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Review paper

Low Salinity Waterflooding in sandstone- A Review

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A b s t r a c t

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Low salinity waterflooding is an emerging EOR technique in which chemistry play major role in improved oil recovery. Numerous laboratory tests from many research groups and some field applications confirmed its effectiveness in enhanced oil recovery. Various mechanisms have been suggested since its appearance to explain the mechanism of the process. Nevertheless, none of them could explain and predict all aspects of the process and possibility of enhanced oil recovery and yet, its mechanism is matter of debate. This paper provides a comprehensive review of low salinity waterflooding. Attempt is made to cover all aspects and features of low salinity waterflooding to shed light on critical and challengeable features and clear the gaps and deficiencies of conducted studies. The proposed mechanisms are discussed and their success and failure are explained. Analytical and numerical modelling of low salinity waterflooding is presented. Both approaches via conventional simulators and geochemical modelling are reviewed. The pore-scale investigation trend is also addressed as a new approach to unveil fundamental phenomena behind the process. Secondary and tertiary low salinity waterflooding are compared in the term of additional oil recovered. Surface forces and rock/fluid/brine interaction and its relationship to wettability are discussed. Results of study of combined low salinity and EOR methods are described which includes simultaneous use of low salinity with polymer flooding, surfactant flooding, CO_2 flooding and also hot water injection. Field applications are demonstrated and necessary conditions to achieve low salinity oil recovery enhancement are discussed. Based on this review, it seems that multi ion exchange and double layer expansion both can contribute in oil recovery enhancement due to low salinity water injection. In fact both proposed mechanisms can reduce oil film thickness which in turn releases more oil. In fundamental point of view, any mechanism that alters surface forces (DLVO theory is starting point) can lead to oil detachment. The process not only is low cost in operation but also can reduce the amount of EOR agents (surfactant, polymer, etc..) and their effectiveness when it combine with other EOR techniques. Accordingly low salinity waterflooding-EOR methods have great potential for enhanced oil recovery in future.

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1. Introduction

Waterflooding is a technique that has been used for a long time to maintain the pressure of reservoir pushing oil from injection well to production well. However the process was physical and no attention was taken to the composition of injected water. Low salinity waterflooding is an emerging EOR technique in which chemistry play major role in improved oil recovery. Improvement of oil recovery by fresh water is observed by Bernard [1] for the first time. But it has not grabbed attention of oil industry till extensive and systematic studies by Morrow's research group confirmed the potential of low salinity waterflooding.

Numerous laboratory tests from many research groups and some field applications confirmed its contribution in enhanced oil recovery. Various mechanisms have been suggested since its appearance to explain the behaviour of the process. Nevertheless, none of them can explain and predict all aspects of the process and possibility of enhanced oil recovery and yet, its mechanism is matter of debate.

In this review, attempt is made to cover all aspects and features of low salinity waterflooding to shed light on critical and challengeable features and clear the gaps and deficiencies of conducted studies. Accordingly, this review provides a basis for further development and defining research topics.

2. Mechanism

As mentioned earlier, Bernard [1], for the first time, noticed importance of fresh waterflooding in oil recovery improvement. He explained the oil recovery enhancement due to sweep efficiency improvement resulted by clay hydration. Clay hydration leads to clay swelling results in pore throat plugging hence reduce pore volume available for oil and water and consequently enhancement in oil recovery.

The first mechanism to explain how low salinity water injection enhances oil recovery was proposed by Tang and Morrow [2]. They found fines in effluent in low salinity water injection. Moreover, when they stabilized the fine by firing at 800 C, low salinity effect was vanished. Consequently, they proposed oil recovery enhancement based on fine migration. The oil drops adhered to clays at pore walls in initial condition. By injection of low salinity water, mixed –wet fines are stripped from the walls and are produced with adhered oil. In fact, when the salinity reduces, the electrical double layer in the aqueous phase between particles is expanded and the tendency for stripping of fines increases and so oil recovery increases.

Albeit it is shown that fine migration is not the main mechanism of enhanced oil recovery in low salinity waterflooding, yet it can enhance it and have contribution in incremental oil recovery partly [3].

A mechanism in which reduction of IFT is responsible of incremental oil recovery was proposed by McGuire et al. [4]. As a result of low salinity water injection, hydroxyl ions are created due to reactions with reservoirs minerals which in turn pH increases. Therefore low salinity acts as alkaline flooding that reduces interfacial tension between oil and water. Likewise, the elevated pH raises reservoir water wettability. In addition, the crude oil acid or polar components are saponified due to contact with raised pH and act as surfactant as shown as follows:

$$(RCOO)_3\ C_3\ H_5 + 3\ NaOH \Longrightarrow 3\ (RCOONa) + C_3H_5\ (OH)_3$$

$$\text{Fat} + \text{alkali} \Longrightarrow \text{soap} + \text{glycerol}$$

Beside these, they pointed out that hardness ions such as calcium and magnesium will precipitate the surfactants in high salinity water injection, hence prevent increasing oil recovery. The reaction takes place as follows:

$$2(RCOONa) + Ca\ (HCO_3)_2 \Longrightarrow (RCOO)_2\ Ca + 2\ (NaHCO_3)$$

$$\text{Soap} + \text{hardness} \Longrightarrow \text{insoluble soap curd}$$

Since low salinity has low concentration of these types of cations, the surfactants keep effective. Various Single well tests in North Slope fields showed that residual saturation decreased between 4 to 11% and oil recovery increases by 6 to 12%.

