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Application of Ion Association Modeling in Prediction of Oilfield Scaling Tendency during Oil Recovery Process by Water Flooding

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Abstract

Considering the maturity and reaching to half-life production in many petroleum reservoirs, development of methods for maintenance and enhanced oil recovery are becoming crucial. One of the most common methods for the mentioned purposes is water flooding. However, there is a drawback to this method that is scaling in reservoirs due to the mixing of injection water with formation water, which would cause formation damage and reduction in production. Before the water flooding begins, it is necessary to simulate the compatibility of brine and injection water. For this purpose, in this study simulations with ion association modeling (ion pairing methods) have been undertaken. To verify the simulations results, X-Ray powder Diffraction (XRD) and Scanning Electron Microscope (SEM) tests have been performed. Acquired data have been compared with XRD and SEM, which show that the simulations are correct. Also, SEM test determined the crystal's structure of scale. Results show that carbonate scale is the major precipitation in this study.

Keywords: *Oilfield scale modeling; Calcium carbonate scale; Ion association; Water injection; Water flooding.*

1. Introduction

Organic and inorganic contents are constituted the scale composition [1]. Scaling phenomena are triggered when supersaturation of a solution is begun, as a result of pH variation, changes in temperature, or if two different water (formation water and injection water) mix together. Scaling can block the porous media or production lines immediately in a few hours [2-3]. Furthermore, scale precipitation damages jeopardize the whole safety and economic asset of production operation [4]. Calcium carbonate, calcium sulfate, barium sulfate, strontium sulphate and carbonates are the most common oilfield scales [5-10]. Calcite is the most common type of oilfield scale [5]. Precipitation of calcium carbonate promptes $CaCO_3$ scale formation as follows:

$$Ca^{2+} + CO_3^{2-} \rightarrow CaCO_3 \downarrow \qquad (1)$$

Calcium sulfate occurs as scales in three different kinds including: gypsum ($CaSO_4 \cdot 2H_2O$), hemihydrate ($CaSO_4 \cdot ½H_2O$), and anhydrite ($CaSO_4$). As same as the barite, the main cause of calcium sulfate formation is the mixing of incompatible of waters. Also, an increase in temperature, steam flooding, and membrane filtration can motivate the precipitation of calcium sulfate [11-13].

Barite scale formation ($BaSO_4$) is a mineral consisting of barium sulfate. Barite precipitation follows through the mixing of high sulfate formation water and injection water. Also, the pressure drop can affect the formation of barite. Kharaka *et al.* [14] investigated the concentration of barium and sulfate for a large group of wells. It is proposed that formation water is saturated with barite in the reservoir which is not flooded. Also, decreasing in pressure and temperature caused the increases of barite saturation index. On the other hand, pressure and temperature diminishing in near wellbore are not plentiful to barite precipitation in wellbore or tubing. Considering this matter, the places should be cautious about it, where the conditions alter very quickly, specially near the wellbore and injection well [15].

Celestite ($SrSO_4$) is a mineral consisting of strontium sulfate. Although, the solubility of celestite is less studied compared to the other known scales [16], a wide range of its solubility has been reported in literature [17-20].

The mechanism of the aforementioned scales is thoroughly comprehended. But, it is crucial to develop precipitation mechanisms of unremarkable scales such as sulfide scales. Sulfide scales have been fully discussed in literature [21-23].

Prediction and modeling of scaling tendency is the first step of oilfield scale management. Therefore, the extensive review of oilfield scale modeling appears to be essential. Vetter *et al.* [24] suggested an aggressive model to predict the sulfate scales precipitation. This model proposes the regulation of sulfate scales. In this regulation, sulfate scales are precipitated in order to insolubility of scales, first barium sulfate, then strontium sulfate and finally calcium sulfate.

Yuan and Todd [25] used the Pitzer [26] activity coefficient in iterative algorithm to promote the Vetter et al model. This model was used to predict the activity coefficient of various ions in high saline waters for typical reservoir pressure and temperature. In a forwarding approach, investigation of carbonate and sulfate scales tendency was conducted through an equilibrium model by Haarberg *et al.* [27]. Oddo and Thomson [28] presented polynomial scaling index (SI) as a function of ionic strength, pressure and temperature.

The primary scale models were established just based on thermodynamic aspects [29-30]. Since it is believed that the calcite scale formation depends not only on thermodynamics, but also on the surface kinetics, it reveals that available scale models do not work properly [31]. Therefore, scale potential models based on both thermodynamics and kinetics aspects have been developed for apply in oilfield purposes.

