

## Static Modeling of Oil Field Mineral Scales: Software Development

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### ABSTRACT

Mineral scale deposition in near wellbore regions of injection wells is one of the main challengeable issues during the water injection process, which magnifies the importance of robust models in predicting the amount of mineral scale deposition such as calcium sulfate. One of the main challenges of CaSO<sub>4</sub> scale is in carbonated reservoirs, in which sensitive behavior is observed in related to the contribution of both calcium and sulfate ions in carbonated and sulfated scale reactions. This defect is mirror of wrong procedure and value in the estimation of first kind/value of precipitant contributed in scale deposition reactions (ions competition) as well as inconsistent temperature/pressure dependent coefficients of prediction model. The objective of this study is to develop a model that can accurately predict the formation and amount of CaSO<sub>4</sub> scale as the dominant scale in multicomponent aqueous systems by three major tools, namely utilization the best temperature- and pressure-dependent thermodynamic interactive ion coefficients (MSE Model: Pitzer), developing our fine-tuned iterative mathematical solver, and verification of the results of the model by accurate experimental data. The results showed that at the optimum value of precipitant (10%) in scale deposition reactions and by defining the best temperature- and pressure-dependent coefficients, we can attain the best accuracy in the prediction of CaSO<sub>4</sub> scale deposited amount (less than 0.06% as a relative error compared to 36% overestimation and 22% underestimation in commercial software). The output of this study is developed software leading to the more accurate prediction of the amount of promising scales in near wellbore regions or pipelines.

**Keywords:** CaSO<sub>4</sub> Scale, Laboratory Static Jar Tests, Iterative Mathematical Solver, Pitzer Thermodynamic Model, Ions Binary Interactive Coefficient

### INTRODUCTION

Seawater injection is one of the common methods used in IOR usually used in offshore oil fields to maintain reservoir pressure and improve secondary recovery. Scale deposition is one of the most serious oil field problems that inflict water injection

systems primarily when two incompatible waters are involved. Scale deposition can occur from one type of water because of super-saturation with scale forming salts attributable to changes in the physical conditions under which the water exists. Super-saturation can be generated in water by changing

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the pressure and temperature conditions or by mixing two incompatible waters. Scale can develop in the formation pores near the wellbore, which reduces porosity and permeability, and consequently productivity and injectivity decrease. It can block flow by clogging perforations or forming a thick lining in production tubing. Scale is also deposited in down-hole pumps, tubing, casing flow-lines, heater treaters, tanks, and other production equipment and facilities. The consequence could be production-equipment failure, emergency shutdown, increased maintenance cost, and overall decrease in production efficiency [1]. The most common oil field scales are listed in Table 1, along with the primary variables that affect their solubility [2].

**Table 1: Common oil field scales.**

Name	Chemical Formula	Primary Variables
Calcium Carbonate	CaCO <sub>3</sub>	Partial pressure of CO <sub>2</sub> , T, TDS, and pH
Calcium Sulfate	CaSO <sub>4</sub> ·2H <sub>2</sub> O	T, TDS, and P
Gypsum		
Hemihydrate	CaSO <sub>4</sub> ·H <sub>2</sub> O	-
Anhydrate	CaSO <sub>4</sub>	-
Barium Sulfate	BaSO <sub>4</sub>	T and P
Strontium Sulfate	SrSO <sub>4</sub>	T, TDS, and P
Ferrous Carbonate	FeCO <sub>3</sub>	Corrosion, Dissolved Gases, and pH
Ferrous Sulfide	FeS	Corrosion, Dissolved Gases, and pH
Ferrous Hydroxide	Fe(OH) <sub>2</sub>	-
Iron(III) oxide-hydroxide	Fe(OH) <sub>3</sub>	-

Based on the industrial reports, the four dominant scales are sulfates, such as calcium sulfate (anhydrite, gypsum), barium sulfate (barite), and strontium sulfate (celestite), and calcium carbonate. Other less common scales have also been reported such as iron oxides, iron sulfides, and iron carbonate. Lead and zinc sulfide scale have recently become a concern in a number of North Sea oil and gas fields [3].

Calcium sulfate scale poses a unique problem for the salts under consideration because it occurs with one of three different phases. Gypsum, the most common scale formed, occurs at relatively low temperatures. At high temperatures (above 100 °C), the stable phase predicted is anhydrite (CaSO<sub>4</sub>). However, hemi-hydrate has been known to form in the temperature range of 100 to 121 °C, especially in non-turbulent systems and in high ionic strength brines [4].

