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Effects of pH and Temperature on Oilfield Scale Formation

Jaber Azizi[1], Seyed Reza Shadizadeh[2*], Abbas Khaksar Manshad[3], and Naghi Jadidi[4]

[1] M.S. Student, Department of Petroleum Engineering, Petroleum University of Technology, Abadan, Iran
[2] Professor, Department of Petroleum Engineering, Petroleum University of Technology, Abadan, Iran
[3] Assistant Professor, Department of Petroleum Engineering, Petroleum University of Technology, Abadan, Iran
[4] Assistant Professor, Department of Petroleum Engineering, Petroleum University of Technology, Abadan, Iran

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Abstract

Water flooding is one of the most influential methods for pressure maintenance and enhanced oil recovery. However, water flooding is likely to develop the formation of oilfield scale. Scale formation in reservoirs, due to the mixing of injection water and formation water, could cause formation damage and production limit. Therefore, it is necessary to simulate the compatibility of brine and injection water. Scale prediction is performed using many thermodynamic and/or kinetic based models. In this study, simulations with speciation (ion pairing) are studied, which is a thermodynamic based tool. The utilization of reservoir conditions, formation water analysis, and sea water analysis as the inputs in this method resulted to the accurate prediction of potential scales. In this study, the factors impacting on the scale potential such as pH, temperature, and mixing ratio were also investigated. The obtained results showed that calcite and aragonite were the major scale potential to precipitate. Finally, the results illustrated the important effect of pH and temperature on different scales formation.

Keywords: Scale formation, Oilfield, Water Injection, Scale Prediction, Calcium Carbonate

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1. Introduction

Pressure maintenance by injection water in reservoirs could be considered acceptable for oil recovery. Due to the incompatibility of water injection and formation water, the precipitation of sparingly soluble salts occurs, which is called scaling. Scaling is one of the various factors affecting formation damage. Based on Civan (2011) work, scale formation categorizes into the fluid-fluid interaction that is one of seven group formation damage mechanisms, including fluid-fluid interaction, rock-fluid incompatibilities, solids invasion, phase trapping/blocking, chemical adsorption/wettability alteration, fines migration, and biological activity. The scale is probably among the three largest water associated production issues in addition to corrosion and gas hydrate (Kelland, 2009). Also, the worldwide price of scale continues to be estimated to be more than billions (Frenier and Ziauddin, 2008). Therefore, it is sensible to prevent the formation of scales. For this reason, scale must be predicted and modeled, and scale inhibitors should then be used. The most common scale types to be encountered in an oilfield are in order of prevalence listed in Table 1 (Kelland, 2009; Merdhah and Badr, 2008). The

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* Corresponding Author:
Email: shadizadeh@put.ac.ir

various forms of calcium carbonate ($CaCO_3$) scales differ only in crystal structure (Frenier and Ziauddin, 2008).

Table 1

Most common oilfield scales (Kelland, 2009; Merdhah and Badr, 2008).

Type	**Name**	**Chemical formula**
Carbonate scales	Calcium carbonate • *Calcite* • *Aragonite* • *Vaterite*	$CaCO_3$
Sulfate scales	Calcium sulfate • *Gypsum* • *Barite*	• $CaSO_4$ • $BaSO_4$
Sulfide scales	Iron sulfide • *Pyrite*	FeS_2
Sodium chloride scale	Salt • *Halite*	NaCl

Distinct changes in states cause sulfate and carbonate scales. For example, carbonate deposits predominantly form as a result of decreasing pressure and growing temperature. These changes in states permit the discharge of carbon dioxide from a solution raising the solution pH, which causes carbonate scale to form, Equation (1. The calcium solubility decreases with a rise in temperature (Nancollas, 1985).

$$Ca^{2+} + 2HCO_3^- \longrightarrow CaCO_3 + CO_2 + H_2O \tag{1}$$

The overall equation given above originates from the set of equilibrium equations defined below:

$$CO_2(g) \rightleftarrows CO_2(aq) \tag{2}$$

$$H_2O + CO_2(aq) \rightleftarrows HCO_3^- + H^+ \tag{3}$$

$$HCO_3^-(aq) \rightleftarrows CO_3^{2-} + H^+ \tag{4}$$

$$Ca^{2+}(aq) + CO_3^{2-}(aq) \longrightarrow CaCO_3(s) \tag{5}$$

$$H_2O \rightleftarrows H^+ + OH^- \quad (water) \tag{6}$$

Sulfate scales usually appear as a result of mixing waters that are incompatible, where one formation water type contains barium ions, while another one contains sulfate ions Equation (7 (Fan et al., 2011). This sea water brine is usually injected to maintain well pressure to allow continuous fluid flow required for production.

$$Ba^{2+}(aq) + SO_4^{2-}(aq) \longrightarrow BaSO_4(S) \quad (7)$$

Equally, Equation (7 could occur for other divalent cations such as Sr^{2+} or Ca^{2+}.