From an experimental study, Lager et al [5] noticed that concentration of Ca^{2+} and Mg^{2+} in effluent is less than that in invading brine which indicating these cations are absorbed by matrix rock. They argued that based on extended DLVO (Deryaguin-Landau-Verwey-Overbeek) theory, eight different mechanisms of organic compounds adsorption to clay is possible which four of them are strongly affected by cation exchange in low salinity waterflooding. As depicted in Figure 1 they are cation exchange, lignad bonding and cation and water bridging. They proposed Multi ion Exchange (MIE) responsible of adsorption and desorption of organic compounds on clay minerals.

Cation exchange occurs when quaternized nitrogen or heterocyclic ring substitute with exchangeable metal cations which initially attached to clay surface. If cation acts as a bridge between exchangeable cations on mineral surface and functional group a weak interaction is created known as cation bridging. In the case of direct bonding of carboxylate group and multivalent cations Ligand bonding is created which is stronger than cation bridging and cation exchange bonds and lead to detachment of organo-metallic complexes from clay surface. Water bridging will occur if exchangeable cation is strongly solvated (i.e magnesium). It involves water molecules solvating the exchangeable cation and the polar functional group of the organic molecule.

Organo-metallic complexes are formed at the clay surface promote oil wetness in reservoirs. The organic materials may also adsorb directly to the mineral surface by displacing the most labile cations present at the mineral surface and thus enhance the wettability alteration toward more oil-wet. When MIE takes place, organic polar compounds and organo-metallic complexes from the mineral surface are removed by replacing them with cations. Desorption of these polar compounds from the clay surface leads to a more water-wet surface, which in turn enhances the oil recovery. The order of affinity of several ions for the clay sites is:

$Li^+ < Na^+ < K^+ < Rb^+ < Cs^+ < Mg^{2+} < Ca^{2+} < Sr^{2+} < Ba^{2+} < H^+$.

Cation exchange capacity is function of pH and ion concentration.

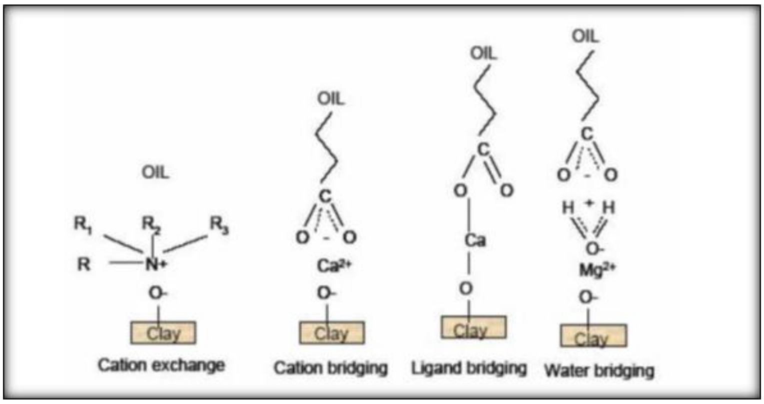

Fig.1: Four mechanism of organic compounds adsorption to clay, cation exchange, lignad bonding and cation and water bridging [5].

Ligthelm [6] proposed double layer expansion as the main mechanism that is responsible of enhanced oil recovery due to low salinity injection. Multivalent metal cations such as Ca^{2+} and Mg^{2+} act as bridge between negatively charged clay minerals and oil compounds. In high salinity condition, high amount of positive cations suppress the electrostatic repulsive forces. This makes the clays more oil-wet. Injecting of low salinity water (especially multivalent cations) yields expansion of double layers which in turn increases repulsive force between the clays and oil. When repulsion exceeds binding forces, oil particles desorbs from the clay surface. By further reduction in electrolyte concentration, the mutual repulsive forces in clay minerals starts exceeding the binding force resulting clay deflocculating and formation damage. However, Austad et al [7] indicated that polar oil components can adsorb onto clay surface without bridging of cations such as Ca^{2+}.

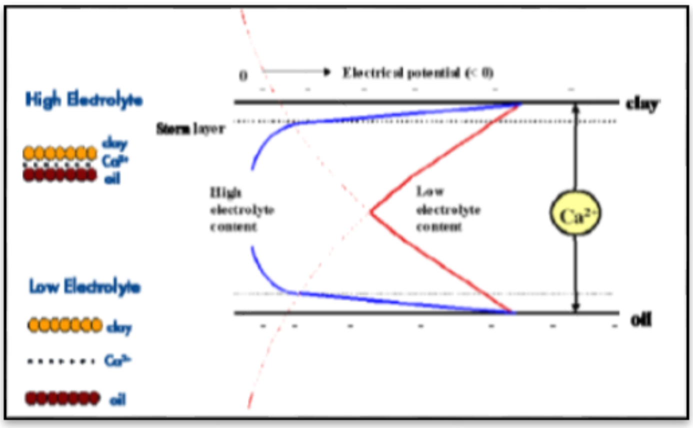

Fig.2: Double layer model [6]

Another mechanism was proposed by Austad et al. [7]. Having large surface area, the clay acts as cation exchanger. Inorganic cations, especially Ca^{2+}, and acidic and basic organic materials are bonded onto the clay in established equilibrium at reservoir conditions. In the case of low salinity waterflooding, the brine-rock equilibrium is disturbed and release of cations, Ca^{2+} in particular, happens. Protons H^+ are replaced by Ca^{2+} results in an increase in local pH. As an illustration:

$$\text{Clay-}\ Ca^{2+} + H_2O = \text{Clay-}\ H^+ + Ca^{2+} + OH^+$$

The increase in local pH results in proton transfer reaction of basic and acidic materials.

$$\text{Clay-}NHR_3^+ + OH^- = \text{Clay} + R_3N + H_2O$$

$$\text{Clay-}RCOOH + OH^- = \text{Clay} + RCOO^- + H_2O$$

Experimental works showed that none of the above suggested mechanisms can explain all phenomena observed in different experiments. They obtained the results that show no additional oil recovery in spite of significant fine production indicating that fine mobilization is not main mechanism of oil recovery. Another experiment showed pressure drop increase without additional oil recovery rejecting flow diversion mechanism [8].