According to Oddo et al. [5], calcium sulfate scale formation is somewhat dependent on temperature, but is typically precipitated because of a decrease in pressure or an increase in the relative concentrations of calcium or sulfate. CaSO<sub>4</sub> solubility is fairly independent of pH, so it can readily precipitate in an acid environment. The solubility of calcium sulfate is also affected by temperature, salinity, and excess common ions.

The case where water injection (seawater, river, aquifer, or produced water) is used for pressure maintenance and sweep, the mixing of incompatible brines can lead to the formation of sulfate scales when the injection water contains sulfate ions [6]. Since there is sensitive behavior in the prediction of CaSO<sub>4</sub> scale deposition due to the contribution of both calcium and sulfate ions in carbonated and sulfated scale reactions, there is a strong motivation for developing accurate modeling tools that can predict the likelihood of CaSO<sub>4</sub> scale formation.

Also, other authors such as Bedrikovtsky (2006, 2009) addressed the injectivity decline due to sulfate scales deposition throughout the PWRI [7, 8]. Mackay discussed that sufficient concentrations of scaling ions delivered to the production well can necessitate squeeze treatments (inhibitor) [9].

Therefore, the authentic estimation of mineral scales precipitation is required. The main difficulty in scale prediction modeling lies in distinguishing the ions competition behavior of multicomponent mixtures. One of the main defects in previous studies is related to wrong procedure in the sequence and value of precipitants taken part in mineral scale deposition reactions at each stage of modeling.

The objective of this study is to develop a model that can accurately predict the formation of  $\text{CaSO}_4$  scale deposition as well as mineral scales in multicomponent aqueous systems by three major tools: huge experimental data bank, including the implemented static tests, gathered temperature and pressure dependent coefficients, and a finely tuned iterative mathematical solver in which the previous described defect was removed. The next parts of this paper are organized in the manner of the following structure. First, we introduce the computational model briefly and determining the used parameters. The mathematical methodology for solving the governing equations has been presented in solving methodology, i.e. section 2. Section 3 contains the results and discussions of the performed Jar tests as the static scale deposition tests as well as the finely tuned mathematical model. In the last section, i.e. section 4, the conclusions are reported to show the accuracy of the proposed mathematical model on the prediction of mineral scale deposition in a specific thermodynamic condition.

## EXPERIMENTAL PROCEDURES

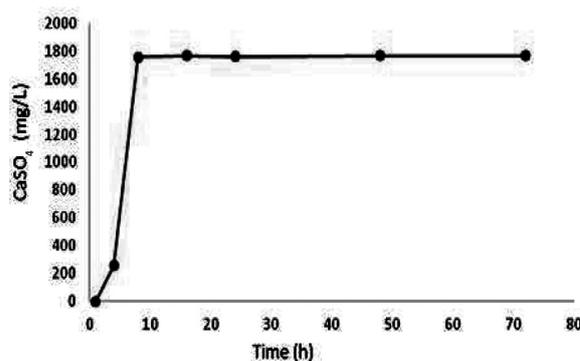
### Experimental Static Test

To obtain the observed scale precipitation results regarding the anhydrite and compare the results of the model as well as other sources (OLI and StimCade) with the experimental ones, static tests were designed and implemented in laboratory.

In order to obtain the anhydrite scales at different ratios of injection to formation water,  $\text{Na}_2\text{SO}_4$  and  $\text{CaCl}_2$  was used to provide required divalents for the water phase (see Table 2 for the compositions of the water phase). After that, the formation water was mixed with injection one at different ratios, and after an optimum time, the solution was filtered with 0.45 micrometer paper filter. The filter was warmed in an oven at optimum time and temperature. The dried filter was weighed and the amount of scales (mg/L) was calculated. Figure 1 shows the stability of the formed scale regarding the time.

**Table 2: Composition of synthetic waters for the prediction of individual scale formation.**

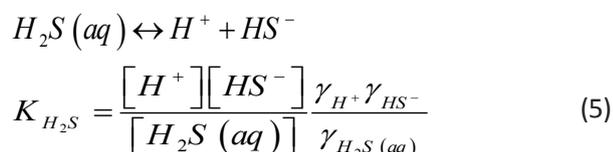
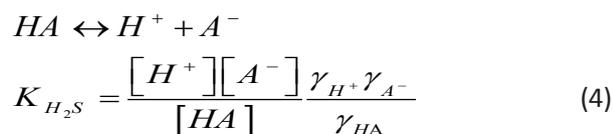
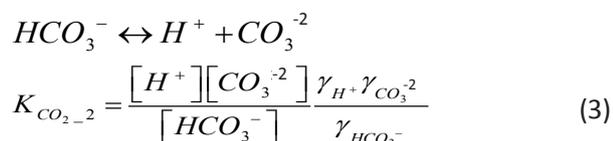
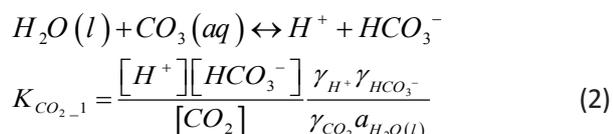
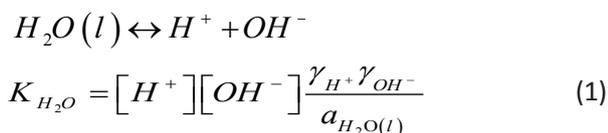
Formation water	Value (mg/L)	Sea water	Value (mg/L)
Calcium	12244	Sulfate	7000
Chloride (Brine)	129758	Sodium	1627
Sodium (Brine)	84068	-	-
Chloride (Calcium)	21734	-	-