Moghadasi *et al.* [32] introduced a model based upon kinetics of oilfield data due to scale formation from water injection. Amiri and Moghadasi, and Amiri et al. [33-35] investigated the effect of temperature, pressure and mixing ratio on different scales. Amiri *et al.* [34] investigated the effect of pressure on calcium carbonate precipitation. As a result of this study, an increase in pressure caused a decrease in calcium carbonate. Also, the aforementioned results were compared to the result of OLI ScaleChem software. Amiri *et al.* [33,36] investigated the calcium sulfate potential to precipitate and the factors that impact on it such as temperature and pressure. These studies investigated the calcium sulfate scaling in both reservoir and production equipment. Amiri and Moghadasi [36] checked into thoroughly the barium sulfate precipitation as insoluble scale formation. Finally, Kamari *et al.* [37] proposed a new model to predict the barium sulfate tendency, based on least square support vector machine (LSSVM), which is a regression method.

2. Theory and modeling

Equilibrium distribution of species for a water associated system such as oilfield brine, waste water, or aqueous solution can be predicted by ion association models. In the past decades, ion association models have been utilized to predict scale indices established by free ion association for resolving of scaling consequences in various industries [38-41]. Ion association models use the ion pairing method to solve the problem of over/under estimation of early indices. Based on Menéndez and Borge [42], there are different types of thermodynamic models including: basic, association, interaction and mixed. Chemical models of ion association are established based on free ions and ion pairing. Mentioned property of ion association models makes the superiority of this methods to the others.

This part of the study discusses the application of ion association model and ion pairing method for determining the saturation level index to predicting scale tendency in oilfield brines.

3. Concept of saturation

The fundamental concept of saturation is the origin of many scale tendency indices. The state of being saturated occurs when a solution will not precipitate or dissolve any solid phase which the state can be stable for a long time. In other words, the solution is at equilibrium for discussed compound and saturation is a dynamic equilibrium between dissolution and precipitation. It should be noted that Solubility product (Ksp) is defined for low salt soluble for which there is a balance between the existing ions in solution and the solid state of matter. This balance for the completely dissolved salt cannot be defined, because of the relationship between them is one sided directed to dissolution. For instance, the solubility of calcium carbonate is defined by the following relationship [39-42]:

$$\mathrm{Ksp} = (\mathrm{Ca})(\mathrm{CO_3}) \qquad (2)$$

where Ksp is the solubility product for calcium carbonate; (Ca) is the calcium activity; (CO_3) is the carbonate activity.

In a certain condition, Ion Activity Product (IAP) is equal to the term $(Ca)(CO_3)$. In other words, the equilibrium condition can be satisfied by the relationship [39-42:

$$\mathrm{IAP} = \mathrm{Ksp} \qquad (3)$$

The state of a solution can be revealed base on the saturation relation as follows:
When a solution is under saturated with a compound:
✓ IAP < Ksp
When a solution is at equilibrium with a compound:
✓ IAP = Ksp
When a solution is supersaturated with a compound:
✓ IAP > Ksp

The index called Saturation Level (SL), Degree of Supersaturation or Saturation Index which is used to determine the scaling tendency is quite similar to the concept of saturation for a solution [39-42]:

$$\text{Saturation Level(SL)} = \frac{\mathrm{IAP}}{\mathrm{Ksp}} \qquad (4)$$

3.1. Ion pairing

Ion pairing formation is ignored in calculation of typical scaling indices. Considering the effect of ion pairing causes to improve the accuracy of the indices, Eq. (5), (6) and (7) show the example of ion pairing for calcium in water:

$$Ca^{2+} + HCO_3^- \Leftrightarrow CaHCO_3^- \qquad (5)$$

$$Ca^{2+} + {CO_3}^{2-} \Leftrightarrow CaCO_3 \qquad (6)$$

$$Ca^{2+} + {SO_4}^{2-} \Leftrightarrow CaSO_4 \qquad (7)$$

Fig. 1 shows the process of saturation level calculation based on ion pairing in a solution. Ion pairing calculation is an iterative process [39-42].

4. Methods and procedures

The procedure of this study is shown in Fig. 2. Before starting the procedure, 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 samples. ICP test was used to determine the Fe^{2+}, Ba_{2+} and Sr^{2+} ions in samples. This ion analysis was used as input for compatibility simulations.