One may find three measures in the precipitation procedure: 1 the accomplishment of supersaturation; 2 nucleation; and 3 the growth of the nuclei to create particles (Gill, 1996). The supersaturation level is a function of changes in temperature (Laing et al., 2003; Sorbie and Laing, 2004), pressure, and pH, all of which may be varied during production. Supersaturation and saturation ratio are interchangeable terms. To get a supersaturated brine system, there are several mechanisms associated with scale formation. These are homogeneous nucleation, heterogeneous nucleation, and subsequent crystal growth (Gill, 1996). These mechanisms occur in equally uninhibited (scale inhibitor (SI) free) and SI-containing systems.

The prediction of calcium carbonate scale in formation water, injection water, and in the mixtures of injection water and formation water is investigated by Amiri et al. (2013b) at different pressures in Iranian oilfields. In this study, the calcium carbonate precipitation was predicted in Siri-C, Siri-D, Siri-E, and Nosrat oilfields, and OLI SCALECHEM software was used in this study. The results of the $CaCO_3$ scale tendency at different pressures of the Persian Gulf water, Siri field formation (Siri C, D, E, and Nosrat) water, and mixtures of injection water and formation water show that the pressure increment drops the amount of $CaCO_3$ precipitation. The effect of temperature on calcium carbonate scale formation is also investigated by Amiri et al. in Iranian oil reservoirs using OLI SCALECHEM software (Amiri and Moghadasi, 2012a). This study shows that the temperature has a great influence on the scaling tendency of $CaCO_3$; when the temperature is increased, the scaling tendency of $CaCO_3$ also rises. The prediction of calcium carbonate and calcium sulfate scale formation by water injection was also investigated by Amiri and Moghadasi (2012b) in some oilfields at different mixing ratios of injection water to formation water using OLI SCALECHEM software for Siri oilfield. The results of the $CaCO_3$ and $CaSO_4$ scale precipitation for the different mixtures of injection water and formation water indicated that when the percent of injected water is increased, the amount of $CaCO_3$ precipitation falls. However, this trend in the $CaSO_4$ first increases, but it then drops. Various authors (Amiri and Moghadasi, 2013; Amiri et al., 2013a; Amiri et al., 2014; Amiri et al., 2013c; Amiri et al., 2012; Moghadasi et al., 2003; Mollakhorshidi et al., 2012) have also investigated the effect of temperature, pressure, and different mixing ratios on calcium sulfate, strontium sulfate, and barium sulfate in Siri Iranian oilfield. In the case of $SrSO_4$ scale, when the Persian Gulf water is injected into Siri oilfield, the $SrSO_4$ scale tendency is increased due to the increment of temperature. In the case of $BaSO_4$, when the Persian Gulf water is injected into Siri oilfield, the $BaSO_4$ scale tendency is declined due to the increment of temperature and pressure.

The DownHole SAT Rx Edition Software calculates all scale potentials and generates all 2D and 3D profile graphs. DownHole SAT also provides “portable” indices in the form of saturation levels. These indices include Stiff-Davis, Oddo-Tomson, Langelier saturation index, Ryznar stability index etc. The saturation levels calculated by the program account for “common ion effects” through a procedure called speciation or ion pairing. This method allows indices to be based upon free ions present rather than total analytical values to provide meaningful common denominators for comparing the scale potential of vastly different water chemistries. The use of ion pairing also allows calculations at a very high ionic strength, where the association of ions becomes as important as (or more important than) the method used to estimate activity coefficients. Moreover, What-if scenario modeling provides one of the greatest benefits from using DownHole SAT. For waterflooding systems, the What-if scenarios allow to evaluate these indicators over a broad ratio range. OLI SCALECHEM is a part of software package named OLI Studio. OLI Studio is a general upstream production chemistry analysis; the software features single point equilibrium calculations, multiple point survey calculations for trend analysis for temperature, pressure, pH, and composition effects, and simple mix and separate capability. There is a big difference between "thermodynamics" and "kinetics". DownHole SAT Rx Edition uses both kinetics and thermodynamics to predict saturation level, but OLI SCALECHEM uses only thermodynamic aspects. In addition, What-if scenarios in DownHole SAT Rx Edition facilitate the simulation of scaling in waterflooding processes.

2. Theory

Ion association models predict the equilibrium distribution of species for cooling water, oilfield brine, waste water, or other aqueous solutions of commercial interest. Scale potential indices based upon the free ion concentrations estimated by ion association models have been used extensively in the past decade to predict scale problems in industrial systems (Ferguson et al., 1995; Ferguson, 2011; Ferguson et al., 2011a; Ferguson et al., 2011b).

Indices calculated by the models have been found to be transportable to waters of diverse composition and ionic strength. They have overcome many of the problems encountered with simple indices which do not account for ion pairing (Ferguson et al., 1995). This part of the current work discusses the application of ion association model saturation level indices to predict and resolve scale formation problems in oil field brines.