**Figure 1: Effect of time on formation of anhydrite scale.**

## Computational Model, Model Description, Aqueous Chemistry

At the first stage, chemical reactions in the liquid phase have been introduced. The dissolved species in the aqueous phase as well as the satisfaction of the mass-action equations for all the possible association reactions between the master species at every transport node are assumed to be in thermodynamic equilibrium. The association reactions between the master species and the corresponding mass-action equations accompanied by the master species are defined in Equations 1-5.

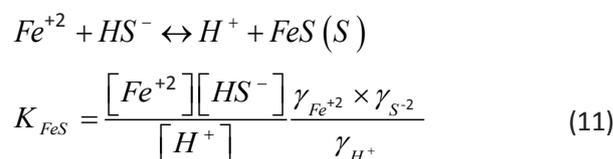
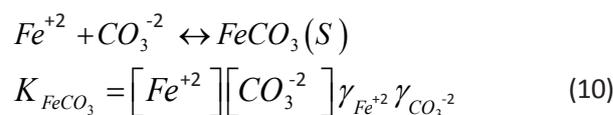
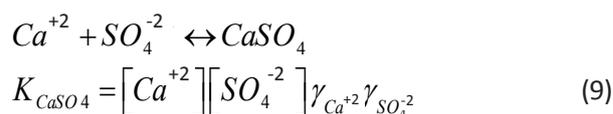
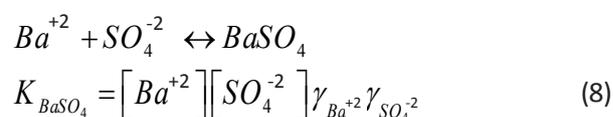
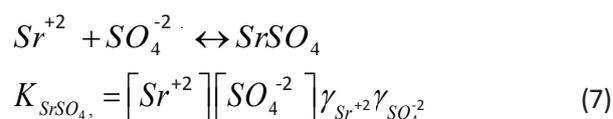
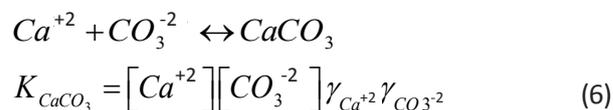


Activity coefficients are calculated with the Pitzer model (See Appendix A) [10].

## Mineral Dissolution and Precipitation

At the second stage, to consider the effects of mineral dissolution and precipitation, promising mineral

phases: calcite ( $CaCO_3$ ), siderite ( $SrSO_4$ ), barite ( $BaSO_4$ ), anhydrite ( $CaSO_4$ ), siderite ( $FeCO_3$ ), and iron sulfide ( $FeS$ ), which can precipitate from the aqueous solution, were considered (Equations 6-11).



The temperature and pressure dependence of the equilibrium constants are calculated using Pederson and Christensen correlations (2006) [10].

## Determination of Parameters in Model

In case of incompatibility between formation and injection water during the water injection process, it is essential to have accurate data on the thermodynamic properties of the mixed electrolyte solutions. The activity coefficients are of primary importance in accurately describing the thermodynamic behavior of aqueous mixed electrolyte solutions. Among recently developed models of electrolyte solutions, ionic interaction models provide the simplest

and most coherent procedures for calculating the thermodynamic properties of electrolyte components. An ion interaction model for predicting the activity coefficients of mixed electrolyte solutions was developed by Pitzer in early 1970s. The Pitzer model extended the Debye-Huckel method, using a virial expansion to account for the ionic strength dependence of the short-range forces in binary and ternary ion interactions [10].