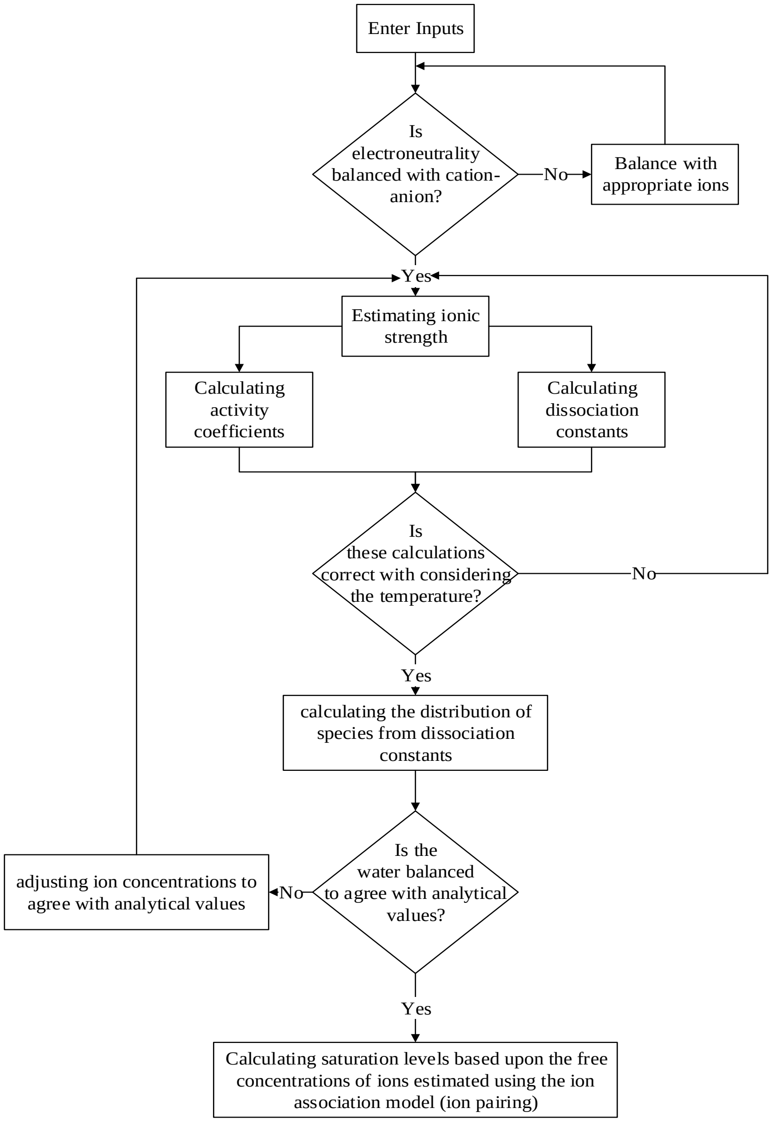

Fig. 1. Saturation level calculation based on ion association modeling

Various types of scale formation was recognized by the aid of simulation result. Also, simulation determined the mixing ratio with the maximum scale potential. Compatibility simulation was performed using DownHole SAT Rx Edition Software. DownHole SAT simulates the compatibility at different ratios, pressures and temperatures. Mentioned effects can be represented in 3-D graphs. These graphs are classified using different colors. These colors represent different conditions which are described as follows:

- Red signifies a definited problem prediction
- Magenta (purple) signifies a problem that is highly likely
- Yellow signifies a problem that will occur with a minimum of change
- Green signifies: No problems are likely.

The first time DownHole SAT is run, it will display a warning that default analytical input units are in affect. This is a reminder to run the Input Units module (which can be selected from the Preferences menu) and choose the analytical reporting units used by user requirement. The first thing in the process of simulation with software is to put the analysis of SW and FW as inputs in the software. Available units of SW and FW are ppm and mg/l, and in this procedure mg/l was used. The compatibility of SW/FW was determined using mix option from what if tab in the software. This tab also helps to determine the effect of pH, temperature, mixing ratio on SL of different scales. The output of this software is saturation levels (SL). The program computes the saturation levels regarding common ion effects through a procedure called speciation or ion pairing.

To verify the simulations result, XRD and SEM-EDX tests were performed. A small (about 0.5 gr) sample of scales was chosen, and XRD analysis was conducted. The XRD test in the study was performed using an apparatus owned by Geological Survey of Iran (GSI). Also, the SEM tests were performed using an apparatus owned by Razi Metallurgical Research Center (RMRC).