2.1. The concept of saturation

A majority of the indices used routinely by water treatment chemists are derived from the basic concept of saturation. A water is said to be saturated with a compound (e.g. calcium carbonate) if it does not precipitate the compound and does not dissolve any of the solid phase of the compound when left undisturbed in the same conditions for an infinite period of time. A water which does not precipitate or dissolve a compound is at equilibrium for that particular compound. By definition, the amount of a chemical compound which can be dissolved in a water and remain in solution for this infinite period of time is described by the solubility product (*Ksp*). In the case of calcium carbonate, solubility is defined by the relationship given below (Ferguson et al., 1995; Ferguson, 2011; Ferguson et al., 2011a; Ferguson et al., 2011b):

$$(Ca)(CO_3) = Ksp \tag{8}$$

where, (Ca) is the calcium activity, and (CO_3) is the carbonate activity; *Ksp* is the solubility product for calcium carbonate at the temperature under study.

In a more generalized sense, the term $(Ca)(CO_3)$ can be called the ion activity product (IAP), and the equilibrium condition described by the relationship reads as (Ferguson et al., 1995; Ferguson, 2011; Ferguson et al., 2011a; Ferguson et al., 2011b):

$$IAP = Ksp \tag{9}$$

The degree of saturation of a water is described by the relationship of the ion activity product (IAP) to the solubility product (*Ksp*) for the compound as follows:

If a water is under saturated with a compound:

- ✓ IAP < *Ksp* (it will tend to dissolve the compound).

If a water is at equilibrium with a compound:

- ✓ IAP = *Ksp* (it will not tend to dissolve or precipitate the compound).

If a water is supersaturated with a compound:

- ✓ IAP > *Ksp* (it will tend to precipitate the compound).

The index called saturation level (SL), degree of supersaturation, or saturation index describes the relative degree of saturation as a ratio of the ion activity product (IAP) to the solubility product (*Ksp*) (Ferguson et al., 1995; Ferguson, 2011; Ferguson et al., 2011a; Ferguson et al., 2011b):

$$Saturation\ Level\,(SL) = \frac{IAP}{Ksp} \tag{10}$$

In actual practice, the saturation levels are calculated by various computer programs. The programs use the activity coefficients used in the IAP for estimating; they differ in the choice of solubility products and their variation with temperature. Also, they differ in the dissociation constants used to estimate the concentration of reactants (e.g. CO_3 from analytical values for alkalinity and PO_4 from analytical orthophosphate) (Ferguson et al., 1995; Ferguson, 2011; Ferguson et al., 2011a; Ferguson et al., 2011b).

2.2. Ion pairing

The saturation index discussed can be calculated based upon total analytical values for the reactants. Ions in water, however, do not tend to exist totally as free ions. Calcium, for example, may be paired with sulfate, bicarbonate, carbonate, phosphate, and other species. Bound ions are not readily available for scale formation. The computer program calculates saturation levels based upon the free concentrations of ions in a water rather than the total analytical values, which include bound ions too. Early indices such as the Langelier saturation index (LSI) for calcium carbonate scale are based upon total analytical values rather than free species primarily due to the intense calculation requirements for determining the distribution of species in a water. Speciation of a water is time prohibitive without the use of a computer for crunching the numbers. The process is iterative and involves (Ferguson et al., 1995; Ferguson, 2011; Ferguson et al., 2011a; Ferguson et al., 2011b):

1. Checking the water for an electroneutrality via a cation-anion balance, and balancing with an appropriate ion (e.g. sodium or potassium for cation deficient waters, sulfate, chloride, or nitrate for anion deficient waters);
2. Estimating ionic strength, calculating and correcting activity coefficients and dissociation constants for temperature, and correcting alkalinity for non-carbonate alkalinity;
3. Iteratively calculating the distribution of species in the water from dissociation constants;
4. Checking the water for balance and adjusting ion concentrations to agree with analytical values;
5. Repeating the process until corrections are insignificant;
6. Calculating saturation levels based upon the free concentrations of ions estimated using the ion association model (ion pairing);

The use of ion pairing to estimate the free concentrations of reactants overcomes several of the major shortcomings of traditional indices. Indices such as the LSI correct activity coefficients for ionic strength based upon the total dissolved solids. They do not account for “common ion” (Ferguson et al., 1995; Ferguson, 2011; Ferguson et al., 2011a; Ferguson et al., 2011b) effects. Common ion effects increase the apparent solubility of a compound by reducing the concentration of reactants available. A common example is sulfate reducing the available calcium in a water and increasing the apparent solubility of calcium carbonate. The use of indices which do not account for ion pairing can be misleading when comparing waters where the total dissolved solid (TDS) is composed of ions which pair with the reactants versus ions which have less interaction with them.