The adjustable parameters for the model are the single electrolyte parameters,  $\beta_{0,MX}, \beta_{1,MX}, \beta_{2,MX}$  and  $C_{\phi,MX}$  as well as the mixed electrolyte parameters  $\theta_{ij}$  and  $\psi_{ijk}$  for the significant minerals. Researches have previously evaluated most of the electrolyte parameters needed for this system using osmotic coefficient data. Table 3 summarizes the references used to extract the required coefficients and parameters in Pitzer model. Selected  $\theta_{ij}$  and  $\psi_{ijk}$  parameters are consistent with the high concentration solubility data while they slightly reduces the accuracy of the equations in the low concentration range. Since terms in  $\theta_{ij}$  and  $\psi_{ijk}$  are relatively insignificant for low concentrations, the loss of accuracy is minimal in the context of solubility prediction.

Different papers have used different empirical functions describing the variation of Pitzer's parameters with respect to temperature and pressure (11, 12, 13, 14, 15, 16 and 17, 18). For computational ease, these functions can be recast into the following ten parameter expression using simple algebraic transformations (Eq. 12). X(T) can be either Pitzer's parameters  $\beta_{0,MX}, \beta_{1,MX}, \beta_{2,MX}, C_{\phi,MX}, \theta_{ij}$  or  $\psi_{ijk}$

$$X(T) = \frac{a_1}{T} + a_2 + a_3P + a_4 \ln(T) + (a_5 + a_6P)T + (a_7 + a_8P)T^2 + \frac{a_9 + a_{10}P}{T - 227} + \frac{a_{11} + a_{12}P}{680 - T} \quad (12)$$

**Table 3: References of the required coefficients and parameters in Pitzer model**

Mixed electrolyte binary interaction parameters ( $\beta_{0,MX}, \beta_{1,MX}, \beta_{2,MX}$ and $C_{\phi,MX}$ ) at 25°C	Harvie et al., 1984 [11]
Mixed electrolyte ternary interaction parameters and ion-neutral interaction coefficient ( $\theta_{ij}$ and $\psi_{ijk}$ ) at 25 °C	Harvie et al., 1984 [11]
Debye–Huckel slope for the activity coefficient, $A^\phi$	Moller, 1988 [12]
Values of the fitting constants Eq.12 for the binary interaction parameters for aqueous electrolytes	Christov and Moller, 2004 [13]; Marion, 2001

### Solving Methodology

In this paper, an iterative algorithm is developed for modeling and the calculation of the amount of precipitates in a super saturated mineral solution. Iterative calculation for the prediction of mineral scales was introduced in previous studies [10]. Yuan and Todd used an iterative model for the prediction of sulfate scales and did not consider the calcite scale. They performed the modeling by applying the sequence of barite, Celestin, and anhydrate precipitation reactions to the calculation procedure. Pedersen firstly analyzed the ion products of the iron minerals ( $FeCO_3$  and  $FeS$ ) against the solubility products. Afterward, the ion product of calcium carbonate is checked against its solubility product. In case of precipitation possibility, double-loop iteration is employed to calculate simultaneous precipitation of calcium carbonate and iron minerals. In a recent study on scales modeling by iterative solvers, Li performed the modeling of calcite and anhydrate scales solely and did not consider barite and celestite scales. He used the iterative calculation on mass and charge change due to scales precipitation with the conditions of lack of change in activity coefficients.

In the specified studies, the calculation algorithms are based on the specific order of reactions and successive calculations some of which are nested. The main challenge in literature, which is the source of error in the estimation of scale precipitation, is lack of possibility to simultaneously calculate the individual solid precipitation in multicomponent mixtures.

To handle the described challenge in literature, two main assumptions are considered; the first one is the simultaneous competition of ions contributed in mineral scale reactions as the common cations/ anions with the different reaction rates. Secondly, the formed mineral scales are proportional to the total concentration. With these assumptions, precipitation ratio coefficient (PRC) would be defined as the ratio of precipitant taken part in reactions at each stage of modeling and is between 0 and 1. The amount of mineral scale for each compound with positive SI is equal to the total potential of precipitation multiplied by PRC. At the end of each stage, the new equilibrium concentrations of ions would be calculated and the updated values of activity coefficients can be obtained by the Pitzer's model (Appendix A). These stages proceed up to the step in which SI changes into zero or negative. The cumulative mineral scale precipitations can be calculated by summation of the experiment steps. In this mathematical solver, convergence speed and calculation error is exactly proportional to the value of PRC. Sensitivity analysis was carried out to obtain the optimum value of PRC in which the modeling results are consistent and matched with the experimental results and are independent of the sequence of scales formation reactions. Figure 2 shows the algorithm applied to the modeling.

