is inevitable trade-off between resolution and system size.

To overcome the challenge posed by the need to accurately discriminate each phase at the three-phase contact between two fluids and the solid, we propose an approach which is grounded on the physical insight that the fluid–fluid interface has a constant curvature. This will enable us to infer the location of the interface as well as the contact angle with the solid without the need to ground the entire analysis on very precise observations of the fluid–fluid interface.

The estimated angle is *effective* in the sense that it is the angle that the fluid–fluid interface would have in the case of a locally