3. Methods

The procedure of this study is shown in **Error! Reference source not found.**. Sea water (SW) sample belongs to the Persian Gulf in Iran, and formation water (FW) sample is provided by West Oil and Gas Production Company (WOGPC) in one of oil wells in Naft-Shahr. A variety of methods are used to determine the concentration of ionic content of samples, but in this study ion chromatography (IC) and inductively coupled plasma (ICP) analyses are chosen. The minimum detection limit of these analyses is about 1 ppb, which is quite enough in this work. IC analysis was included to investigate the concentration of the following ions: Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, SO_4^{2-}, and HCO_3^-. Also, the concentration of the other ions (Fe^{2+}, Ba^{2+}, and Sr^{2+}) was analyzed by ICP. The prepared sample of SW for this purpose was 100 ml, and 100 ml of FW was prepared to analyze ions.

Before starting the experimental procedures, injection water and brine samples were analyzed by IC and ICP tests to determine their ions. IC test determined Na^+, K^+, Ca^{2+}, Mg^{2+}, Cl^-, SO_4^{2-}, and HCO_3^- in the samples. ICP test was used to determine the Fe^{2+}, Ba^{2+}, and Sr^{2+} ions in the samples. These ion analyses were used as the input to the compatibility simulations.

It is necessary to simulate the compatibility of brine and injection water. This simulation helps with the identification of different types of scales that will form in the solution of injection water and brine. Furthermore, the simulation determines the mixing ratio (mixing ratio is the proportion of each material over the overall volume/mass) in which the maximum scale potential will occur. Compatibility simulation was performed using DownHole SAT Rx Edition Software (Software, 2015). DownHole SAT simulated the compatibility at different ratios, pressures, and temperatures; the output of this software is saturation levels (SL). The mentioned effects can be represented in 3-D graphs classified using different colors. These colors represent different conditions which are described as follows:

RED signifies a definite problem prediction. Corrective actions such as treatment, pH adjustment, or concentration ratio decrease (blowdown) should be taken to eliminate, or alleviate, the problem. Red signifies loss of control in *treated saturation level profiles*.

MAGENTA (purple) denotes that a problem is highly possible. Some systems might be able to carry the degree of supersaturation indicated by magenta without deposition occurring. Most systems will precipitate if operated in the magenta range without treatment.

YELLOW signifies that a problem will occur with a minimum of change. Make sure of your measurements. A slight error in pH could mean that you are really operating in a RED zone. The system is borderline and should be watched.

GREEN means no problems are likely. Significant changes (e.g. a 0.1 pH increase, a 10 °C temperature increase) could result in a problem.

BLUE signifies a safe operating range where deposition from the scale evaluated would not be expected. Blue is also used to signify a successful treatment in *treated system profiles*.

The program computes the saturation levels regarding common ion effects through a procedure called speciation or ion pairing. DownHole SAT (Software, 2015) allows to evaluate the scale potential for common scales over the range of water chemistry, temperature, pressure, pH, and pCO_2 anticipated in a surface water, brine, or mixture. DownHole SAT can evaluate the scale potential for common scales at both the lowest and highest temperatures anticipated and at the lowest and highest pH values or pCO_2 expected, and can assist in predicting scale problems downhole, or when a water is brought to the surface. DownHole SAT also evaluates mixtures of waters at the ratios, pressures, partial pressures, and temperatures we select (SAT, 2013).

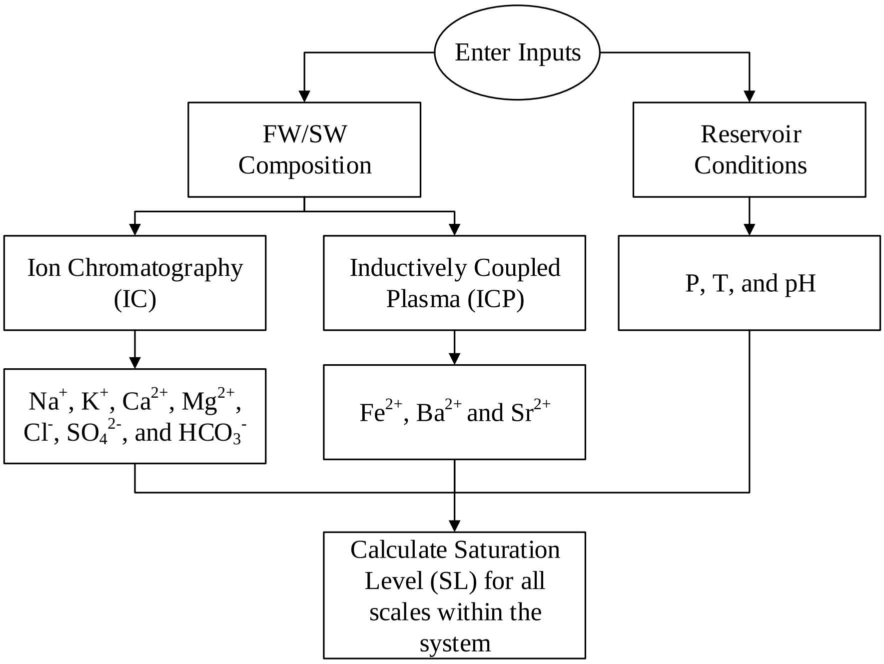

Figure 1
Flowchart of the study for assessing scaling tendency due to mixing ratio changes.