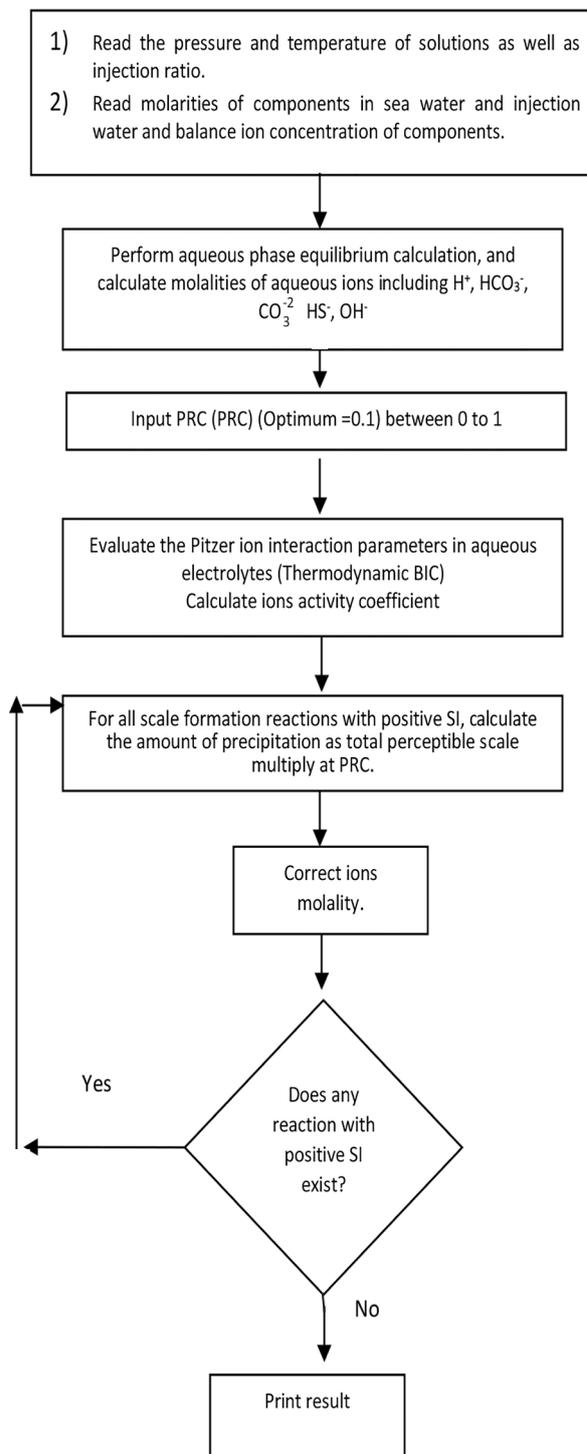


Figure 2: Algorithm applied to mineral scale precipitation modeling.

## RESULTS AND DISCUSSION

A specified algorithm (Figure 1) was written in Visual Basic (V.B.) programming language and the following results have been obtained. Figure 3 shows the executive input window of the developed software.

In this window, all of the required parameters for the model, including ions compositions and TDS as well as pH, are inserted and pushing the check data button, the charge balance will be carried out to commence the ions equilibrium reactions calculations. Figure 4 shows the executive output window, in which calculation procedure will be done in a specific thermodynamic condition and different mixing ratios for the case study.

Also, the graphical charts will be plotted in the output window simultaneously.

To validate the parameters used in the model, the solubility of one individual solid (anhydrate) was calculated and compared to the literature results at different temperatures and at the vapor pressure by applying the algorithm in Figure 5.