Recently Brady and Krumhansl [9-10] proposed a surface complexiation model in which a diffuse-layer model is used to account for double-layer interactions at kaolinite oil surface. The model is based on diffuse double layer and pH-dependent surface charge. Surface charges include both kaolinite basal-plan and edge interaction with oil. The most abundant charged oil-surface complex are calculated to be $–COO^-$, $-NH^+$ and $–COOCa^+$ and the most abundant kaolininite-edge complex is $Al:Si\text{-}O^-$. They assumed that oil adhesion take place by oppositely charged species in oil and kaolinite surfaces. The concentration of the mentioned complexes and their products in both clay and oil is calculated as function of pH. They stated that the waterfloods that reduce the number of oppositely charged products concentration should enhance oil recovery. Regarding oil kaolinite-edge interaction, they found that below $pH<5\text{-}6$ low salinity injection decreases the number of $[-NH^+][>Al:Si\text{-}O^-]$ pairs which increases oil recovery. Moreover, above pH 5.3 the number of [-$COOCa^+$][>Al:Si-O^-] pairs decreases due to low salinity injection which in turn favors oil recovery (although it is balance out by increases of [-NH^+][>Al:Si-O^-] to some extent). Their semi quantitatively calculation showed that low salinity waterflooding will increase exchange of –NH^+ onto basal plane while reduces –$COOCa^+$ exchange to basal plane. The net calculated effect due to low salinity waterflooding will be enhanced oil recovery in pH<6 and >7.

Due to similarity in eqilibria of Ca^{2+} and Mg^{2+}, they argued that effect of Mg^{2+} is same as Ca^{2+} . Based on their calculations, the sandstones that contain carbonate minerals carbonate dissolution dumps the effect of low salinity waterflooding. According to similarity in basal plane exchange capacity of kaolinite, ilite and chlorite, their responses to waterflooding is estimated similar.

Based on simulation model (their model is not, in fact, a mechanistic model, residual oil saturation and relative permeabilities are considered as function of brine salinity) and summary of 411 coreflooding experiments, Aladasani et al (2102) concluded that low salinity waterflooding recovery mechanism is function of initial and final states of wetting properties. In weak water-wet conditions, low capillary pressure is responsible of incremental oil recovery of LSWF. In the case of weak oil-wet conditions, LSWF works due to increased oil relative permeability and change of the non-wetting phase to oil. But in strong oil-wet and water-wet conditions, LSWF mechanism is increased oil relative permeability alone.

Researchers regarding pore scale studies pointed out that double layer expansion is main mechanism of low salinity waterflooding [11-15]. In the same approach, Berg et al [16], argued that double layer expansion or ion exchange are the mechanisms and more studies are needed to reach final idea (see section pore scale study).

It is observed that the samples containing low amount of kaolinite respond to low salinity waterfloopding greatly. In these conditions mineral dissolution was considered as main mechanism of process enhancement resulting higher sweep efficiency [17].

Other mechanisms such as mineral dissolution, osmotic pressure, salinity shock…have been proposed [18] that have been less discussed as responsible mechanism.

Lessons from other fields of sciences and technologies will help us to understand interaction of oil/brine/rock system. There are some studies that unveil the role of clays in release of bitumen from sand. Taking the lessons from these researches is useful since the mechanism behind it is same as that behind low salinity waterflooding in nature. Study of Clark and Pasternak [19] showed presence of 2-10% clay in sample containing bitumen decreases release of oil by Clark Hot Water Extraction process. Coagulation of clay-bitumen is enhanced by presence of multivalent ions such as Ca^{2+}, Mg^{2+} and Fe^{2+}. Kaolinite coagulate with bitumen greatly in pH less than 6. But increase in pH results in lower coagulation. Presence of 10^{-3} mol/l of multivalent ions causes complete coagulation of bitumen with kaolinite over the entire pH. By measuring of zeta potential and coagulation density, they argued that surface charges govern the process. On the other hand, citric acid addition decreases coagulation of kaolinite significantly. In their discussion adsorption of citrate anions on the surface of kaolinite was introduced as reason of lower coagulation [20].

It is reported that particle coagulation enhances when the cations are in form of mono hydroxyl species such as $CaoH^+$ and $MgoH^+$ indicating hydrolysis and surface adsorption is responsible of coagulation. The edge and basal planes of mineral responds to cations differently. Yan et al [21] measured zeta potential of talc and muscovite at various Mg^{2+} and Ca^{2+} concentration at pH 8.5 for edge and basal planes separately. For both planes, zeta potentials increase when divalent ions concentration rises. Double layer compression was recognized responsible of basal plan charges variation and adsorption dominated the edge surface changes. Stronger impact of Ca^{2+} than Mg^{2+} was discussed to less hydrated characteristic of Ca^{2+} compared to Mg^{2+}.

3. Surface forces and low salinity waterflooding

DLVO is fundamental theory that is starting point for understanding the behaviour of colloidal systems [22]. Classical DLVO theory contains two force components, electrostatic (diffuse double layer) and Van Der Waals terms. While the extended DLVO contains the term named structural force.

