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Historical perspective

Understanding the role of brine ionic composition on oil recovery by assessment of wettability from colloidal forces

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ABSTRACT

The impact of injection brine salinity and ionic composition on oil recovery has been an active area of research for the past 25 years. Evidence from laboratory studies and field tests suggests that implementing certain modifications to the ionic composition of the injection brine leads to greater oil recovery. The role of salinity modification is attributed to its ability to shift wettability of a rock surface toward water wetness. The amount of trapped oil released depends on the nature of rock, oil, and brine surface interactions. Reservoir rocks exhibit different affinities to fluids. Carbonates show stronger adsorption of oil films as opposed to the strongly water-wet and mixed-wet sandstones. The concentration of divalent ions and total salinity of the injection brine are other important factors to consider. Accordingly, this paper provides a review of laboratory and field studies of the role of brine composition on oil recovery from carbonaceous rock as well as rationalization of results using DLVO (Derjaguin, Landau, Verwey and Overbeek) theory of surface forces. DLVO evaluates the contribution of each component of the oil/brine/rock system to the wettability. Measuring zeta potential of each pair of surfaces by a charged particle suspension method is used to estimate double layer forces, disjoining pressure, and contact-angle. We demonstrate the applicability of the DLVO approach by showing a comprehensive experimental study that investigates the effect of divalent ions in carbonates, and uses disjoining pressure results to rationalize observations from core flooding and direct contact-angle measurements.

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1. Introduction

Alteration of injection water salinity has the potential to increase oil recovery and it is emerging as a relatively new improved/enhanced oil recovery method. The role of water salinity to increase oil recovery in sandstone reservoirs was identified more than 65 years ago. It was observed that fresh water injection in a sandstone core containing clays gives greater recovery when compared with brine injection [1]. The recovery increase was accompanied with a lowering of permeability and development of relatively high pressure drop [2]. The findings of these earlier studies were revisited in the 90s and the effect of the salinity was considered an opportunity to maximize oil recovery [3,4]. Following these studies, a great deal of research has been focused on attempting to assess the salinity effect on oil recovery, primarily in sandstones [5–8].

Research on the salinity effect on oil recovery from carbonates is relatively new compared with sandstones. Increasing the concentration of the divalent ions of the injection seawater, such as Mg^{2+} , in chalk cores was found to increase oil recovery during spontaneous imbibition experiments [9]. Reducing the total salinity was effective in increasing oil recovery as well when forced core flooding was performed on limestone cores. Significant reductions in salinity as well as more moderate reductions influenced recovery positively. Yousef et al. [10] flooded a composite core with seawater of 57 kppm until the residual oil saturation was reached. Additional oil was recovered when this seawater flooding was followed by twice diluted seawater.

Interestingly, studies examining the salinity effect in carbonates have not definitively differentiated the role of very dilute salinity as compared to more concentrated brines with key additions of certain divalent species. The former is expected to result in increasing importance of double-layer forces and dissolution of rock species whereas the latter suggests that ion exchange may be relevant.

Nevertheless, several mechanisms were proposed to understand the effect of the key conditions that lead to a successful effect of brine ionic composition modification on oil recovery for both sandstone and carbonate rocks. The main mechanism that was suggested based on the DLVO theory is double layer expansion [11–13]. Other mechanisms explained the interactions in terms of non-classical DLVO approach such as the multi-ion exchange [6] for sandstone rocks, and ion-substitution-adsorption [9,14,15], and anhydrite dissolution [16–18], for carbonate rocks. Assuming negligible effect of DLVO surface forces in carbonate systems, it was hypothesized that rock dissolution of sites contacting oil droplets is the cause of the oil release [19,20]. Most of the mechanisms were proven to take place but the degree to which they cause an increase in oil recovery is not settled yet. These studies generally agree that the main cause of the additional oil recovery is the shift in wettability toward water wetness that is induced by ionic interactions of oil, brine, and rock interfaces while they differ in explaining the details of the wettability alteration process. The change in zeta potential of the brine/rock interface was observed in carbonate systems and considered an indirect proof for the role of surface forces to alter wettability at the double layer region [16,9] as opposed to the rock dissolution hypothesis that was not verified.

This paper presents a wettability assessment method of thin films from zeta potential measurements and predictions of contact-angle and disjoining pressure. An experimental case study is discussed that illustrates the impact of altering ionic composition of injection brine on oil recovery and contact-angle. The DLVO approach correctly describes the trend of wettability alteration and shift in contact-angle

that was observed from experiments for dilute brines. Also it provides a better understanding for the role that each component of oil, brine, and rock play to alter wettability.

In the following section, we review the key findings of studies that examined the role of ionic compositions in carbonate systems. The next section reviews the proposed scenarios for wettability development and alterations with respect to thin films. Then we review the DLVO theory and governing equations of surface forces and their relationship to wettability. After that we discuss a comprehensive experimental study that shows the role of divalent ions in altering wettability and increasing oil recovery. Conventional forced core flooding and direct contact-angle measurements were conducted using different brines. The following sections demonstrate utilization of DLVO theory as reviewed, to assess wettability by calculating disjoining pressure and reconstructing contact-angle from zeta potential measurements.

2. Studies of carbonate systems

Carbonates refer to rocks that are composed primarily of calcium carbonate. Calcite is a stable polymorph of calcium carbonate and is rarely found in nature in its pure form. An example of mineral calcite is Iceland spar that is a transparent cleavable crystal. A synthetic calcium carbonate can be processed to form a powder of small size particles, sub-micron to nanometer. A relatively pure form of calcium carbonate is found in chalk reservoirs, that are common in the North Sea fields. The super-giant fields in the Middle East have carbonate reservoirs containing dolomite and anhydrite impurities [21–23].

Experimental studies have found that different schemes of salinity modification of the injection brine are able to produce additional oil from carbonate porous media. Spontaneous imbibition tests conducted on chalk cores were sensitive to the amount of divalent ions in the invading brine. Manipulation of the amounts of Ca^{2+} , Mg^{2+} , and SO_4^{2-} ions in solution boosted oil production and increased oil recovery by more than 10%. The additional oil recovery correlates with the increase of SO_4^{2-} ions. Zeta potential was measured as a function of ion concentrations in aqueous chalk suspensions. The observed zeta potential values are linearly proportional to the amount of Ca^{2+} and Mg^{2+} ions, and inversely proportional to SO_4^{2-} . It was concluded from the tests that these potential determining ions influence chalk surface charges and shift wettability to stronger water wetness [24,25,9].

Modeling of surface chemistry and disjoining pressure confirmed the impact of divalent ions on wettability [19,26]. It was argued, however, that the primary cause of wettability alteration is from dissolution of the rock surface on sites where oil droplets are trapped. It is worth noting that these studies evaluated the change of zeta potential for calcite/brine suspensions but did not, however, account for the zeta potential changes in the oil–water emulsion suspension.

It is possible to increase oil recovery by reducing the total salinity of the injection water without any modification of the initial ions relative concentrations [10,20]. Forced imbibition tests conducted on carbonate reservoir cores from the Middle East show that additional oil was released during injection cycles of diluted brines. The experiment began by injecting seawater of 57 kppm until the residual oil saturation was reached and no more oil was produced at the effluent. Production of trapped oil was observed when the injection brine was diluted with distilled water of the same volume. Further dilution of the injection brine, 20 times dilution, prompted the release of trapped oil. Contact-angle measurements display a shift in wettability conditions from mixed-wet to water-wet [10]. The oil recovery increase was attributed

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