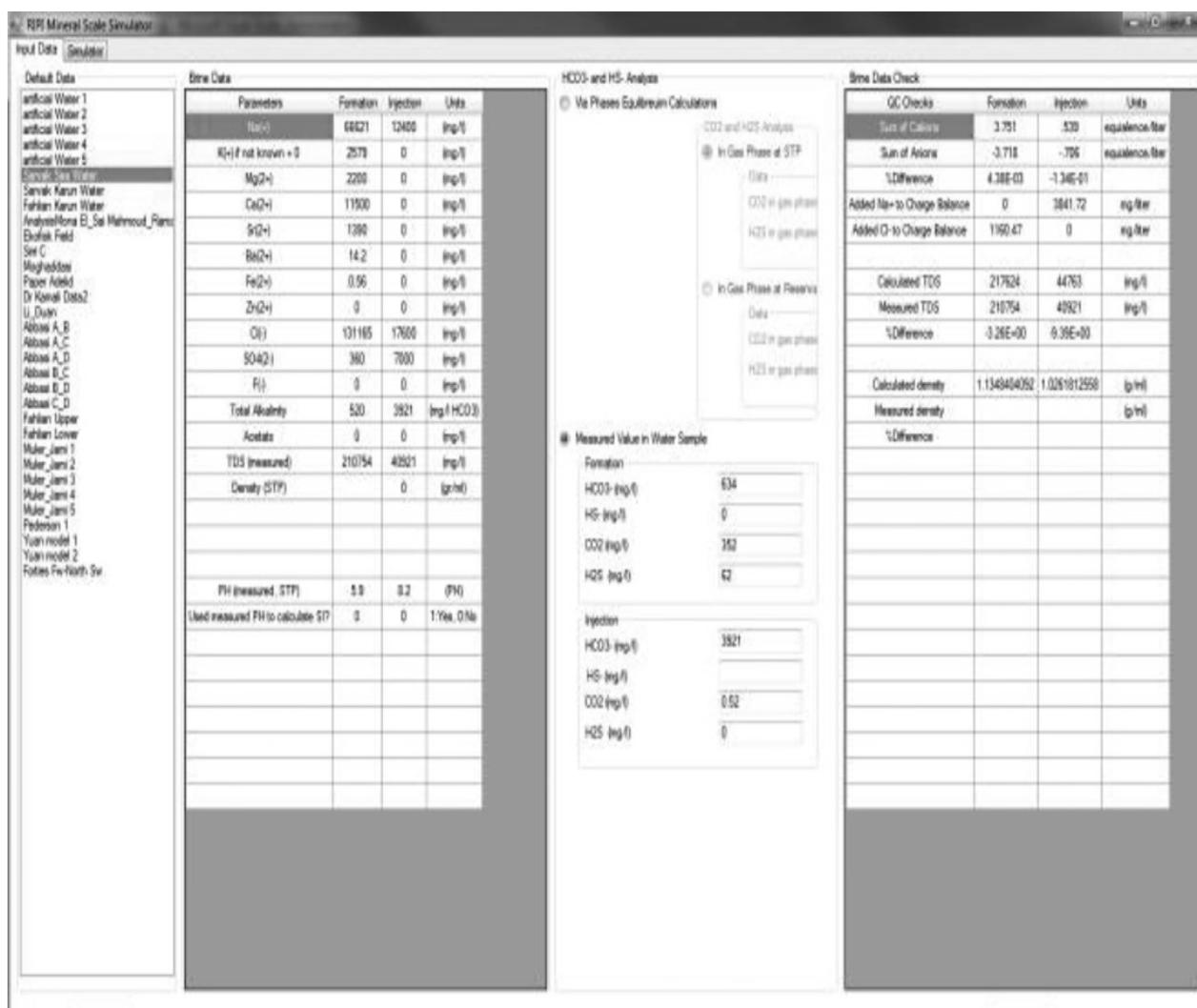


Figure 3: Input window of the developed software.

