



Full Length Article

Modeling the characteristic thermodynamic interplay between potential determining ions during brine-dependent recovery process in carbonate rocks



Adedapo N. Awolayo^{a,*}, Hemanta K. Sarma^a, Long X. Nghiem^{a,b}

^a Chemical and Petroleum Engineering Department, University of Calgary, Canada

^b Computer Modelling Group Ltd., Calgary, Canada

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ABSTRACT

Brine-dependent recovery processes in carbonate reservoirs have been receiving much attention for the past two decades, and wettability alteration has been identified among the dominant process mechanisms. Most studies suggest that the wettability alteration in carbonate rocks is caused by desorption of oil acid groups from rock surfaces by the adsorbed sulfate, with concomitant co-adsorption of the divalent cations. Although such symbiotic interaction between active ions (Ca^{2+} , Mg^{2+} , and SO_4^{2-}) and the rock surface has been established, their systematic interplay under different conditions remains to be better explored and understood. In this study, we propose a reactive transport model that considers various reaction sets to investigate the affinity of these active ions toward the rock surface. The important thermodynamic properties were obtained by using the model to interpret single-phase experiments. The model results were in excellent agreement with the produced ion histories reported from single-phase flow through experiments. Using the same thermodynamic parameters, our model also replicated the produced ion histories and oil recoveries obtained during brine-oil flooding experiments. The established thermodynamic parameter can be applied to predict various brine-dependent recovery processes in different carbonate lithology as no significant difference was observed for the interplay between active ions and either chalk or limestone rocks. Accordingly, the Derjaguin–Landau–Verwey–Overbeek (DLVO) theory of surface forces was used to rationalize active ions interactions and to evaluate the contribution of each force component to the wettability change in the oil-brine-rock system. The results emphasize that disjoining pressure and electrostatic interaction energy are sensitive to $\text{SO}_4^{2-}/\text{Ca}^{2+}$ ratio as well as brine salinity.

1. Introduction

In recent years, brine-dependent recovery processes have seen much R&D efforts, in particular for carbonate reservoirs because of their potential to improve oil recovery through a relatively inexpensive means and ease of operation. A major thrust in these research efforts has been to tweak and optimize the brine chemistry that delivers more effective and efficient oil-brine-rock interactions leading to an improved oil recovery. Several studies ranging from laboratory to field tests suggest that brine-dependent recovery leads to additional oil production in both sandstone and carbonate reservoirs [1–12]. The effectiveness of the recovery process is mostly governed by the composition of flowing fluids in the porous media and the mass transfer between such flowing (aqueous/oleic) fluids and the immobile (rock) phase. In addition, this process is often explored on two major fronts: ionic composition and strength modification [13], the latter having

been a successful scheme in sandstone reservoirs. To date, a significant volume of research studies has been conducted to resolve pending arguments about the applicability of the process in sandstones; however, research on carbonates reservoirs still lies in its emerging phase.

Results from most experimental studies in carbonate rocks (for example, displacement tests, spontaneous imbibition, contact angle, NMR, zeta potential, effluent analysis tests) point to the ability of the process to shift rock wettability towards water wetness as the principal fundamental mechanism. However, there appears to be no unanimity as to how the wettability alteration occurs, and hence, this remains a gray area and needs to be studied. A review of mechanisms that influence wettability changes has been presented by several authors, including electrostatic interactions between rock-brine and oil-brine interfaces, multi-ion exchanges between surface active ions, rock mineral alteration in terms of dissolution/precipitation, complexation between oil phase components, multivalent ions and rock surface

* Corresponding author.

E-mail addresses: adedapo.awolayo@ucalgary.ca (A.N. Awolayo), hemanta.sarma@ucalgary.ca (H.K. Sarma), long.nghiem@cmgl.ca (L.X. Nghiem).

