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In-situ characterization of wettability and pore-scale displacements during two- and three-phase flow in natural porous media

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ABSTRACT

We establish a unique approach to measure in-situ contact angle from micro-CT images acquired during two- and three-phase miniature core-flooding experiments in order to overcome the uncertainties associated with conventional contact angle measurement techniques. The measurements are used to quantify the wettability behavior of the rock and explain pore-level displacement events occurring in three-phase flow. Six two-phase experiments are performed on individual core samples with three pairs of fluids, i.e., oil-brine, gas-oil, and gas-brine, and under two thermodynamic conditions: (a) binary-equilibrated, when only the two respective phases are at equilibrium and (b) ternary-equilibrated, when all three phases are equilibrated and only the two desired fluids are injected into the core. A three-phase experiment set is also performed under ternary-equilibrated conditions, which includes gas injection, a waterflood, and an oilflood process. All experiments are performed on Berea miniature core samples using a nonspreading brine-oil-gas fluid system.

We measure receding and advancing contact angles at arc menisci and main terminal menisci for the two-phase binary-equilibrated experiments and characterize contact angle hysteresis for each fluid pair. Contact angle hysteresis values are almost identical for all fluid pairs. The results of the two-phase binary- and ternary-equilibrated experiments show similar contact angle distributions for each fluid pair. Contact angle distributions during the three-phase flow experiment are analyzed to develop new insights into relevant complex displacement mechanisms. The results indicate that, during gas injection, the majority of displacements involving oil and water are oil-to-water events. It is observed that, during the waterflood, both oil-to-gas and gas-to-oil displacement events take place. However, the relative frequency of the former is greater. For the oilflood, gas-water interfaces only slightly hinge in pore elements. Pore-scale fluid occupancy maps and the Bartell–Osterhoff constraint verify the above-mentioned findings.

1. Introduction

Among all factors affecting multiphase flow in porous media, wettability undoubtedly plays a key role. It is a manifestation of complex rock-fluid interactions and has a significant impact on the microscopic distribution of fluid phases and fluid-fluid interface movement in the pore space. Wettability substantially influences multiphase flow properties, such as remaining oil saturation, relative permeability, and capillary pressure (Alizadeh and Piri, 2014b; Anderson, 1986a; Hirasaki, 1991; Morrow, 1990; Morrow and Nguyen, 1982; Zhou and Blunt, 1998). The fact that wettability of oil reservoirs can fall into a broad wetting spectrum ranging from strongly water-wet to strongly oil-wet (Cuiec, 1984; Treiber et al., 1972) motivated researchers to quantify this surface property through some measurable parameters, such as contact angle. Contact angle is the most universal and accepted measure of the wettability of a surface (Morrow, 1990) and is determined at the contact line where three phases (i.e., two fluids and the solid) meet. By convention, it is measured through the denser fluid.

Although various techniques, such as sessile drop, captive bubble, and dynamic Wilhelmy plate (Buckley, 2001), have been proposed over the years to measure the contact angle, they all suffer from some drawbacks. Perhaps, the main common drawback of these methods is the selection of the solid surface. This arises from the fact that all contact angle measurements, even at reservoir conditions and with reservoir fluids, are made on smooth, flat, and pure surfaces (Anderson, 1986b). While the selection of the substrate is based on the predominant mineral component of the reservoir rock, e.g., quartz as a representative of sandstones, this approach may not consider the mineral heterogeneities observed in the pore space even in a single pore element. In other

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Nomenclature

C_{so}	spreading coefficient of oil on water in the presence of gas
FOV	field of view
IFT	interfacial tension
K	permeability
SGI	secondary gas injection
SP	saturation point

Greek letters

γ	interfacial tension
θ	contact angle
ϕ	porosity

Subscripts

abs	absolute
a	advancing
g	gas
h	hinging
o	oil
r	receding
w	water (brine)

Superscripts

AM	arc meniscus
eq	equilibrium
i	initial
MTM	main terminal meniscus

words, in mineralogically complex systems such as natural rocks, the minerals lining the pore walls and their distributions may vary from one location to another, leading to different wetting properties (Morrow, 1990). This, however, is overlooked in conventional laboratory measurements of contact angle. Additionally, the substrates used for contact angle measurements have smooth surfaces whereas the pore walls in natural rocks have microscopic roughness – one of the main reasons causing contact angle hysteresis (Israelachvili, 2011; Morrow, 1975; Oliver et al., 1980), the difference between advancing contact angle (when the wetting phase displaces the non-wetting phase) and receding contact angle (when the non-wetting phase displaces the wetting phase). Furthermore, the substrates used in these measurements have flat surfaces, while pores are often formed with curved walls. Curvature of the solid surface may significantly impact contact angle measurements. All the above-mentioned drawbacks imply that contact angle measurements made through conventional techniques may not precisely represent the wetting characteristics of natural rocks, unless made directly inside the pore space under relevant conditions. This goal, however, was unattainable for many years.