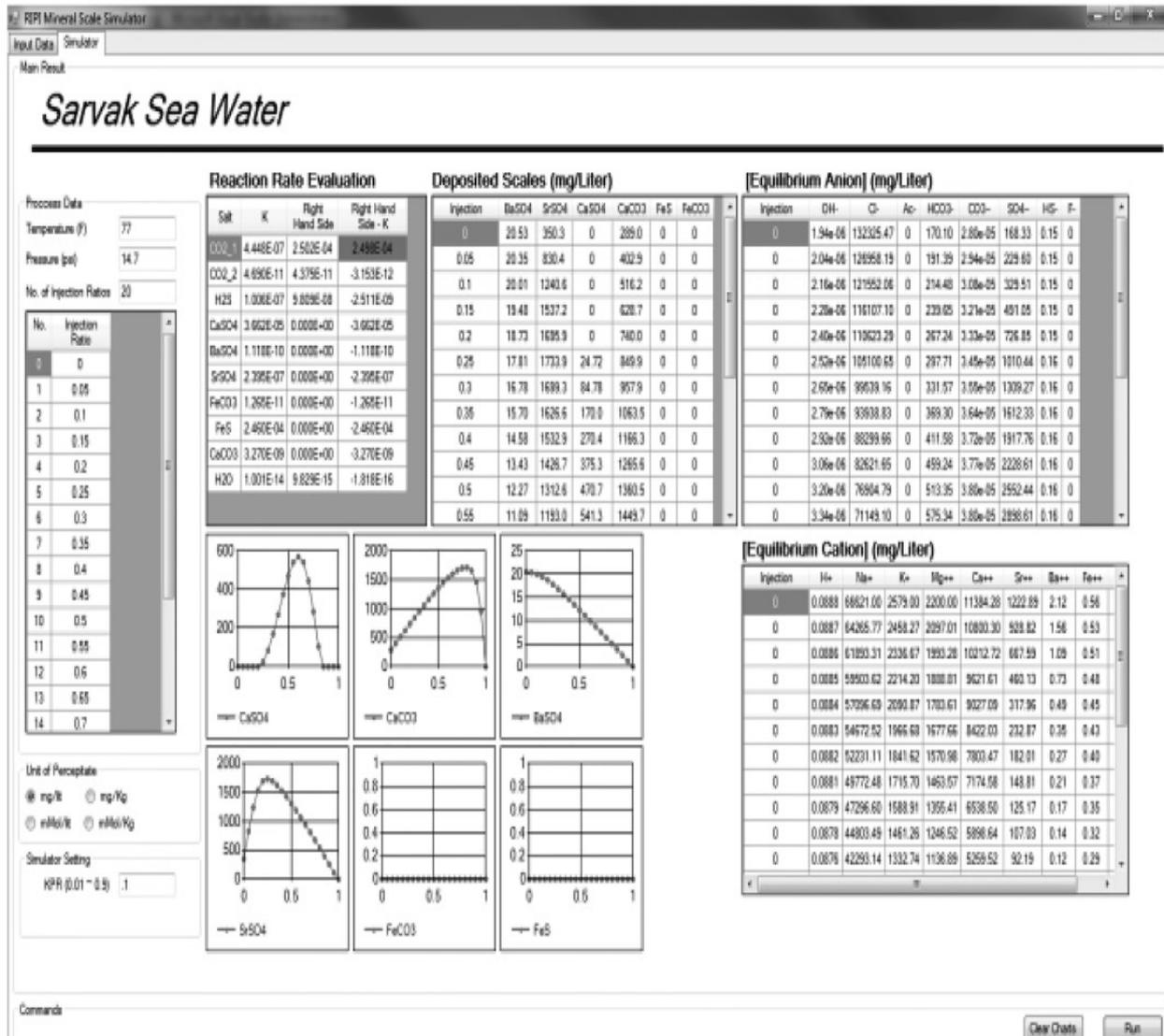


Figure4. Executive output window of developed software

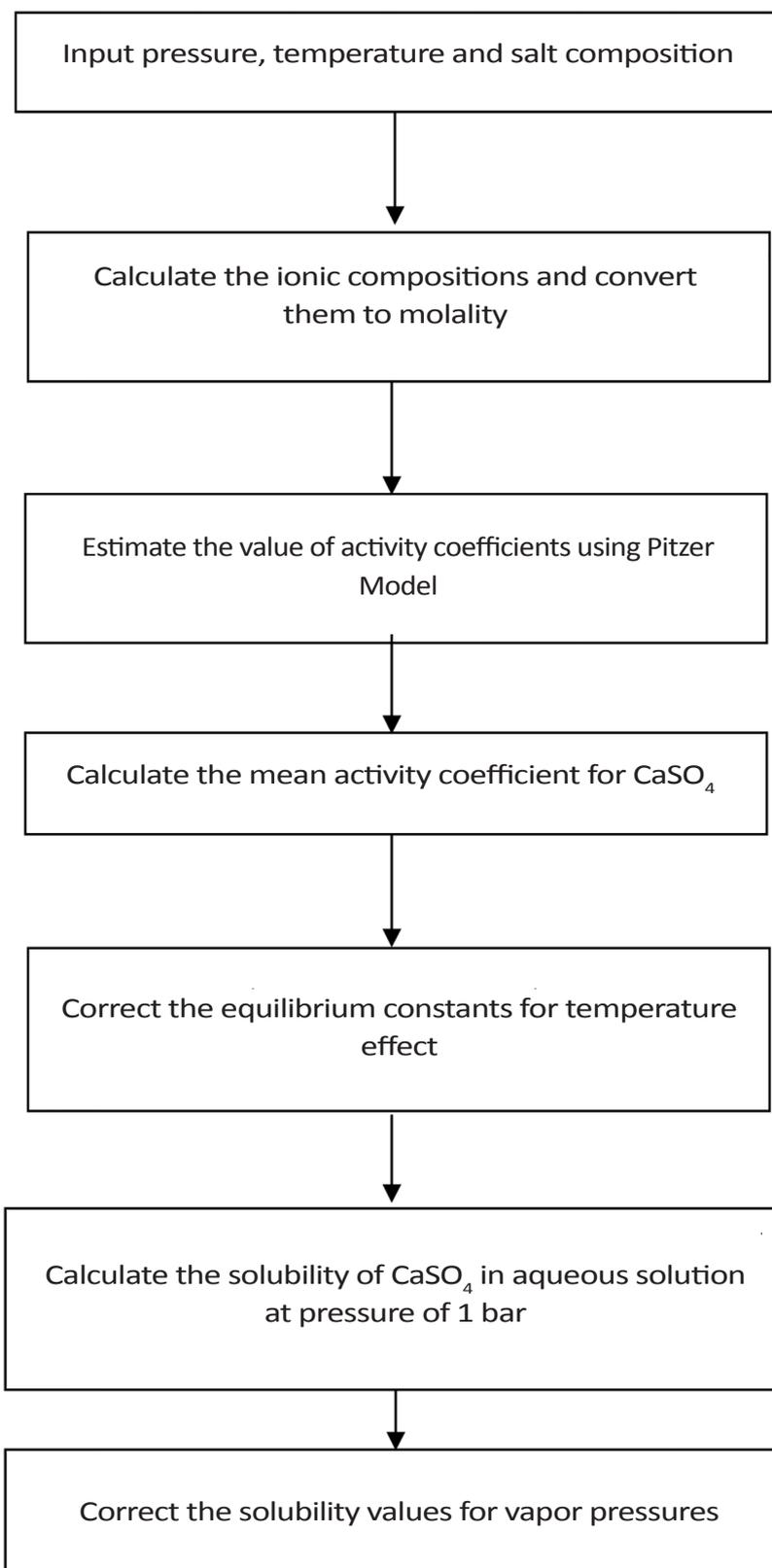


Figure 5: Algorithm Applied to solubility calculation.

The result of the comparison of  $\text{CaSO}_4$  solubility in pure water is shown in Figure 6. As can be observed, there is a good match between our model and the previously developed models, which reflects using of suitable pressure and temperature dependent parameters as well as adjustable parameters for the model.

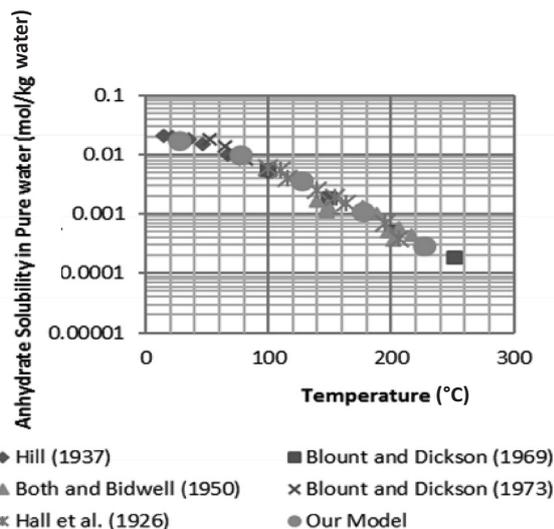


Figure 6: A comparison of the calculated anhydrite solubility in water using our model with the experimental data.

Since there is  $\text{CaSO}_4$  as one of the dominant scales in blending the used case studied water (Sarvak) with sea water (Table 2), in the second stage, the scale precipitation of individual solid ( $\text{CaSO}_4$ ) has been assessed with the valid proposed model. In order to solely monitor the