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Thin liquid films in improved oil recovery from low-salinity brine

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ABSTRACT

Low-salinity waterflooding is a relatively new method for improved oil recovery that has generated much interest. It is generally believed that low-salinity brine alters the wettability of oil reservoir rocks towards a wetting state that is optimal for recovery. The mechanism(s) by which the wettability alteration occurs is currently an unsettled issue. This paper reviews recent studies on wettability alteration mechanisms that affect the interactions between the brine/oil and brine/rock interfaces of thin brine films that wet the surface of reservoir rocks. Of these mechanisms, we pay particular attention to double-layer expansion, which is closely tied to an increase in the thickness and stability of the thin brine films. Our review examines studies on both sandstones and carbonate rocks. We conclude that the thin-brine-film mechanisms provide a good qualitative, though incomplete, picture of this very complicated problem. We give suggestions for future studies that may help provide a more quantitative and complete understanding of low-salinity waterflooding.

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1. Introduction

Despite society's best efforts towards developing renewable energy sources, more than 70% of the global energy consumption in the coming decades is expected to come from fossil fuels [1]. To meet this high demand, the petroleum research community has continually strived to develop innovative methods for improved oil recovery. One such method that has gained much attention in the past two decades is low-salinity waterflooding (LSW). The improved recovery from LSW is referred to in the literature as the low-salinity effect (LSE). LSW has attracted great interest partly because of its deceptively simple nature. As its name implies, LSW involves the injection of only low-salinity brine. No additional chemicals, which may be costly to the operation, are necessary to observe the LSE, although there have been recent efforts to couple LSW with polymer flooding [2]. Improved recovery has been observed for both secondary-mode LSW, which involves injection of brine into rock saturated with oil, and tertiary-mode LSW, which involves injection of low-salinity brine after the rock has already been flooded in secondary mode with brine of a different composition.

Studies on LSW date back to at least the 1940s, although most of the modern work on this topic began in the 1990s [3,4]. Since then, the number of papers on LSW has rapidly increased, especially in the past several years. Based on these studies, it is generally believed that LSE

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occurs because LSW alters the wettability of oil reservoir rocks towards a state more favorable to oil recovery [4,5]. Other processes, such as an increase in the elasticity of brine/oil interfaces that we discuss briefly in Section 4, may also contribute to the improved oil recovery from LSW, but wettability alteration is currently thought to be a major factor. To understand the wettability alteration, it is important to note that the pores of rock saturated with oil are wetted by a residual thin brine film, with oil occupying the rest of the pores. Saturating rock with the oil may cause the rock to become more oil-wet through a variety of brine/oil/rock interactions [6]. LSW studies on sandstones conclude that a high oil recovery is observed in rocks that are weakly waterwet [3,7]. LSW alters the wettability of the sandstones from an oil-wet state towards an optimum weakly water-wet state, thereby leading to an improved recovery. The LSW literature can be classified into studies on sandstones and studies on carbonates. This is a natural choice of division because the surface charge and the chemical reactivity of the two rock types, which consequently affect their wetting behavior, can be very different under the same reservoir conditions [8,6,9[•]]. In fact, until the recent work of Yousef et al., LSE was not even observed in carbonates [10,11[•]]. Yousef et al. report that LSW alters the wettability towards more water-wet conditions, and the maximum LSE is observed for rocks that are weakly water-wet. In summary, LSW in both sandstones and carbonates may alter the wettability towards an optimum state that lies in the weakly-wet region of the spectrum. A glaring question that remains is how does this alteration occur?

The mechanism(s) by which the wettability alteration occurs is currently a major topic of debate in the LSW literature. Various

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mechanisms have been proposed, but none of them have definitively been shown to be the primary one [12]. This is due to the complex nature of the brine/oil/rock interactions, and is further complicated by a number of conflicting observations from experimental studies [4]. The low-salinity effect likely results from a combination of more than one mechanism. This review focuses on mechanisms which change the stability of thin brine films that wet the surface of oil reservoir rocks. We pay particular attention to the so-called double-layer expansion (DLE) [13,14",15"]. DLE is described by classical Derjaguin-Landau-Verwey-Overbeek (DLVO) theory. In DLE, injection of low-salinity brine increases the electrostatic repulsion between the film's brine/oil and brine/rock interfaces via the expansion of two electrical double layers, one formed at each of the two interfaces. Consequently, the film becomes thicker and more stable, resulting in a more water-wet rock. Our review also includes studies on chemical mechanisms in low-salinity brine which affect the non-DLVO interactions between the brine/oil and brine/rock interfaces. For example, wettability alteration may occur because of multicomponent ionic exchange (MIE) involving divalent cations near the clay surfaces of sandstones [16[•]]. Crude oil forms organometallic complexes with divalent cations adsorbed on the clay surface. In LSW, MIE occurs so that the complexed cations are replaced with uncomplexed cations from the brine film, leading to release of the organometallic complexes and oil recovery.

