



Full Length Article

Interactions of DTPA chelating agent with sandstone rocks during EOR: Rock surface charge study

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ABSTRACT

Enhanced oil recovery (EOR) using low salinity water has been proven experimentally and at the field scale. One of the mechanisms behind recovery enhancement due to low salinity water injection has been related to the change of rock surface charge which might lead to wettability alteration. Low salinity water EOR has its own disadvantages such as the high cost due to the need for enormous amounts of fresh water and it may cause scale precipitation and fines migration. Therefore, efficient and alternative low-cost EOR methods are needed. In this work, chelating agent diethylenetriaminepentaacetic acid (DTPA) for EOR application in sandstone rock was investigated. DTPA is an efficient metal ion control agent and can seize certain cations (especially Fe^{3+}) from rock surfaces. DTPA chelating agent is very stable up to 400 °F at high pH values. The effect of DTPA injection on the surface charge of the sandstone rocks was investigated. Zeta potential measurements, ions analysis after interaction with the rock, and coreflooding experiments were performed. Three different types of sandstone rocks were used to investigate the changes in rock surface after conditioning with DTPA. Several scenarios were tested to compare low salinity water, deionized water, and seawater with DTPA chelating agent. The comparison was made by measuring the zeta potential of the rock powder. Coreflooding experiment was conducted along with effluent analysis for SO_4^{2-} , Ca^{2+} , Mg^{2+} , Fe^{3+} , and Al^{3+} ions to observe the effect of injecting DTPA with seawater on oil recovery and permeability. The results showed that the addition of DTPA resulted in Fe^{3+} ions absorption from the rock surface. Absorbing such trivalent cation resulted in a significant change in surface charge. Zeta potential dropped towards a more negative value for all sandstone rocks samples used in this study. Zeta potential results showed that mixing Berea sandstone with 5 wt% of DTPA in seawater led to similar surface charge value as if it was mixed with low salinity water for the same period. This means that DTPA combats the effect of the concentrated salts in seawater. DTPA chelated all multivalent cations in seawater and eliminated their effects on the surface charge. Also, DTPA increased the pH of the seawater from 7 to 11 and this will drag the surface charge to more negative. From coreflooding experiment, we observed that injecting 5 wt% DTPA solution in seawater after seawater injection enhanced the oil recovery from 56% to 75% in Berea sandstone cores at 100 °C. The additional oil recovery can be explained by more negative values of the zeta potential of the rock surface. We expect that chelating agent EOR using DTPA is a viable alternative to low salinity water flooding where huge fresh water is required to dilute the seawater.

1. Introduction

The improvement in oil recovery caused by wettability alteration has been proven in laboratories and at the field scale. Many factors contribute to set the wettability preference for a rock. By tuning these factors, the desired wettability of the rock can be obtained that could result in maximum oil production. Different surfactants and low salinity water (LSW) has been used to alter the wettability of the rock [1–6]. It has been observed that the ionic content of the rock and the injected

water plays a major role in wettability alteration and hence in oil recovery [7,8]. The LSW yields better recovery factors due to the low salt concentration of specific ions which could result in lowering the capillary pressure and changing the wettability of the rock to a more water-wet conditions [9–12].

Chelating agents are organic compounds that form soluble, complex molecules with metal ions that can control the reactivity of multivalent metal ions by inactivating the ions. Use of chelating agents as injection fluids for enhanced oil recovery (EOR) could capture certain ions from

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the injected water and formation brine. Ions capturing will result in the release of certain ions from the rock surfaces to achieve the equilibrium in the bulk solution. Chelating agents can be added to seawater without dilution and can be used at very low concentrations. Chelating agents can affect the rock dissolution process that could result in the release of oil from the rock surface and result in improved oil recovery. Chelating agents have been successfully used as an additive in the oil and gas industry in scale removal process, iron control, and matrix stimulation. Recently, they were introduced as standalone fluids for EOR, stimulation, and wettability alteration applications [13].

Wettability of the rock is usually determined by contact angle measurements. Zeta potential measurements can be related to the rock wettability and rock surface charges [14]. Zeta potential is the potential at the shear plane of the electrical double-layer. Zeta potential value is related to the thickness of the double layer and the charge of the interface surface between the mineral and brine. The change in zeta potential indicates qualitatively the change in rock surface charge and rock wettability. For sandstone rocks, the more negative value of zeta potential indicates that the surface is more water-wet [14]. It was found that the rock wettability is strongly related to the water film stability located in-between rock surface and crude oil [15,16].