4. Results and discussion

Table 2 provides the results derived from the preliminary analysis of FW and SW samples by IC and ICP. **Error! Reference source not found.** demonstrates the saturation level of probable scales as a function of mixing ratio of sea water. The index called saturation level (SL) describes the relative degree of saturation as a ratio of the ion activity product (IAP) to the solubility product (Ksp) (Ferguson et al., 1995; Ferguson, 2011; Ferguson et al., 2011a; Ferguson et al., 2011b). Saturation level increases as the mixing ratio of sea water decreases; therefore, the maximum precipitation occurs when mixing ratio of sea water is 10%. As can be seen in Figure 2, saturation level in 0% of mixing ratio has the highest value. However, this study aims to investigate scale potential in a water flooding process. Mixing the injection water and formation water is the main way for scale formation; thus, the sea water (injection water) must be mixed into formation water. Also, a 10% mixing ratio is chosen for the highest saturation level. Hence, for all the experiments, this mixing ratio of sea water is chosen. As it can be seen in this graph, calcite and aragonite have the most precipitation potential in this study.

Table 2

Water analysis of formation water and sea water.

	Formation water	**Sea water**
Na^+	35387	15000
K^+	287	372
Ca^{2+}	1505	372
Mg^{2+}	1216	-
Ba^{2+}	0.02	-
Sr^{2+}	7.768	2.3
Fe^{2+}	0.016	0.005
SO_4^{2-}	8492	5450
HCO_3^-	1267	67
Cl^-	52112	22167
pH	7.76	7.10
Density	1.07080	1.02330
TDS	55600	31400

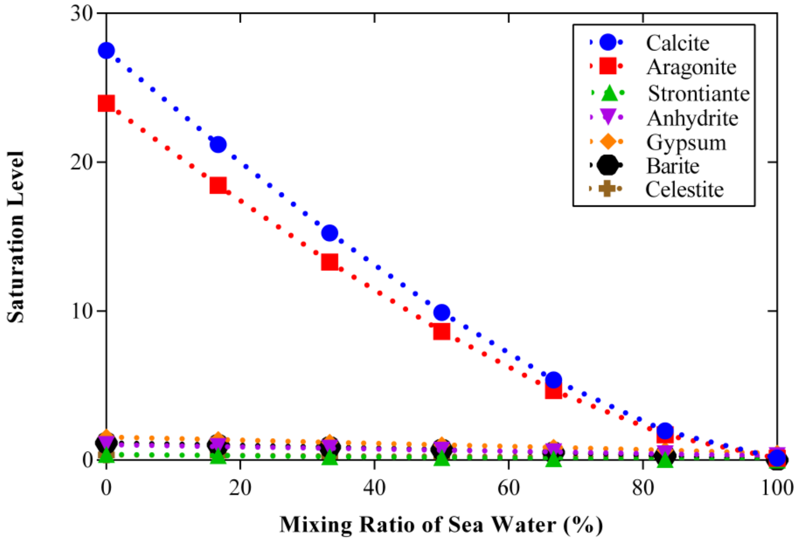

Figure 2
Saturation level as a function of mixing ratio for different scales in solution.

Figure 3 shows the effect of pH and temperature (°F) on calcite saturation level at a 10% sea water mixing ratio. The vertical axis (*z* axis) is the saturation level, and the *x* axis is pH; the *y* axis is temperature. The index called saturation level (SL) describes the relative degree of saturation as a ratio of the ion activity product (IAP) to the solubility product (*Ksp*) (Ferguson et al., 1995; Ferguson, 2011; Ferguson et al., 2011a; Ferguson et al., 2011b). At any constant temperature, the saturation level of calcite increases sharply as the pH rises. With a pH less than 8.33, saturation level increases gradually as the temperature rises; at pH values greater than 8.33, this increase of saturation level is drastic. Scale formation in a hard water as $CaCO_3$ precipitation process is an equilibrium and slowing process. An increase in pH due to CO_2 release in solution will push the precipitation of $CaCO_3$ as presented in Equation 1. The change in precipitation with pH is due to a change in buffer intensity which varies inversely with pH, in agreement with calcium carbonate precipitation potential (CCPP) theory; also, aragonite acts like calcite.