Section 2 discusses recent (published 2009 or later) studies in detail which show that DLE and the chemical mechanisms, which we refer to collectively as thin-brine-film mechanisms, can play an important role in low-salinity wettability alteration of sandstones. Section 3 discusses studies on thin-brine-film mechanisms in carbonates. We conclude with some closing remarks and suggestions for future studies in Section 4.

2. Wettability alteration involving thin brine films on sandstones

2.1. Chemical composition of sandstones

Sandstones are primarily composed of quartz, which has the same chemical formula as silica (SiO₂), but may also contain other minerals (e.g., feldspars, anhydrite, mica, calcite) as well as various clays (kaolinite, illite, chlorite, montmorillonite). The clays are particularly important for LSW because they are commonly found along the surfaces of the pores in which the oil and brine reside. Silica has an isoelectric point at a pH value of 2, and its surface becomes more negatively charged as the pH increases above this value [8,6]. Reservoir brines are typically at pH values above the isoelectric point. Clays are also negatively charged on their faces. As a result, sandstones are negatively charged along the surface of the pores.

These negative charges play a central role in the wettability alteration mechanisms described in Section 2.2.

2.2. Double-layer expansion (DLE) and two chemical mechanisms

The wettability of the reservoir rock depicted in Fig. 1 depends on the stability of the thin brine film that wets its surface. The film's stability is influenced by interactions between its brine/oil and brine/rock interfaces. In sandstones, these interactions include: 1) electrostatic interactions between charged groups on the oil surface of the brine/oil interface and charges on the rock surface of the brine/rock interface; 2) hydrogen bonding between polar functional groups in the crude oil, such as those present in asphaltenes, and polar groups on the rock surface; 3) Lewis acid/base interactions between charged basic groups (e.g., NH₄⁺) on the oil surface and negatively-charged groups on the rock surface; 4) formation of organometallic complexes between charged acidic groups (e.g., COO⁻) on the oil surface and divalent cations (usually Ca^{2+} and Mg^{2+}) adsorbed on the rock surface. The first interaction is described by classical DLVO theory, while the last three, which are depicted in Fig. 1(b), can be thought of as non-DLVO interactions between the brine film's two interfaces [17,16,18"]. All of these interactions contribute to the disjoining pressure $\Pi(h)$ in the film, a quantity that depends on its thickness h. The average thickness of the wetting brine films is thought to be less than 10 nm, and may be much smaller than that value [8,19]. Attractive interactions between the two interfaces produce negative contributions to Π that cause the film to collapse, decreasing h. Repulsive interactions produce positive contributions to Π that stabilize the film and increase h. Stable, thick brine films are indicative of a water-wet state. The LSW mechanisms described in this section either weaken attractive interactions or strengthen repulsive interactions, thereby resulting in a more stable, thicker brine film and a transition from an oil-wet state to a more water-wet state that results in improved oil recovery.

One of the most simple, yet effective ways in which low-salinity brine may alter the wettability is double-layer expansion [15^{*}]. Counterions in the brine film adsorb to the negatively-charged brine/oil and brine/rock interfaces, whose electrostatic potentials we represent by ψ_1 and ψ_2 , respectively. The counterions form an electrical double layer at each interface and screen the repulsion between the two negatively-charged interfaces [Fig. 2(a)]. A characteristic length of this screening is the Debye length κ^{-1} given by

$$\kappa^{-1} = \left(\frac{\varepsilon_r \varepsilon_0 k_B T}{2N_A e^2 I}\right)^{1/2},\tag{1}$$

where ε_r is the relative permittivity (dielectric constant) of the brine, ε_0 is the permittivity of free space, k_B is the Boltzmann constant, *T* is the

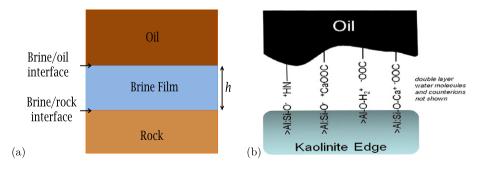


Fig. 1. (a) The wettability of oil reservoir rocks is influenced by interactions between the brine/oil and brine/rock interfaces of thin brine films that wet the surfaces of the pores in which the fluids reside. Repulsive interactions produce a positive contribution to the disjoining pressure Π in the film that increases the film's thickness *h* and leads to a more water-wet state. The average thickness of these wetting films is thought to be less than 10 nm, and may be much smaller than that value [8,19]. (b) Is a schematic of non-DLVO interactions between brine/oil and brine/clay (kaolinite) interfaces, and is adapted from the Fig. 6 of [20°], with permission.

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