XRD analysis indicated the type of formed scale/precipitations and SEM-EDX revealed morphology and elemental analysis of the scale crystals. To produce precipitation (to use for XRD test) 1500 mL of solution with 10% SW-90% FW was maintained at room temperature (Equal to 25°C) for 24 hours. Since the scale was stuck at the bottom of the container, the container was removed of its contents and was placed in an oven in order to be dried. Then the scales were separated. Due to the evaporation taking place in the oven, there were a probability of the evaporate scale formation. To prevent this error, elemental analysis through SEM was utilized in order to verify XRD result. For SEM test, 100 mL of desired solution was filtered with 0.22μm filter paper after 24 hours. Therefore, elemental analysis was a better criterion to verify XRD analysis.

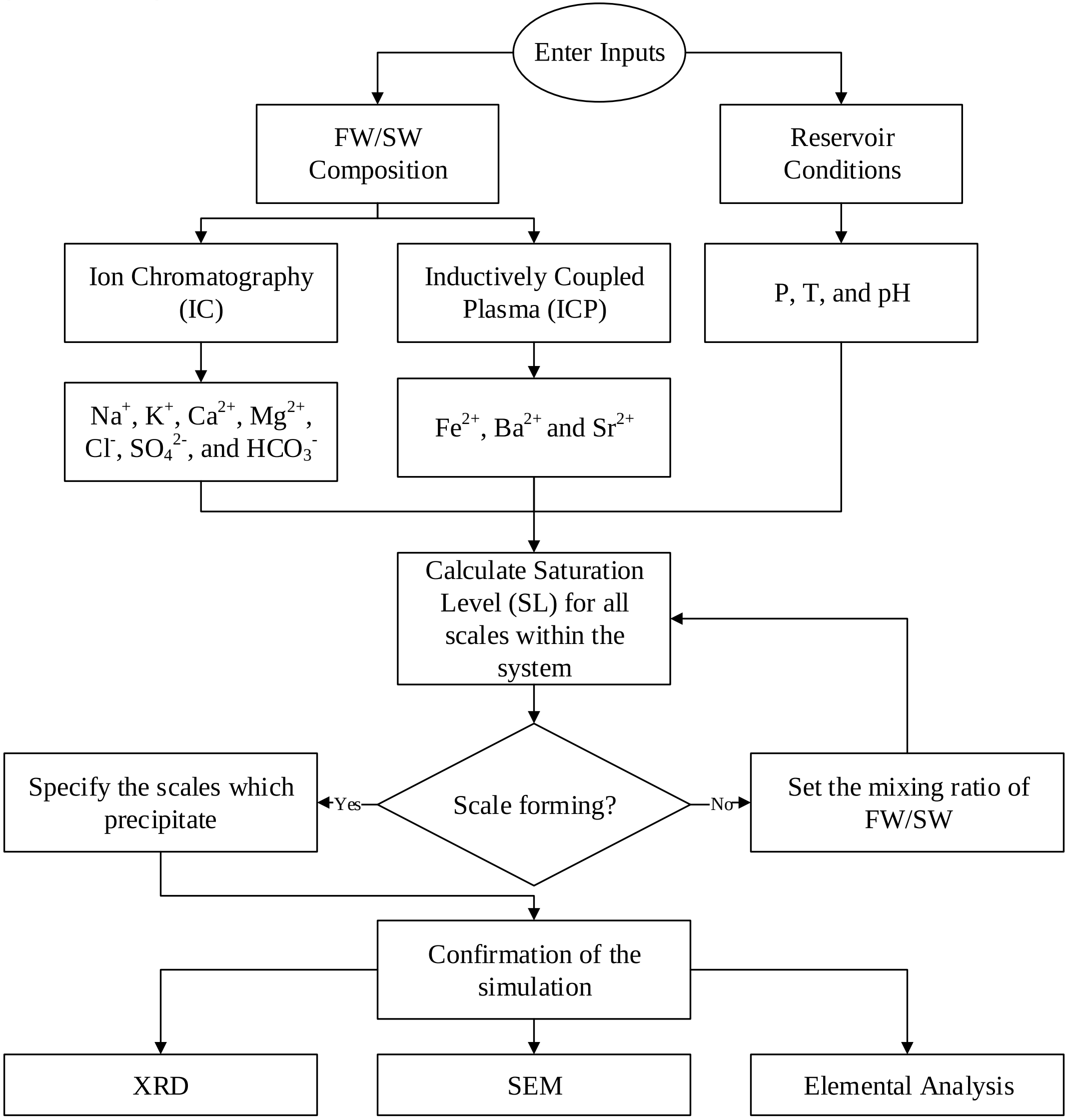

Fig. 2. Flowchart of the study for assessing scaling tendency due to mixing ratio changes

5. Results and discussion

5.1. Formation water and sea water analysis

Table 1 provides the results derived from the preliminary analysis of FW and SW samples by IC and ICP.