Since sandstone rocks usually contain clay minerals such as illite, kaolinite, chlorite, and iron minerals such as siderite and ankerite, the presence of iron can significantly affect the surface rock charge. Mahmoud and Abdelgawad (2015) found that hydroxyethylethylenediaminetriacetic acid (HEDTA) chelating agent recovered more than 20% additional oil from Berea sandstone rocks after seawater injection [17]. They concluded that, rock dissolution, wettability alteration, and IFT reduction are possible mechanisms for improved recovery using HEDTA chelating agents in Berea sandstone rocks. In a previous publication from our group, we found that addition of 10 wt% of diethylenetriaminepentaacetic acid (DTPA) solution in seawater showed much less IFT (0.8 mN/m) than distilled water (4.2 mN/m) where both were at pH = 11 (the pH of distilled water was adjusted by sodium hydroxide) [17].

In continuation, our previous work, the effect of DTPA chelating agent on the surface charge of the sandstone rocks was investigated in this work. The stability constant value represents the bond strength between the chelating agents and metal ions (Fe^{+3} , Ca^{+2} and Mg^{+2}) [18]. For DTPA chelating agent, the values of Log (Stability Constant) for Ca^{+2} , Fe^{+3} , Mg^{+2} are 10.9, 28 and 9.3, respectively. This shows that DTPA has a higher potential in seizing the Fe^{+3} compared to other ions. Several zeta potential measurements were performed to compare zeta potential of rock using LSW, deionized water (DW), synthetic seawater (SW) and DTPA. The main objective of this work was to observe the change in zeta potential for three different sandstone rocks when treated with DTPA for different time periods. Coreflooding experiment was conducted to prove the effect on oil recovery and permeability of Berea sandstone rock after injecting DTPA. Also, the effluent analysis was conducted to observe the effect of the chelating agent on rock dissolution.

2. Experimental

2.1. Materials

Three different sandstone rocks with different mineralogy were used in this study (Table 1). Quartz constitutes the major fraction of these sandstone rocks. The three sandstone rocks contain a different ratio of ions. The composition of different ions in these sandstone rocks is given Table 2. Formation water (213,734 ppm), Arabian Gulf seawater (57,670 ppm), and low salinity water (5767 ppm) were prepared by dissolving analytical grade salts in deionized water. The salts such sodium chloride, calcium chloride, magnesium chloride, sodium sulfate, and sodium bicarbonate were purchased from Sigma Aldrich. A commercially available chelating agent DTPA was used. The chemical

Table 1

Mineralogy of sandstone cores in wt% [19].

Mineral	Berea	Bandera	Kentucky
Quartz	87	58	66
Dolomite	1	16	–
Calcite	2	–	–
Kaolinite	4	3	Trace
Illite	1	10	14
Chlorite	2	1	–
Potassium feldspar	3	–	3
Plagioclase	–	12	17
MW*	98.94	149.47	156.89

* Average molecular weight of the rock in g/mol.

Table 2

Ions weight percent in sandstones.

Sandstone	Fe^{+3} (wt%)	Ca^{+2} (wt%)	Al^{+3} (wt%)	Mg^{+2} (wt%)	Total (wt%)
Berea	0.23	1.02	0.18	0.46	1.89
Bandera	0.12	4.37	0.09	2.45	7.03
Kentucky	0.00	1.26	0.00	0.26	1.52

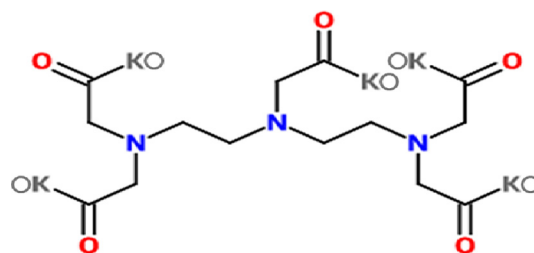


Fig. 1. Chemical structure of DTPA Chelating Agent [17].

Table 3

Stability constants of DTPA with different cations [20].

Ions	Al^{+3}	Ca^{+2}	Fe^{+3}	Mg^{+2}
Log K values	18.6	10.8	28	9.3

Table 4

Oil composition.

Component	Mol%
C ₅	1.23
C ₆	4.23
C ₇	10.66
C ₈	15.81
C ₉	14.51
C ₁₀	14.43
C ₁₁	11.15
C ₁₂₊	27.98
Total	100%

structure of DTPA is shown in Fig. 1. The molecular weight, pH, and density of DTPA ($\text{C}_{14}\text{H}_{18}\text{N}_3\text{O}_{10}\text{K}_5$) was 583.8 g/mol, 11, and 1.25 g/cm³, respectively. The stability constants of DTPA are given Table 3. The oil used was Arabian medium oil and its composition is listed in Table 4. The oil density, gravity, and viscosity was 0.87 g/cm³, 31, and 13.1 cP, respectively. Fluid properties are given in Table 5.

2.2. Inductively coupled plasma-mass spectrometry (ICP-MS)

The change in the mineralogy of the sandstone rocks sample by DTPA and seawater injection was monitored using inductivity coupled

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