3.1. Electrostatic (diffuse double layer)

Particles and plates carry both permanent and induced charges. This surface charge induces an electrostatic field combined with the thermal motion of the ions, creates a counter charge, which cover the electric surface charge. Electric potential decrease almost exponentially, which based on "Debye length" is introduced which mainly depends on "ionic strength" [23]. When double layer surfaces reach each other, electrostatic interaction will occur. These interactions can be either attractive, repulsive. The forces are dependent of the pH and salinity in the aqueous phase.

3.2. Van der Waals

The van der Waals force is the second component in the DLVO theory**.** While this force not as strong as electrostatic or H-bonding interactions, they are always present and plays an important role at small and large distances. This attractive force is a function of the Hamaker constant. Hamaker constant is not in fact constant and it varies depends on conditions [23].

3.3. Structural force

In many cases classical theory has not been capable to explain the surface-related phenomena. As a result, the theory was upgraded to extended DLVO theory which contains third term known a structural force [22]. When the distance between oil/brine and brine/solid interfaces are separated by a distance of few molecular diameters, the short range interactions appearing in the system is described by short range forces. These forces are repulsive forces and are called solvation, structural or hydration forces (hydrogen bonds) when the medium is water, and are a result of the intermolecular structure of the water.

These three forces constitute surface forces that when two particles or plates reach each other interact due to them. These forces cause adhesion of oil/brine to mineral. Contact angel increased with increase of adhesion to surface [24].

3.4. Zeta potential

As stated before, zeta-potential is the electrostatic potential at the interfacial layer located at the slipping plane which is measurable parameter. The magnitude of the zeta-potential gives an indication of the potential stability of a colloidal system, *i.e.* crude oil/brine or crude oil/solid system. The pH at which the total net charge is zero is termed the Zero Point of Charge (ZPO). It determines the pH at which minerals exhibits cation or anion exchange. In other pH, depends on sign of charges, force between surfaces may be attractive or repulsive.

It is reported that kaolinite carried negative charges in the tested pH range from 3 to 12. With increasing pH from 4 to 8, the zeta potential became more negative and above pH 8 it stabilized at about –50 mV. In the presence of Ca^{2+}, zeta potential became much less negative between pH 6 and 12. Approximately, the same trend followed in presence of Mg^{2+} from pH 6 to 10. However, above pH 10, it soared steeply and reached zero at pH 10.4, then took positive value in higher pH [20].

Experimental study by Min [25] also showed zeta potential gradually increases and reaches its maximum at pH 8 and then tends to decrease with the pH value increasing. With Ca^{2+} concentration increasing, the absolute value of the surface zeta potentials of kaolinite gradually decreases.

The suspension particle surface ion adsorption and the change of surface bonds of Si–O, Al–O and Al–OH are the underlying reasons to change surface zeta potential of kaolin suspension particle. Repulsive energy between kaolinite-kaoloinite particles was more than that between kaolinite-quartz particles.

Buckley et al [26] measured zeta potential of crushed silica glass as function of pH and salinity. Increasing of pH reduces zeta potential of solid surface and lower NACL concentration decreases it as well. The measured zeta potential value for crude oil cleared that ZPO take places in PH around 4 for various salinity. In pH higher than 4, zeta potential became negative and for pH lower than 4 positive. Lower salinity made zeta potential more negative in pH higher than 4 and more positive in pH lower than 4.

Nasralla et al. [14] measured zeta potential at rock/brine and oil/brine interfaces. Results showed that zeta potential was positive at pH less than 3 then reached zero at pH 3-6. At pH higher than 6, it become negative and the more the reduction of water salinity, the higher negatively charged the oil/brine and rock/brine interfaces. In another study, they found that injection of low salinity water changes brine/mineral to more negative charges which generate repulsive force resulting in release of more oil at pH higher than 7. In this study, however, they considered cation exchange responsible of additional oil recovery in low salinity waterflooding [27].

3.5 Mechanism of oil/brine/mineral interaction

Buckley and Liu [28] argued four ways of oil/brine/mineral interactions by which wetability is governed.

- **Polar interaction**: This interaction takes place between polar functional group of oil and polar surface sites if mineral under clean and dry surface conditions.
- **Surface precipitation**: If the oil does not have high level of solubility for the asphaltenes, the system trend for wetting alteration increases.
- **Acid/base interactions**: In the presence of water, Polar functional groups of both mineral and crude oil phases can behave as acids and bases creating charges in both the solid and oil interface. PH is the main governing factor of the level of surface charge.
- **Ion –binding interactions**: The ions such as Ca^{2+} can acts as a bridge making three possible interactions. Oil-Ca-oil, mineral- Ca-mineral and oil-Ca-mineral. The first two can limit wettability alteration while the last promotes it. PH is not the essential factor and temperature is supposed to affect interactions greatly.

In a oil/brine/rock system, more than one interaction occurs. Surface precipitation is less likely to take place due to usual presence of water as first fluid occupying the pore space. Roughly, combination of three properties, API gravity (solvent quality of the oil) acid and base numbers give a qualitative index for wetting alteration.

But experimental study by Basu and Sharma [29] showed that polar components (resin and asphaltens) play an important role in disjoining pressure of brine film between mineral and oil. In presence of resin and asphaltens, film stability decreases with increasing brine salinity and decreasing of pH while without them increasing the pH and salinity leads to more stable film. The later consequence was not in agreement with calculation of DLVO theory as authors suggested the role of variation of hydrophobic force with salinity.

Silica surface adhesion experiments showed that higher pH (pH=8) results in more water-wet condition than lower pH at 4 in same brine condition. Behaviour of silica was same as core containing quartz [30].