Table 1. Water analysis of formation water and sea water

	Formation water	Sea water		Formation water	Sea water
Na^+	35 387	15 000	SO_4^{2-}	8 492	5 450
K^+	287	372	HCO_3^-	1267	67
Ca^{2+}	1505	372	Cl^-	52 112	22 167
Mg^{2+}	1216	-	pH	7.76	7.10
Ba^{2+}	0.02	-	Density	1.07080	1.02330
Sr^{2+}	7.768	2.3	TDS	55 600	31 400
Fe^{2+}	0.016	0.005			

5.2. Simulation results

Fig. 3 demonstrates the saturation level of probable scales as a function of mixing ratio of sea water. 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) [39-42]. Saturation level increases as the mixing ratio of sea water decreases, therefore the maximum precipitation occurs when mixing ratio of sea water is 10 %. Thus, for all the experiments this mixing ratio of sea water was chosen. As It can be seen in this graph, calcite and aragonite have the most precipitation potential in this study.

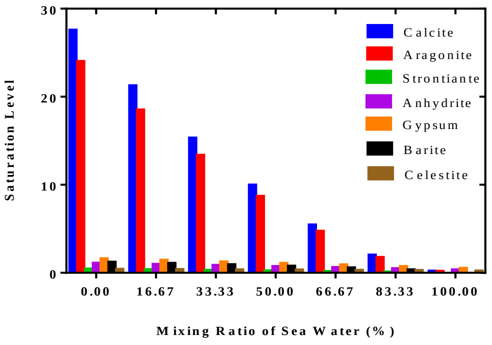

Fig. 1. Saturation level as a function of mixing ratio for different scales in solution

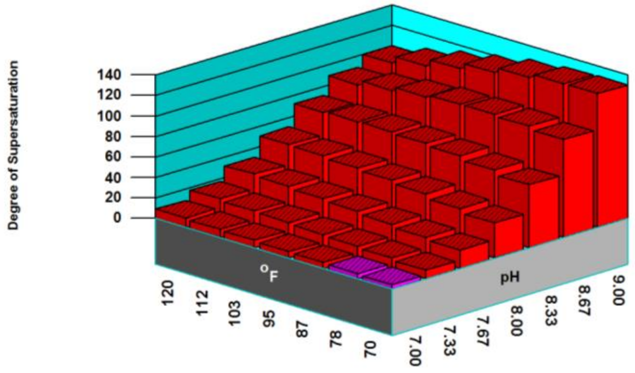

Fig. 2. Effect of pH and temperature on calcite saturation level

Fig. 4 shows the effect of pH and temperature (°F) on calcite saturation level at 10 % of sea water mixing ratio. The vertical axis (z axis) is the saturation level, the x axis is pH and the y axis is temperature. At any constant temperature, saturation level of calcite increases sharply as the pH increases. With pH less than 8.33, saturation level increases gradually as the temperature increases, after that (pH greater than 8.33) this increase of saturation level is drastic.

Fig. 5 shows the effect of pH and temperature (°F) on aragonite saturation level at 10 % of sea water mixing ratio. The vertical axis (z axis) is the saturation level, the x axis is pH and the y axis is temperature (Fig. 5 shows similar results as Fig. 4).

Based on simulating the study for pH of 7 to 8.33, saturation level of calcite and aragonite increased as the temperature went higher. In 8.33 to 9 pH, these amounts decreased which according to Amiri and Moghadasi [43] on $CaCO_3$ tendency for Siri oilfield in Iran were true.

Fig. 6 shows the effect of temperature (°F) and mixing ratio of SW/FW on calcite saturation level. The vertical axis (z axis) is the saturation level, the x axis is temperature and the y axis is mixing ratio of SW/FW. Calcite saturation level increases as the temperature increases and mixing ratio decreases. Thus, it can be inferred that mixing ratio is the key factor that controls the calcite saturation level.

