



Direct pore-scale reactive transport modelling of dynamic wettability changes induced by surface complexation

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ABSTRACT

Laboratory experiments have shown that oil production from sandstone and carbonate reservoirs by waterflooding could be significantly increased by manipulating the composition of the injected water (e.g. by lowering the ionic strength). Recent studies suggest that a change of wettability induced by a change in surface charge is likely to be one of the driving mechanism of the so-called low-salinity effect. In this case, the potential increase of oil recovery during waterflooding at low ionic strength would be strongly impacted by the inter-relations between flow, transport and chemical reaction at the pore-scale. Hence, a new numerical model that includes two-phase flow, solute reactive transport and wettability alteration is implemented based on the Direct Numerical Simulation of the Navier–Stokes equations and surface complexation modelling. Our model is first used to match experimental results of oil droplet detachment from clay patches. We then study the effect of wettability change on the pore-scale displacement for simple 2D calcite micro-models and evaluate the impact of several parameters such as water composition and injected velocity. Finally, we repeat the simulation experiments on a larger and more complex pore geometry representing a carbonate rock. Our simulations highlight two different effects of low-salinity on oil production from carbonate rocks: a smaller number of oil clusters left in the pores after invasion, and a greater number of pores invaded.

1. Introduction

Reactive transport in porous media is an essential field of study in a broad range of applications including oil and gas production, carbon dioxide (CO₂) sequestration, nuclear waste storage and transport of pollutant in the subsurface (Steeffel et al., 2005). Porosity and wettability changes induced by chemical reactions is relevant to a range of production-related applications for oil and gas reservoirs, including diagenesis, well stimulation and Enhanced Oil Recovery (EOR). These processes are particularly important in the case of carbonate reservoirs for which recovery factors generally do not exceed 30%. EOR processes such as CO₂ flooding (Han and Gu, 2014) and Low Salinity Flooding (LSF) (Mahani et al., 2015b) have the potential to significantly increase the recovery factors by changing flow properties such as viscosity and wettability, and increase the overall sweep efficiency in the reservoir.

The coupling between chemical reactions and transport is traditionally described at the macro-scale by an Advection-Dispersion-Reaction Equation (ADRE). However, at the macro-scale, the ADRE has been shown to give inaccurate predictions of experimental results (Gramling et al., 2002). This discrepancy has been linked with

incomplete mixing of the injected and resident fluids (Alhashmi et al., 2015; Karadimitriou et al., 2017). Therefore, modelling the reactive transport at the pore-scale, where the mixing can be resolved on a pore-by-pore level, is a crucial tool to better constrain macroscopic reactions (Zaretskiy et al., 2010).

Although pore-scale non-reactive (Bijeljic et al., 2013; Hosa et al., 2016; Kallel et al., 2016) and reactive transport (Alhashmi et al., 2015; Soulaïne and Tchelep, 2016; Zaretskiy et al., 2012) have been studied extensively in recent years, only a few studies have included wettability alteration. Kallel et al. (2017) developed a model for wettability alteration during primary drainage based on polar compound adsorption. Watson et al. (2017) included wettability alteration with increasing tracer concentration following a Heaviside function for the contact angle. None of these studies included the geochemical reactions that are taking place at the water/mineral interface and can influence wettability. One of the reasons for this is that wettability change induced by manipulating the injected water composition (e.g. lowering the ionic strength) is a mechanism that is not fully understood yet (Mahani et al., 2015b; Tang and Morrow, 1997). This effect has been associated in the past with a reduction of the salt content of water (the so-called Low

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Nomenclature*Variables*

<i>c</i>	concentration (mol/L)
<i>D</i>	molecular diffusivity (m ² /s)
<i>f</i>	interior force (N/m ³)
<i>H</i>	Henry constant
<i>I</i>	ionic strength (mol/L)
<i>J</i>	molecular diffusion flux (mol/m ² /s)
<i>K</i>	stability constant
<i>L</i>	characteristic length (m)
<i>m</i>	surface concentration (mol/m ²)
<i>p</i>	pressure (Pa)
<i>r</i>	bulk chemical reaction molar rate (mol/m ³ /s)
<i>s</i>	surface chemical reaction molar rate (mol/m ² /s)
<i>T</i>	temperature (K)
<i>t</i>	time (s)
<i>U</i>	characteristic velocity (m/s)
<i>u</i>	velocity (m/s)
<i>v</i>	surface complex charge
<i>Z</i>	ionic charge
α	phase volume fraction
Γ	surface sites density (mol/m ²)
γ	interfacial tension (N/m)
κ	interface curvature (m ⁻¹)
μ	viscosity (Pa.s)
ψ	surface potential (V)
ρ	mass density (kg/m ³)
σ	surface charge density (C/m ²)
θ	contact angle