The wetted surface increase the time of adsorption compared to dry surface and usually occurs in two stage of fast adsorption followed by slow adsorption. Increase in temperature leads to longer adsorption time [31].

Oil film stability is function of pH. An experimental evidence of such a relationship was shown by Liu and Buckley [32] for both mica and glass, oil film is stable at most of the range of pH. By decrease in salinity it oil film become stable/conditional stable at less range of pH.

Buckley et al [26] showed that DLVO theory can predict the behaviour wettability and adhesion of crude oil to glass surface. Except, in some conditions lack of data to assess the value of hydration force resulted in fail of the theory to match with experimental results.

4. Wettability effects and its alteration

It is believed that enhanced oil recovery in low salinity waterflooding is related to wettability change. Furthermore there is direct evidence that wettability altered to more water-wet during low salinity waterflooding [14]. Accordingly, initial and final wettabilities are key indices which have been evaluated. However, results are controversial.

The initial oil-wet condition is essential to observe low salinity effect [33-34] since water-wet sample produced no additional oil [34]. Even, as Hadia et al [35] reported, samples of initial neutral-wet does not show additional oil recovery in low salinity waterflooding while those with initial oil-wet shows. As well, it is observed that the more initial oil-wet condition cause better oil recovery even the clay content being low [36].

The type and level of salinity water injection is important to create wettability of a reservoir. In presence of plagioclase mineral, higher salinity water cause PH below 7 which in turn create mixed-wet condition which is favourable condition for low salinity waterflooding. On the other hand, in moderate low salinity water injection this mineral causes pH>7 which create unfavourable water-wet state [37].

The desired final wettability is also matter of debate. Some researchers reported that low salinity waterflooding change the wettability into neutral state rather than weter-wet state [38-39]. Ashraf et al [40] pointed out same results, but they also reported that highest S_{or} reduction took place in resulted water-wet condition.

It is argued that although residual saturation in low salinity flooded water-wet condition is higher than that mixed-wet condition, but higher oil relative permeability leads to net preference of final water-wet condition than the final mixed-wet condition [41].

It is clear that change in wettability correspond to change in relative permeabilities curve which in turn results in change in fluid flow in porous media. Experiment by Webb et al [42] showed due to low salinity water injection ,the relative permeabilities curves have similar motilities with the previous ones, but are shifted in saturation, benefitted the low salinity waterflooding resulted in incremental dry oil recovery. They attributed this quick kinetically oil recovery to be in consistent to the double layer/multiple ion exchange theory.

5. Pore scale studies

Pore scale studies have been done by many researchers, as a more directly approach, to find impact of low salinity water injection in wettability state or surface forces of mineral/brine/oil system.

Berg et al [16] performed microscopic experiment to look for mechanisms involved in low salinity waterflooding. Oil droplets were located on clay mineral (especially montmorillonite) and the system exposed to flow of composition designed water. They point out that the emulsification, IFT reduction, fines migration and selective plugging of water-bearing pores via clay swelling are discarded as main relevant enhanced oil recovery. Additional oil recovery due to low salinity waterflooding was attributed to wettabillity modification, but there was not any evidence to distinguish between double layer expansion and cation exchange or detachment of clay layer as responsible mechanism. They also reported that at least in reservoir containing montmorillonite there is an optimum low salinity concentration that lower than that the clays are swelled result in formation damage.

A pore scale study by Hassenkam et al [13] deal with the determination of the level of adhesion force between water and crude oil by Atomic Force Microscopy (AFM). In this imaging technique, AFM tips are functionalized by organic compounds passing above a surface in aqueous solution of various salinity while adhesion force between them is recording. For both outcrop and reservoir cleaned samples the experiments showed the low salinity decreased adhesion force from 20% to 45%. However in one set experiment low salinity didn't change the adhesion force. Through a discussion, they concluded that their results are in consistent with van der walls dispersion forces in combination with the effects of the electrical double layer and possible ion bridging.

A study in which effect of salinity on contact angel investigated carried out by Nasralla et al [14] revealed that reducing salinity decrease crude oil contact angel with mica (both Moscovite and Biotite). Except for seawater that with salinity of 55000 mg/l has high contact angel of 76° (formation brine with salinity of 174000 mg/l showed contact angel of 60°), decreasing the salinity of water reduced the contact angel. These experiments directly show that lower salinity make the rock more water-wet. They also showed that decreasing the pressure decreases oil droplet contact angel indicating altering wettability of mica to more water-wet. Furthermore, increasing the temperature made mica less water-wet. They stated that wettability alteration is result of double layer expansion but not mechanism of incremental oil recovery. Double layer expansion is argued as primary mechanism of additional oil recovery. As well, through an experimental study by Mahani et al. [15], it was found that low salinity brine reduces contact angle (or in other word increase wettability to more water-wet) followed by detachment of oil drops at critical angle of 40-50º. Detachment time for three different low salinity brines was same. A diffusion and electrokinetic mechanism is supposed to predict time scale of the process. The mechanism support double layer and surface charge effects. There is evidence that relate contact angel and water film thickness which support double layer expansion mechanism. The smaller contact angle was found to correspond to thicker water film [43].

Small angle scattering experiments by neutron and x-ray cleared that the thickness of water film increases with a fall in salinity. Effect of divalent ions is more profound than the monovalent ions. These observations are explainable by double layer expansion mechanism. Interestingly, however, water film thickness reduced in case of fresh water that is inconsistent with the double layer expansion model [11].

6. Secondary versus tertiary mode

Both secondary and tertiary low salinity waterflooding contribute to incremental oil recovery [44]. But usually secondary injection is more desired,[45] and in some conditions tertiary low salinity water injection didn’t result in extra oil recovery [27,39].