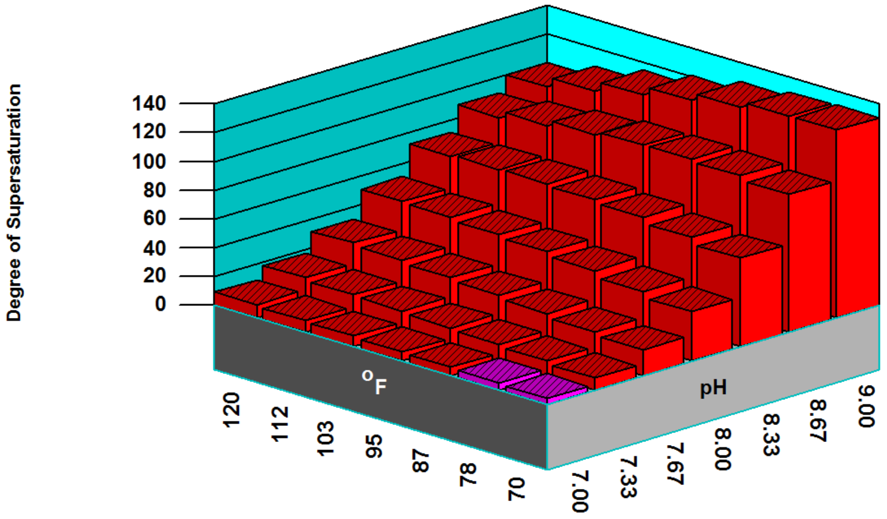

Figure 3
Effect of pH and temperature on calcite saturation level.

Figure 4 shows the effect of pH and temperature (°F) on aragonite saturation level at a 10% sea water mixing ratio. The vertical axis (*z* axis) is the saturation level, and the *x* axis is pH; the *y* axis is temperature. This graph shows results similar to Figure 3.

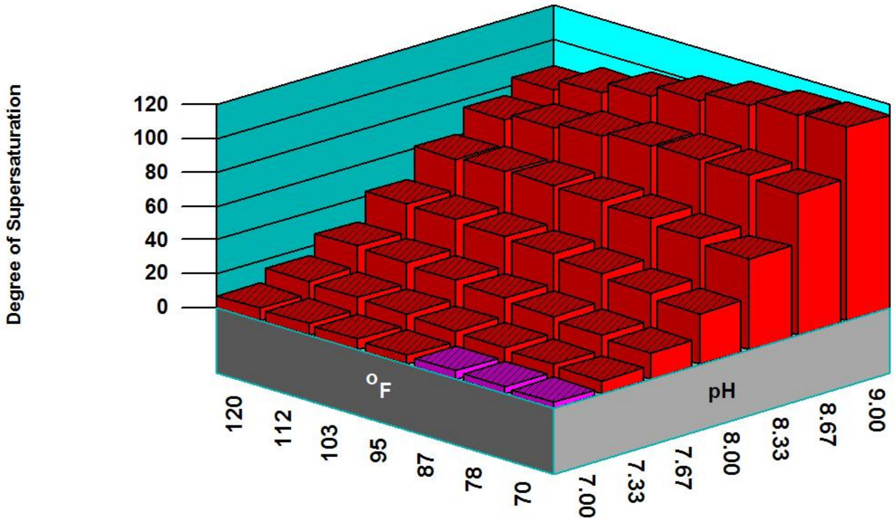

Figure 4
Effect of pH and temperature on aragonite saturation level.

Based on the simulating study for pH of 7 to 8.33, the saturation level of calcite and aragonite increased as the temperature rose. At pH values of 8.33 to 9, this amount decreased, which is true according to Amiri et. al. (Amiri and Moghadasi, 2012a) study on $CaCO_3$ tendency for Siri oilfield in Iran.

Figure 5 shows the effect of pH and temperature (°F) on strontianite saturation level at a 10% sea water mixing ratio. The vertical axis (*z* axis) is the saturation level, and the *x* axis is pH; the *y* axis is temperature. At any temperature, the saturation level of strontianite increases with increasing pH. At pH values up to 8.33, saturation level slightly increases while temperature drops. At pH values greater than 8.33, this trend gradually increases.