Constants

<i>F</i>	Faraday constant (96490 C/mol)
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<i>g</i>	gravity constant (9.81 m/s ²)
<i>R</i>	universal gas constant (8.314 J/mol/K)
<i>e</i>	dielectric constant of pure water (78.41 at 25 °C)
ϵ_0	vacuum permittivity (8.854 × 10 ⁻¹² C/V/m)

subscripts

<i>i</i>	component index
<i>j</i>	phase index
<i>s</i>	surface
<i>s_i</i>	surface component index
<i>st</i>	surface tension

Abbreviations

25dSW	25 × diluted Sea Water
ADRE	Advection-Diffusion-Reaction Equation
CSF	Continuous Surface Force
CST	Continuous Species Transfer
EOR	Enhanced Oil recovery
FW	Formation Water
HS	High Salinity
IHP	Inner Helmholtz plane
LBM	Lattice-Boltzmann Method
LSE	Low-Salinity Effect
LSF	Low-Salinity Flooding
NSE	Navier–Stokes Equations
OHP	Outer Helmholtz plane
PDI	Potential Determining Ions
PNM	Pore Network Modelling
PV	Pore Volume
SCM	Surface Complexation Modelling
SSF	Sharp Surface Force
SW	Sea Water
VOF	Volume-Of-Fluid

Salinity Effect, or LSE) (Tang and Morrow, 1997). However, recent advances in experimental work, in particular in the domain of fluid visualisation, greatly improved the understanding of this phenomenon.

For sandstones, Tang and Morrow (1997) observed that LSE is linked to the presence of clay minerals, and also depends on a number of factors such as oil composition, formation water composition and salinity of the injected water. A number of potential low-salinity mechanisms have been proposed in the literature such as fines migration (Tang and Morrow, 1999), interfacial tension reduction and emulsification (McGuire et al., 2005), selective plugging via clay swelling (Zhang and Morrow, 2006) and wettability alteration by multi-component ionic exchange (Tang and Morrow, 2002) and/or electrical double-layer expansion (Austad et al., 2010). Berg et al. (2010) directly visualised detachment of crude oil from clay minerals. They observed a release of crude oil when changing the brine from high to low salinity even when the amount of clay swelling was insignificant.

For carbonate rocks, the effect cannot be related to the presence of clay, as most of these rocks do not contain any, and if they do, they occur only at very low levels. Moreover, the exact chemical composition of the injected water seems to play a more important role than just salinity and ionic strength (Zhang et al., 2007). Mahani et al. (2015b) observed a LSE on calcite patches in the absence of dissolution and with an increase of interfacial tension. They could link the change of contact angle to a change of ζ -potential, defined as the local electrical potential at the slipping plane (shear plane) that separates the stationary and mobile phases in tangential flow of the liquid with respect to the surface (Hunter, 1981). More recently, Jackson et al. (2016a); 2016b) observed that the increase of oil recovery from carbonate core-plugs obtained by

manipulating the injected water composition depends greatly on the sign of the ζ -potential at both water/mineral and water/oil interfaces. Both these studies suggest that LSE in carbonate rocks is driven by a wettability change induced by a change of surface charge and the double-layer expansion. To analyse this effect further, Surface Complexation Models (SCM) (Goldberg, 2013) can provide a link between the water composition and the change of potential.

SCM are chemical models that provide a molecular description of adsorption phenomena using an equilibrium approach. One of the major advancements of SCM is that it considers the charges on both the adsorbing ion and the solid adsorbent surface (Goldberg, 2013). Hence, SCM can provide the link between water composition and change of surface potential. SCM for clay minerals (Bradbury and Baeyens, 1997), calcite and dolomite (Pokrovsky et al., 2000) have been proposed. In the context of EOR, Brady and Krumhansl (2012) and Brady et al. (2012) have used such models to investigate reactions at the water/oil and water/mineral interfaces that lead to oil adhesion on mineral surfaces. More recently, Mahani et al. (2016) developed a calcite SCM, adapted from Pokrovsky et al. (2000), and modified so that it can explain the impact of sulphate concentration on the ζ -potential at the water/calcite interface.

The objective of this work is to develop an internally consistent pore-scale numerical model that (1) captures the effect of surface complexation on the surface potential and wettability of carbonate rocks, (2) can explain experimental observations at the micro-scale and (3) can be easily parameterized to study the impact of different water, mineral and oil compositions. For this, we use direct numerical simulation of the Navier–Stokes Equations (NSE) (Patankar and

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