7. Effect of brine composition

An extensive study by Shell researchers [33] examined the role of formation composition and imbibing brine as well as type of crude oil in low salinity imbibition experiments. They found that presence of Ca^{2+} and Mg^{2+} in formation brine makes the sample more oil-wet and also the more Ca^{2+} and Mg^{2+} concentration in formation brine, the more oil-wet behaviour the samples show. An interesting finding was that the samples with high level of Ca^{2+} and Mg^{2+} when surrounded by high and low Nacl salinity exhibited same oil recovery. On the other hand, the sample with Na^{+} concentration in formation brine didn't show any additional oil recovery even upon exposed to very low salinity brine (0.01 diluted Dagang brine). Some other imbibition experiments using different crude oil indicated those samples aged with crude oil that make the samples more oil-wet produced more incremental oil in the case of low salinity imbibition. From the results mentioned, they argued that the more initial oil-wet the samples have, the more oil available for low salinity waterflooding.

8. Effect of/on core mineralogy

Presence of clay minerals is essential for having a good potential of low salinity waterflooding. Review of 411 coreflooding experiments revealed a strong correlation between residual oil saturation to chlorite and kaolinite contents and wettability index as well [46]. The experiments by Wickramathilaka and Morrow [47] confirmed this type of correlation.

Low salinity waterflooding changes the composition of cores and associated rock properties. Experimental study of Pu et al [48] showed the cores that are rich in anhydrite cement, low salinity waterflooding dissolved the cements. Presence of sulphate ions in effluent water and thin section pictures confirmed this conclusion. However it is not clear that is a cause, effect or accompany phenomenon.

During low salinity waterflooding permeability increase of 100% was happened, [49]. On the other hand 10% decrease of permeability was observed in case of 100 diluted water flooding.

8. Effect of temperature

Effect of temperature seems complicated. Flooding tests of samples from Varg field showed that low salinity waterflooding effect is observed when aging temperature was 90 C while no extra oil was recovered when aging temperature was 60 and 130 C [50].

As well, impact of temperature showed varies from substantial impact on contact angle of crude oil with mica to non-significant in muscovite and biotite surfaces pre-treated with pH 4, [NaCl] = 0.1 *M* brine Buckley [51].

Low salinity waterflooding illustrated more sensitive than high salinity waterflooding. Higher aging and flooding temperature resulted in higher oil recovery due to low salinity waterflooding compared to high salinity waterflooding [52].

9. Effect of low salinity on Interfacial Tension

Exploring the impact of salinity on interfacial tension is crucial since a mechanism is proposed by McGuire et al. [5] indicating lower salinity water reduced IFT between water and oil which in turn enhances oil recovery.

Water/oil interfacial tension is greatly affected by oil composition. Large number of components in crude oil makes difficult to predict IFT of water/oil. According to Hughey et al [53], there are around 11000 different components in one crude oil. Undoubtedly, this number for all types of oil will be considerably higher. In general, it can be stated that value of IFT is governed by many parameters and it is not fully understood.

Buckley and Fan [54] measured the IFT of crude oil and made a correlation between IFT and some crude oil parameters. Based on statistical analysis, they proposed a correlation indicating higher amounts of n-C_7 asphaltenes, base number and viscosity of crude oil results in higher value of IFT. On the other hand, higher values of pH and acid number lead to lower value of IFT.

Controversial results have been reported in literature regarding effect of salt concentration on IFT. Salinity increase in aqueous phase in the case of Athabasca bitumen resulted in a decrease in IFT [55]. Same result was observed in experiments carried out by Cai et al. [56].

Likewise, Okasha and Al-Shiwaish [57] observed same behaviour for both dead and live oil of Arab-D carbonate formation, Sudi Arabia.

Based on study of Kumar [58], the more salt in pure hydrocarbon, the more the IFT between oil and brine. In case of crude oil, different behaviours were observed. Increase in salt concentration resulted in increase in IFT of medium crude oil. On the other hand, IFT of heavy crude oil diluted with decane decreased with increase in salinity at any given concentration of decane.

Experimental study on dead oil of Yates field revealed that with dilution of the brine, the IFT decreased initially, but it increased by further dilution of brine. Accordingly, a critical salinity was recognized in which the IFT has the lowest value[59]. An optimum IFT (lowest value) in 5% NACL brine concentration also reported by Alotaibi and Nasr-El-Din [60].

Type of brine composition affects the IFT. Presence of $CaCl_2$ increase the IFT compared to NaCl or formation brine composition [61]. While, No difference was observed between effects of three salts sodium chloride, Calcium chloride and magnesium chloride [56].

10. Necessary condition for low salinity EOR effects

Based on systematic experiment by Tang and Morrow [2], the necessary condition to observe low salinity oil recover enhancement are presence of mobile fine clay, polar components in crude oil and initial water saturation. Moreover, many experiments showed that oil-wet or mixed-wet initial condition is crucial for applicability of low salinity waterflooding (see section). However, these requirements are not without exception.

As a general rule, it can be stated that presence of clay is essential for a formation to have potential of low salinity waterflooding. Although there is a exception for this rule [62], it is observed that incremental oil recovery is function of percentage of formation clay. Yet the relationship is not simple and type of clay is important. Clays with positive zeta potential and those swell with fresh water are detrimental for the process [63]. Kaolinite is a clay that is effective to enhance oil recovery for the process even though its cation exchange capacity is low.

By simulation using Phreeqc , it is found that low salinity injection into rock/oil/brine system in which low salinity waterflooding give lower concentration of divalent onto clay surface, may be indication of good candidate for low salinity waterflooding [41].