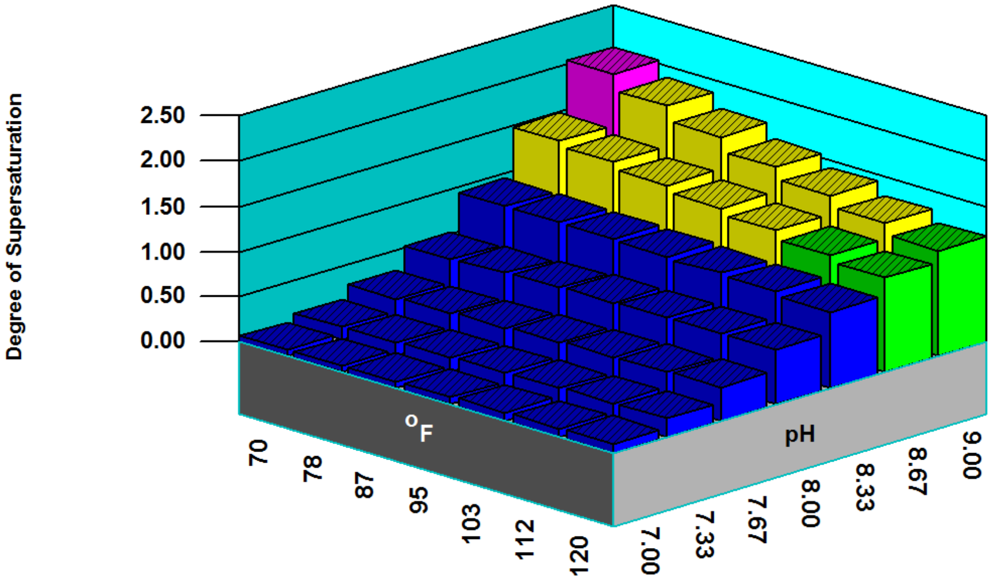

Figure 5
Effect of pH and temperature on strontianite level.

Figure 6 shows the effect of pH and temperature (°F) on anhydrite saturation level at a 10% sea water mixing ratio. The vertical axis (*z* axis) is the saturation level, and the *x* axis is pH; the *y* axis is temperature. At any pH values, the saturation level of anhydrite increases with raising temperature. It can be seen that temperature affects the saturation level of anhydrite much more than pH. Lopez-Salinas et al. (2011) investigated the anhydrite saturation index for several wells in different formations at room temperature, 85 °C, and reservoir temperature (120 °C). They observed the increasing trends with respect to increasing temperatures and expressed that the saturation index, for most of the minerals, is sensitive to the pH, anhydrite being an exception. The finding of the mentioned work agrees with the result of this study.

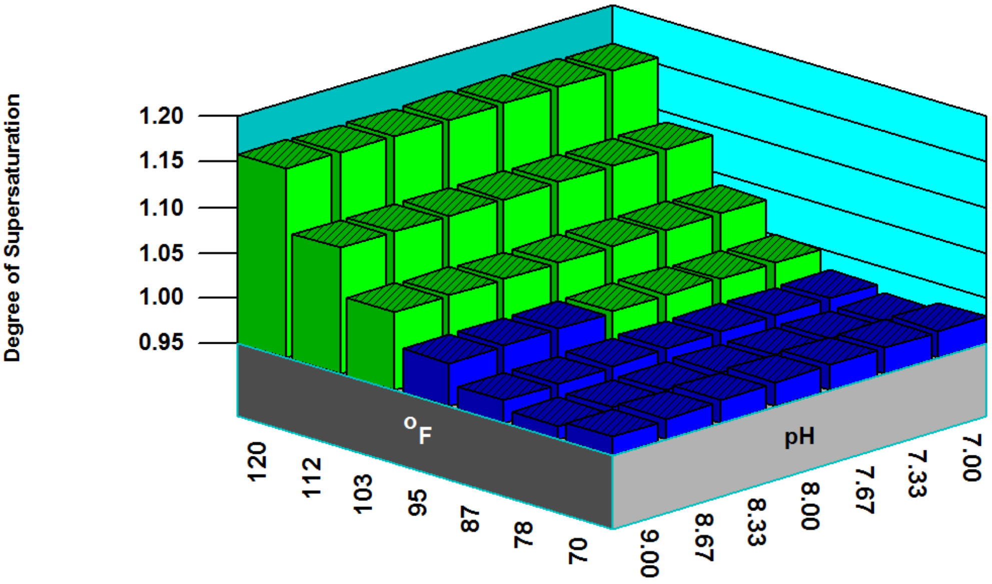

Figure 6
Effect of pH and temperature on anhydrite saturation level.

Figure 7 shows the effect of pH and temperature (°F) on gypsum saturation level at a 10% sea water mixing ratio. The vertical axis (*z* axis) is the saturation level, and the *x* axis is pH; the *y* axis is temperature. At any pH, saturation level decreases with decreasing temperature up to 103 °F; it then increases with lowering temperature. It is obvious that temperature mainly controls gypsum saturation level.