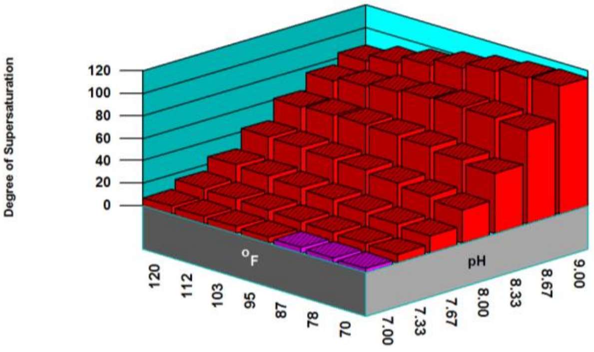

Fig. 3. Effect of pH and temperature on aragonite saturation level

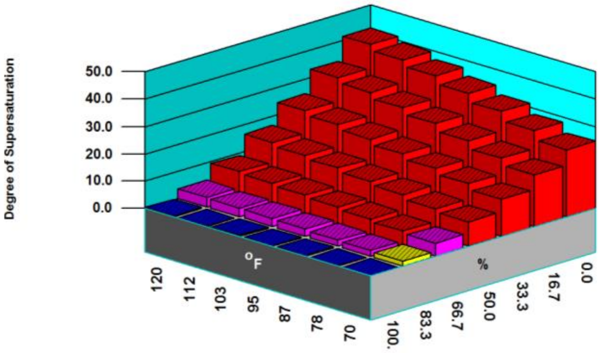

Fig. 4. Effect of temperature and SW/FW mixing ratio on calcite saturation level

Fig. 7 shows the effect of temperature (°F) and mixing ratio of SW/FW on aragonite saturation level. The vertical axis (z axis) is the saturation level, the x axis is mixing ratio of SW/FW and the y axis is temperature. Aragonite saturation level acts in the same way as calcite saturation level.

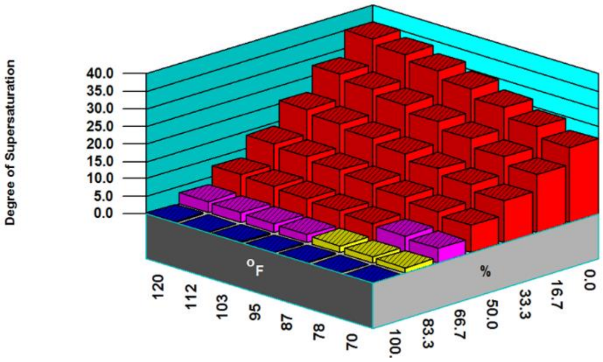

Fig. 5. Effect of temperature and SW/FW mixing ratio on aragonite saturation level

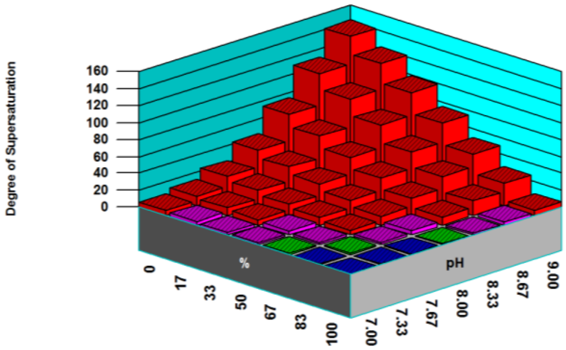

Fig. 6. Effect of SW/FW mixing ratio and pH on calcite saturation level

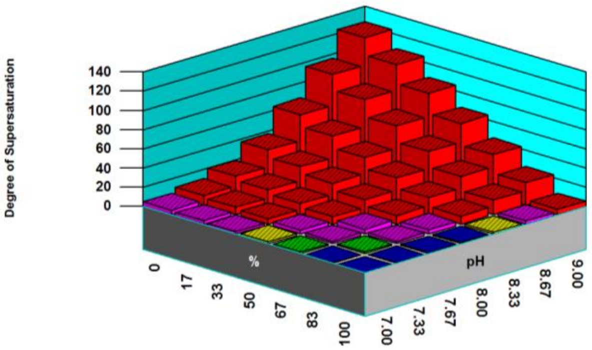

Fig. 7. Effect of SW/FW mixing ratio and pH on aragonite saturation level

Fig. 8 and Fig. 9 show the effect of SW/FW mixing ratio and pH on calcite and aragonite saturation levels. The vertical axis (z axis) is the saturation level, the x axis is pH and the y axis is mixing ratio of SW/FW. At any constant pH, saturation level increases as the mixing ratio decreases. As pH increases, this increase of saturation level is more sharp.

Fig. 10 shows XRD analysis for precipitated scales. The x axis is 2θ (θ is the angle of diffraction) and the y axis is the intensity. It shows two main peaks that Geological Survey of Iran (GSI) (the owner company of XRD) interprets them as calcite and halite based on BS EN

13925-1:2003 standard. XRD analysis determines that halite crystals are in fact NaCl in cubic form and calcite crystals are $CaCO3$ in hexagonal form. Simulations on this matter predicted only the formation of carbonate scales in this experimental solution, then to verify XRD results the EDX test within SEM apparatus was conducted.