11. Analytical and numerical modelling

Two approaches have been considered to model low salinity waterflooding. In the first one, the mechanism of rock/oil/brine interaction is ignored and just relative permeability, capillary pressure or other main parameters are functioned on salinity concentration.

In this approach it is showed that modelling of low salinity waterflooding can be considered same as other waterflood-based EOR in which fractional flow equation should be modified to reach appropriate model for process [64].

Wu and Bai [65] modelled low salinity waterflooding mathematically and numerically for both sandstone and fractured media. Salt was considered as a component transported and contained only in the aqueous phase. However, the salt can absorb on rock grains and transportation take place by advection and diffusion. Both relative permeability and capillary pressure were modelled as function of salinity that results in enhancement in oil recovery. The analytical equations were discretized to reach numerical formulation and were implemented in a general simulator MSFLOW. The simulator was matched successfully with experimental results.

Without considering the mechanism behind the process, [66] coupled UTECHEM and Iphreeqe simulators and tested it. To model wettability alteration relative permeability and capillary pressure are interpolated based on two sets of carves. UTECHEM was used for species transportation and Iphreeqe to find rock-fluid equilibrium state. Consequently he relative permeaqbility and capillary pressure curves are constructed.

In the second approach, rock/oil/brine (geochemical) interaction is modelled to results in wettability alteration which can be regarded as mechanistic model.

A numerical model that couples a Buckley-Leverett two-phase model and multiple ion exchanges as the main mechanism of wettability [67]. The model is created for one dimensional low salinity waterflooding suitable for lab experiments. The ions calcium, magnesium, and sodium can desorbs or adsorb to the rocks that in turn determine the wettability and new relative permeability curves. The cations release depends on several factors such as clay content and connate water and injected brine composition.

In the same approach, Omekeh et al. [68] developed a mathematical model for one dimensional experimental study. The model is based on dissolution/precipitation of various carbonate minerals and ion exchange between clays and aquatic phase that affect relative permeability and consequently the amount of oil recovered. The model was successfully matched with experimental observations in terms of ion concentration and pH and oil recovery. It was demonstrated that carbonate chemistry may decrease the beneficial impact of low salinity waterflooding.

12. Field application and study

Some field scale, both single well and double wells, experiments have been conducted worldwide. As well, some reservoir samples were waterflooded in reservoir conditions. Results were promising while some showed no incremental oil recovery.

Webb et al. [44] tested low salinity in a production well in a giant field in Middle East. Pulsed neutron capture log was used to determine oil and water saturation. Low salinity waterflooding improved oil recovery considerably. Its effect depends on perforated intervals. At top perforated interval 50% decrease in remaining oil was observed, while at the middle and bottom 10-20% reduction was reported.

By investigating of historical field data of some waterflooding projects in Wyoming state USA, Robertson [69] showed that lower ratio of injected brine salinity to formation salinity resulted in higher oil recovery.

Lager et al. [70] presented results of their field application of low salinity waterflooding in single hydraulic unit in an Alaska oil field in May 2005 till May 2006. Injection switched from high salinity injection of 16640 ppm to non-optimized low salinity, as mentioned by authors, of 2600 ppm. Oil production rate increased from 150 bbl/d to a peak of 320 bbl/d followed by a decrease to 200 bbl/d while the water cut dropped from 92% to 87% at the same time. Due to constant water injection, they inferred that no fine generation took place. Chemistry of produced oil shows decline in Ca^{2+} and especially Mg^{2+} ions. Recording dramatic decline in Mg^{2+} ion, constant injectivity and negligible change in PH, they attributed the low salinity effect to the MIE mechanism. However, they mentioned that the used non-optimized low salinity brine reduced S_{or} by 2% and in the case of the optimized low salinity brine 10% reduction in S_{or} is achievable. More study indicated that 40% slug of low salinity water is more effective than continuous waterflooding (Seccombe et al, 2008). A low salinity waterflooding test involving an injector and producer 1040 feet apart in the same field showed a reduction in water-cut from 95% to 92% and 10% OIP incremental oil recovery with 28% expected decrease in residual oil saturation [62].

A field test of low salinity in the Omar field in Syria showed the rocks are changed to more water-wet condition. The indicators of this change were direct connate water banking measurements and dual steps in water-cut development. These observations were supported by lab-scale spontaneous imbibitions and Log-Inject-Log field test [72].

Thyne and Gamage [73] conducted low salinity waterflooding of cores from Minnelusa formation, Wyoming, USA. No additional oil recovery was reported from these experiments by low saline water. Analysis of waterflooding of many fields from same formation showed that some of them flooded with low salinity water with no sign of incremental oil recovery due to lower salinity waterflooding. They concluded that lack of additional oil production was due to lack of mobile clay fines. This conclusion was made since experiments with same oil and water using Berea sandstone, which contained mobile fines, resulted in incremental production [45].

Laboratory low salinity waterflooding of plugs from an extensively faulted and heterogeneous western Africa field showed a 5-8% incremental oil recovery compared to high salinity one. However, after simulated history matching of historical water injection and forecasting for low salinity injection, additional oil recovery of 2-3.4% was predicted [74].

An extensive study has been done to find the potential of low salinity waterflooding in Snorre field, Norway. Laboratory experiments at reservoir conditions resulted in no considerable incremental oil recovery (2% for Statfjord formations sample and 0% for Lunde core) in tertiary state. The pilot test also indicates no difference oil saturation after seawater flooding and low salinity waterflooding. The marginal or no response due to low salinity waterflooding was attributed to proper initial wettability of formations as low salinity waterflooding does not change it to release the attached oil [75].