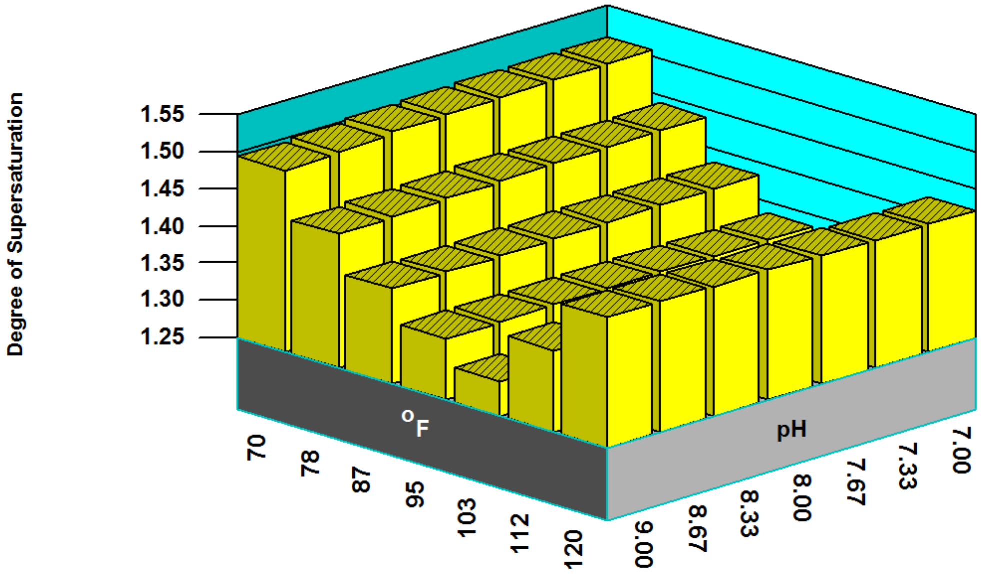

Figure 7
Effect of pH and temperature on gypsum saturation level.

Figure 8 shows the effect of pH and temperature (°F) on barite saturation level at a 10% sea water mixing ratio. The vertical axis (*z* axis) is the saturation level, and the *x* axis is pH; the *y* axis is temperature. The variation of saturation level for barite is the same as Figure 7. Our results are in agreement with the work of Amiri et. al. (Amiri and Moghadasi, 2012a) showing that increasing temperature leads to a fall in $BaSO_4$ saturation level. According to Skillman et al. (1969), *Ksp* of barite is increased with raising temperature. Considering the inverse relation between *Ksp* and saturation level (Equation 10), the saturation level of barite decreases at higher temperatures. Moreover, pH has no striking impact on barite saturation level.

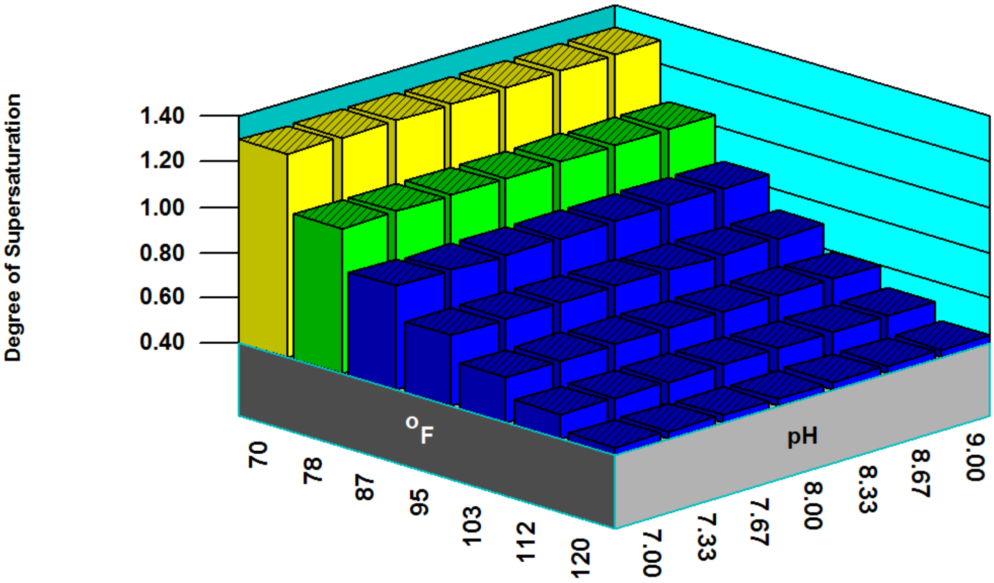

Figure 8
Effect of pH and temperature on barite saturation level.

5. Conclusions

In this study, the evaluation of scale potential at different mixing ratios of SW and FW were firstly conducted. This study used the ion analysis of Naft Shahr oilfield formation water to simulate the scale formation potential in a probable water flooding project. The results of this study can be used in Naft Shahr water flooding implementation in future. According to the obtained results, the following conclusions and/or recommendations are presented:

- Simulation in this study has shown that calcite and aragonite were the major scales in this investigation.
- The results of the simulation indicate that at specific mixing ratios, pH was generally the main parameter that impacts on the calcite and aragonite scales.
- The results of this study indicate that at specific mixing ratio, temperature was generally the main parameter that influences the gypsum, anhydrite, and barite scales.

Nomenclature

FW	: Formation water
IAP	: Ion activity product
IC	: Ion chromatography
ICP	: Inductively coupled plasma
K_{SP}	: Solubility product
SL	: Saturation level
SW	: Sea water
TDS	: Total dissolved solid
WOGPC	: West Oil and Gas Production Company

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