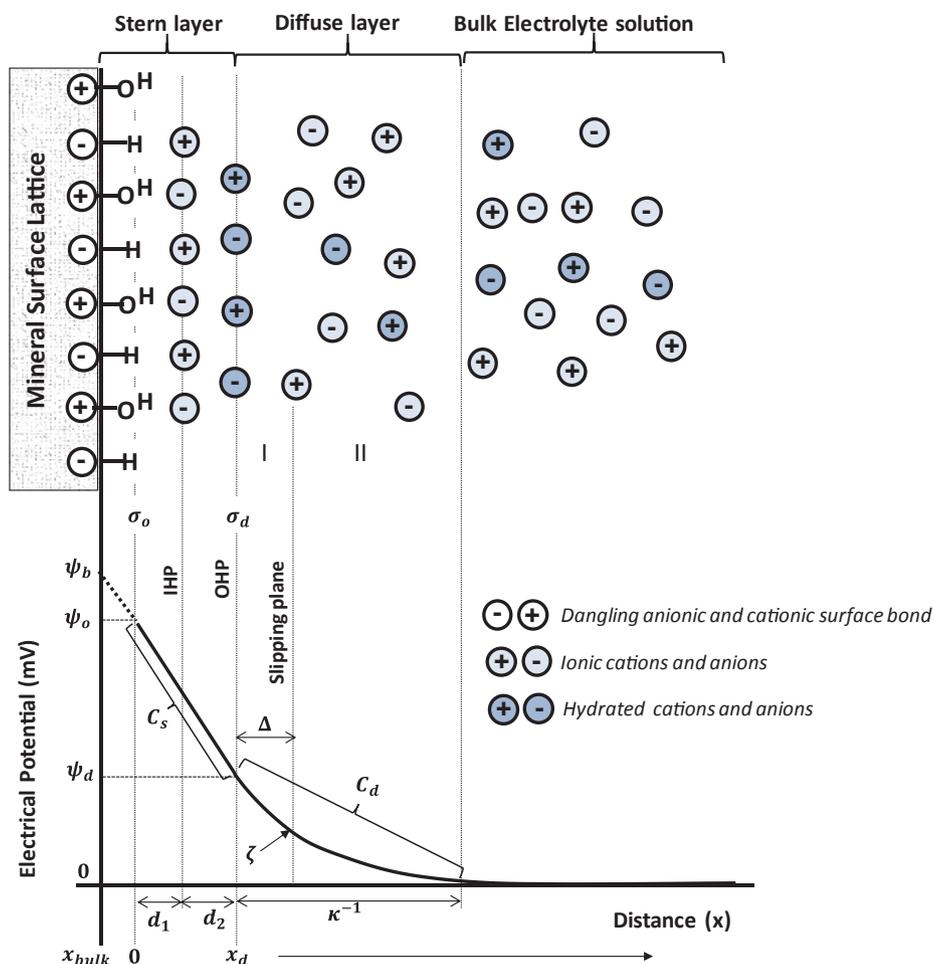


Fig. 1. Schematic representation of the electrical double layer (EDL) and electrical potential at the rock–brine interface. The sketch shows the variation of electrical potential as a function of distance from the rock surface, partitioned by charged planes – inner Helmholtz plane (IHP), outer Helmholtz plane (OHP) and Slipping plane. The potential developed within the EDL declines with distance linearly through the Stern layer, exponentially through the diffuse layer and drops to zero in the bulk electrolyte solution. The partial charge on the dangling surface ions left behind at the bulk solid is represented by ψ_b ; ψ_0 represents the potential of the surface; ψ_d stands for the potential at the Stern layer and ζ represents zeta potential. While σ_o and σ_d are the surface and diffuse layer charge density (C/m^2) respectively. The stern layer potential difference is characterized by constant capacitance, C_s , while the diffuse layer has variable capacitance, C_d . At plane $x = 0$, which corresponds to the hydrolysis layer, H and OH of the water molecules are chemisorbed to the dangling surface ions. At $x < 0$, the potential is so high that attaching ions do not bond to the surface ions. The inner-stern layer is characterized by d_1 length; the outer-stern layer is characterized by d_2 length, and the diffuse layer is characterized by κ^{-1} length, also known as Debye length [17,18].

[2,3,10–12,14,15]. It is important to note that the magnitude of the forces existing between reservoir fluids (with their dissolved constituents) and the rock surface often controls wettability. Hence, the wettability changes can only be fully understood by considering a detailed account of the intermolecular interactions between fluids in contact with the rock surfaces [16].

Moreover, many of these mechanisms combine to launch various interfacial phenomena occurring concurrently during fluid flow in porous rock. These phenomena involve intermolecular forces acting on the oil-brine-rock system, which is composed of rock-brine and oil-brine interfaces interacting through the water-film layer. At the interface between a charged surface and an electrolyte solution, an electrical potential is developed such that the surface charge is neutralized via coulombic interaction with oppositely charge ions (counter-ions) attracted to the surface from the adjacent solution. Subsequently, layers of ions are developed separating the charged surface from the bulk solution in a special arrangement known as the electrical double layer (EDL; shown in Fig. 1). The first region next to the charged surface is the stern layer; the counter-ions are specifically attached in the inner stern layer defined by inner Helmholtz plane (IHP). The hydrated counter-ions that do not specifically attach are in the outer stern layer (also defined by outer Helmholtz plane – OHP). The second layer, known as the diffuse layer, contains ions of the same sign (co-ions) and

counter-ions that are not attached and move freely under the influence of thermal motion and electric attraction [17–20]. These ions are present to ensure electrical neutrality of the EDL, at any circumstances that the stern layer failed to neutralize the surface charge. The diffuse layer extends from the OHP to a point where electric attraction is not felt by ions in the bulk electrolyte solution and the thickness of this diffuse layer is termed Debye length (κ^{-1}). The shear or slipping plane, to which measured zeta potentials are attributed, are often displaced by a distance Δ from the OHP into the diffuse layer [21]. The specific distance is unknown and generally used as an adjustable parameter to fit zeta potential data, within 0.245 - 1 nm has been reported [22–25]. The distribution of ions in the region displaced by the distance Δ in the diffuse layer (see region I in Fig. 1) is not influenced by tangential stress arising from bulk fluid flow while ions in the second region (see region II in Fig. 1) of the diffuse layer are distributed based on flow of the bulk fluid.

1.1. Theory of film stability

When two interfaces approach and interact with each other, the distance separating them is considered to influence the energy of the oil-brine-rock system and as a result, the stability of the water-film layer between the interfaces. These interactions are mostly dictated by the

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