13. Combined EOR and Low salinity waterflooding

Combination of many EOR methods and low salinity waterflooding can be considered to use their benefit simultaneously.

Of the advantage of surfactant low salinity waterflooding are lower adsorption and retention and higher solubility of surfactant resulting better oil recovery and economic than only surfactant waterflooding [76]. In the case of surfactant low salinity waterflooding, intermediate-wet condition showed more favourable condition than the water-wet condition [77].

Combination of polymer and low salinity waterflooding was examined by Mohammadi and Jerauld [78] numerically. They found that the combined method benefits equal the summation of each technique. In addition, the polymer low salinity flooding reduces the necessary amount of polymer somehow one-third of polymer flooding alone. In fact low salinity condition increases viscosity of solution compared to high salinity condition making it more favourable for a such process. Moreover, it reduces retention of polymer to rock. The same numerical study by Alzayer and Sohrabi [79] shows effectiveness of polymer low salinity waterflooding for heavy oil recovery. Analyses of some experimental data were consistent with above statements [80]. Besides, the low salinity media, polymer is more stable at higher temperature and shear degradation. It also provide less production chemistry issue such as scaling and souring, These all reduce the expenditure for this process compared to conventional polymer flooding. On the opposite side, it cause some disadvantage such as clay swelling and breakthrough delay. Both sides should be considered in economic of the process [80].

Injection of polymer low salinity water in secondary mode (at initial water saturation) is more beneficial than tertiary (at residual oil saturation) mode [81].

An effort is taken by Abass and Fahmi [82] to apply low salinity experiments in extracting heavy oil by hot water injection. Their experiments showed 20%, 29% and 24% (three experiments with heavy viscose oil of 1700 cp @65 C, >1000 cp @ 60 C and 700 cp @ 35 C respectively) OOIP increase by low salinity hot water injection over the high salinity hot water injection. Moreover, 8% incremental oil recovery was observed by steam injection after low salinity hot water injection.

IFT of brine-CO2 was measured at reservoir condition by new technique, It reduces with decrease of salinity indicating decreasing of CO2 solubility in brine [83]. Combining of CO2 and low salinity waterflooding needs many attentions. Higher pressure, lower salinity and lower temperature increases solubility of CO2 in water (see for example [84]). however, considering this rule in all ranges of parameters needs more attention. In fact, there are controversial results in effect of CO2 solubility as well as salinity in oil recovery. In one hand, there are some numerical and experimental studies that shows considering CO2 solubility and also the parameters that enhance it decreases oil recovery in CO2 flooding or WAG–CO2 flooding [85] (numerical study of Co2 flooding), [86] (numerical study of CO2 flooding), [87] (numerical study of WAG-CO2 flooding), [88] (experimental study of WAG and CO2 flooding, especially lower salinity led to lower oil recovery) [89] mentioned no effect of low salinity in CO2 flooding. On the other hand, there are some studies that indicate higher CO2 solubility in water (and considering it in calculation) increases oil recovery, [for example [90] (experimental study of SWACO2 and WACO2 flooding, especially lower salinity led to higher oil recovery), [91] (numerical study of CO2 flooding in carbonate). Beside these, Yan and Stenby [92.93] numerically showed that higher CO2 solubility in water results in higher oil recovery in CO2 flooding but lower oil recovery in WAG-CO2 flooding. Accordingly, this effect is not understood thoroughly and is subject of study.

Kulathu et al [94] proved effectiveness of cycling low salinity waterflooding rather than continuous waterflooding through experimental study. Same residual saturation was achieved by 3-4 pore volume injected in cyclic water injection compared to 6-7 pore volume in continuous water injection.

14. Conclusion

- The starting point to investigate mechanism of the process can be DLVO theory. Although it failed to explain some phenomena, but extended DLVO theory was successful to explain surface-related phenomena [22]. In the same way, classical DLVO theory failed in some conditions to predict behavior oil/brine/rock system [26, 29]. However, it is believed that by considering hydration force the behavior can be explained [26, 29]. By determination of three forces (forces in DLVO equation: van der Waals, electrostatic and hydration) in various conditions the effect of many parameters such as low salinity injection is possible. Hydration force need great attention since there are not too much studies to reveals its value. However, the role of electrostatic is crucial and somehow complicated. Electrostatic force is function of surface charge which itself is function of other parameters such as pH, mineral and oil composition. In fact, three mechanisms of fine migration, Ion exchange and double layer expansion were stated explainable by DLVO theory. Yet, the details, conditions and contribution of each are not clear.

- The conducted researches in other fields such oil extraction from mined oil sands, underground water engineering, soil science, effluent environmental science, paper industry and those industry that clays play important roles can be useful to determine surface complexes and interactions.
- Comprehensive studies are needed to cover all aspects of low salinity waterflooding from atomic and droplet scales to reservoir scale to assess the EOR potential of low salinity waterflooding. Such a workflow is sketched by Suijkerbuijk et al, [43] in three main levels. At single droplet scale the DLVO and non-DLVO forces in rock/brine/oil system should be considered. Channel scale study is needed to quantify hydrodynamics of the process. At the end reservoir scale couple geochemical processes with two-phase flow processes.
- Economic part of the process is important to evaluate the applicability of the process. Low salinity waterflooding is same as waterflooding in process except it has additional unit of desalination to reach proper salinity. Furthermore, environment issue due to release of high salinity water and process by-product sludge should be assessed.
- Although the wettability modification due to double layer expansion or cation exchange is supposed in sandstone as mechanism of low salinity waterflooding, but it is believed that mechanism in carbonate formation shares the same reason except the geochemical reactions occurred in carbonate are different [43].

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