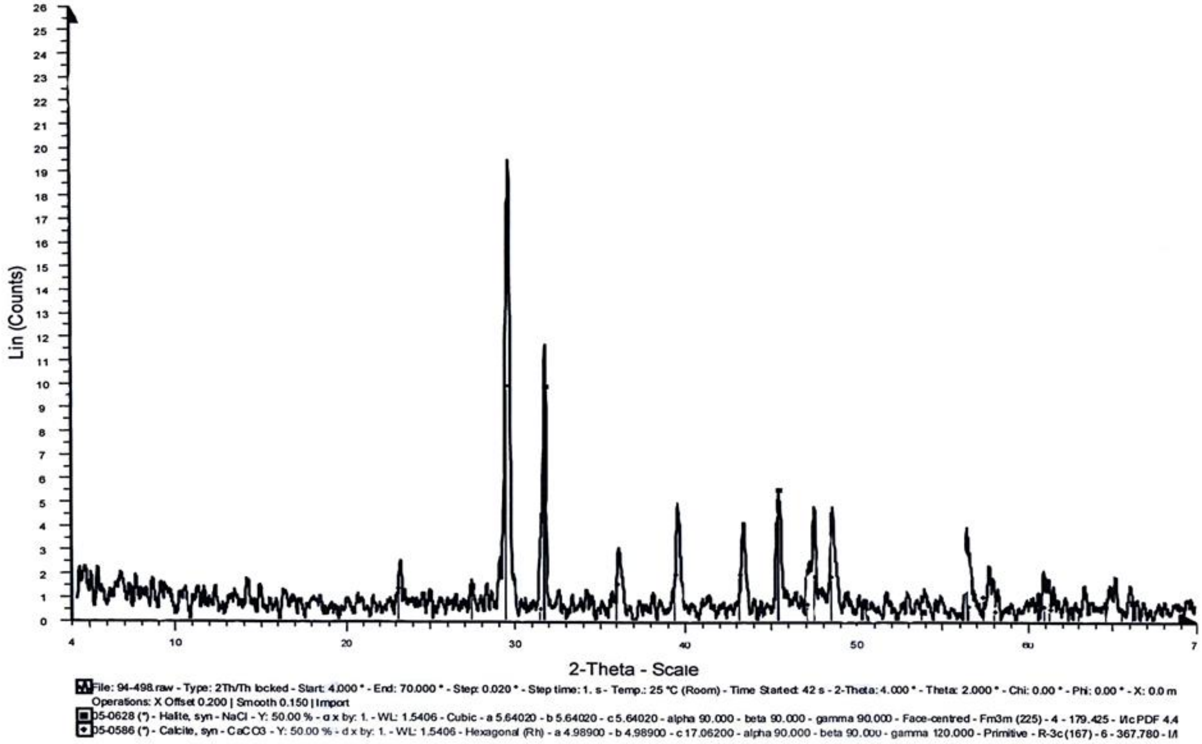

Fig. 8. XRD spectra of precipitated scales

EDX test in Fig. 11 shows the elemental analysis that verifies the simulation precision. The x axis is energy (keV) and the y axis is the intensity. As can be seen in Table 2, only carbon, oxygen and calcium were found through elemental analysis and only carbonate scales in the solution have been detected. The halite is probably formed because of drying the scale sample in oven and evaporation of sample's water. Halite (NaCl) scale is the most common scale type that is formed in this manner in HT/HP wells [44].

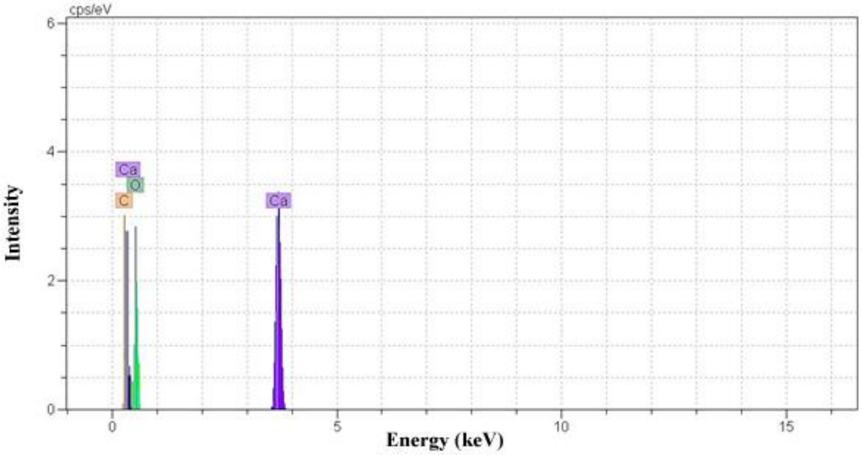

Fig. 11. EDX elemental analysis of precipitated scale

Table 2. Quantitative EDX elemental analysis of precipitated scale

Element	Series	unn. C (wt.-%)
Carbon	K series	8.53
Oxygen	K series	46.36
Calcium	K series	27.57
	Total	82.5 %