The advent of imaging techniques, in particular X-ray computed tomography, in the past few decades has created a turning point for fundamental studies in environmental engineering and petroleum engineering. The use of X-ray computed tomography (CT) to determine physical and chemical properties of porous media and their fluid contents with spatial resolutions on the sub-millimeter (micron) level has created invaluable opportunities to develop insights into fundamentals of fluid flow through porous media. This novel technology enables us to visualize spatial distribution of fluids at the pore level by means of miniature core-flooding setups and micro-CT scanners (Alizadeh et al., 2014). The use of micro-CT imaging is increasingly becoming pervasive in various areas of reservoir engineering, such as pore-scale characterizations (Arns et al., 2005), investigation of trapped non-wetting phase clusters (Iglauer et al., 2010; Khishvand et al., 2016), visualization of fluid occupancy in fractures (Karpyn et al., 2007), and capillary pressure characterization (Armstrong et al., 2012). Wildenschild and Sheppard (2013) have provided more details regarding the applications of X-Ray microtomography. Micro-CT images have recently also been used to measure contact angle at the pore level (Andrew et al., 2014; Khishvand et al., 2016). Andrew et al. (2014) carried out a flow experiment on Kenton limestone using a supercritical CO_2-brine fluid system and measured pore-scale contact angles at the end of imbibition using X-ray microtomography. The measured contact angles covered a range of values (a distribution), which was ascribed to contact angle hysteresis and surface heterogeneity. Optical thin section and SEM images were used to examine surface roughness. No attempt was made to discern the difference between hinging and advancing contact angles. Measurement uncertainties (particularly voxelation error) and contact angle relaxation due to the cessation of flow prior to scanning could have influenced the results. During relaxation, as the system is allowed to come to rest, the advancing (or receding) angle may approach toward an equilibrium value (Bartell and Merrill, 1932; Hassanizadeh and Gray, 1993). Khishvand et al. (2016) addressed some of these uncertainties and improved wettability characterization techniques during their measurements on Gambier limestone and Bentheimer sandstone. To avoid contact angle (interface) relaxation, flow of fluids was not stopped while scanning. All the contact angles were measured at steady state. They used a new segmentation approach (and adequate phase contrasts) instead of conventional watershed segmentation to minimize the voxelation error. It was also tried to distinguish between Main Terminal Meniscus (MTM) and Arc Meniscus (AM) contact angles and characterize receding and advancing angles during drainage and imbibition processes. The investigators employed their technique to measure in-situ oil-brine contact angles and used the data to explain capillary de-saturation behavior of different rock samples. In the present study, we used a similar technique to characterize contact angle distributions for three different pairs of fluids and then employed the measurements to develop an improved insight into subtle complexities of pore-scale displacement mechanisms during two- and three-phase flow processes that had only been proposed theoretically and not been examined experimentally at the pore scale in natural porous media.

We performed two- and three-phase flow experiments on Berea sandstone rock samples and imaged fluid configurations inside the pore space to measure contact angles at arc menisci and main terminal menisci. This study was aimed at establishing a superior workflow, which could overcome some of the errors associated with conventional contact angle measurement techniques, such as overlooking mineral heterogeneity and surface roughness. We provided a perfect framework including a robust experimental procedure and a comprehensive image analysis approach to quantify the wettability behavior of the rock during two- and three-phase flow experiments. We evaluated the effect of relaxation on the interface movement and highlighted the importance of continuous flow during in-situ contact angle measurement. For the first time, in-situ receding, hinging, and advancing contact angle distributions were characterized for different pairs of fluids, i.e., oil-brine, gas-oil, and gas-water, in a natural rock. These values are representative of the wettability behavior of Berea sandstone. We also explained displacement mechanisms responsible for the observed contact angle trends. Moreover, a series of two-phase oil-brine, gas-oil, and gas-brine experiments were performed under different thermodynamic conditions to examine the effect of fluid equilibration on contact angle behavior. Finally, we conducted a three-phase flow experiment to determine contact angle distributions and use them, along with pore-scale images, to find dominant double displacement mechanisms occurring during the three-phase saturation path taken in the experiment.