Fig. 12 shows a carbonate crystal formed in solution, featuring a regular hexagonal shape and compact structure. This photo was taken by an apparatus owned by Razi Metallurgical Research Center (RMRC). Its scale is 20µ and has been magnified 1500 times.

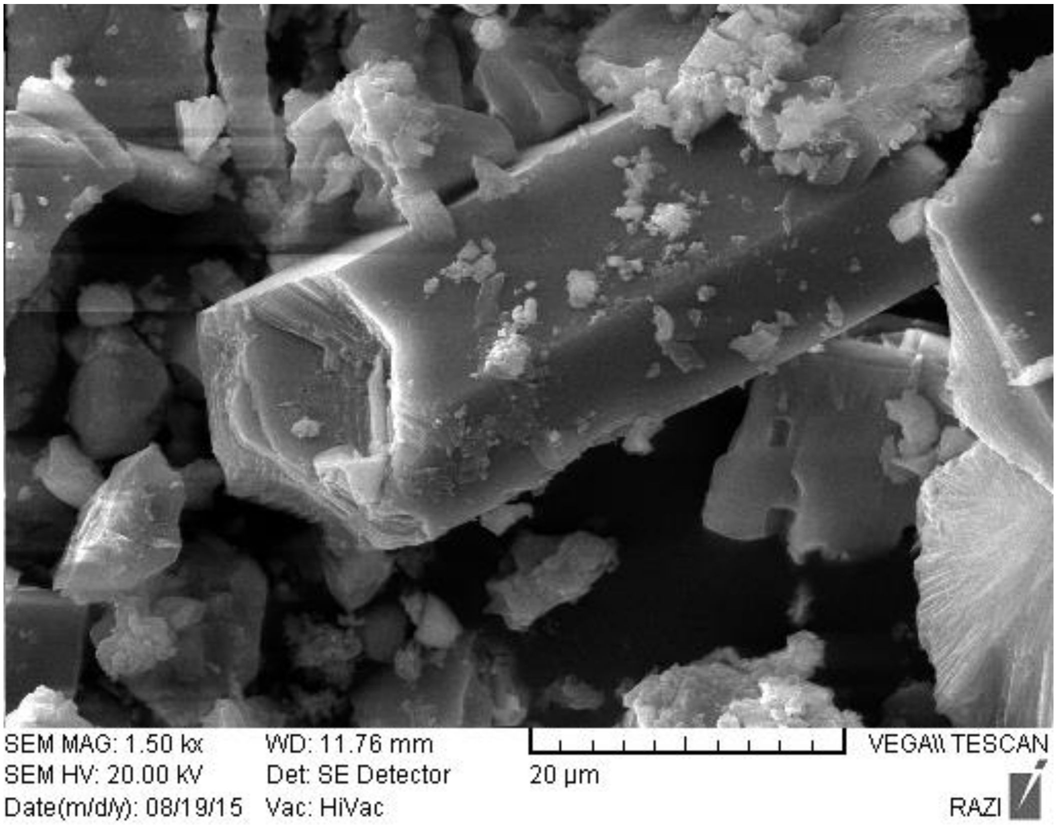

Fig. 9. SEM image of carbonated crystal formed in solution

6. Conclusion

In this study, first evaluation of scale precipitation potential in different mixing ratios of SW and FW were conducted. Then a mixing ratio of SW and FW with harsh scaling condition was determined. XRD and SEM tests were conducted to assess the model prediction. According to the obtained results, the following conclusions have been obtained:

- Simulation has shown that calcite and aragonite were the two leading scales in the investigation.
- Results demonstrate that the behavior of calcite and aragonite scale formation is very much alike.
- The results of the simulation indicate that generally in specific mixing ratio, pH was the main parameter that impacts the calcite and aragonite scales.
- XRD and SEM test result shows the crystal structure of calcite in the study is a regular hexagonal shape.
- XRD and elemental analysis confirms the simulation accuracy, which shows the ion association modeling is a reliable model to predict the formation of oilfield scales.

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