formation of one type of mineral scale, synthetic waters have been used as the formation and injection sea waters (see Table 3 for their compositions). By blending the used waters at different mixing ratios, the results for  $\text{CaSO}_4$  scales at different ratios are obtained in both modeling and experimental sections. The comparison between the experimental data and the model results as well as commercial software ones is shown in Figure 7.

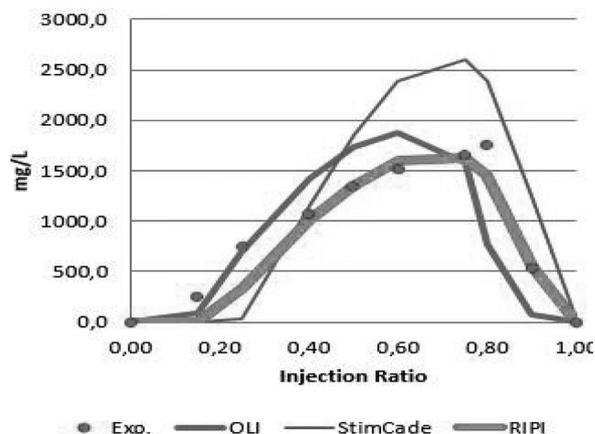


Figure 7: Comparison of mineral scale precipitation in the developed model, different common tools, and laboratory tests (at  $T=25\text{ }^\circ\text{C}$ ,  $P=1\text{ bar}$ ).

Different software packages use different solvers but one model, i.e. Pitzer/modified Pitzer, to predict the precipitation and most of the difference in the results is referred to different algorithms defining ions competition through the scaling reactions.

To be sure about the kind of formed scale as individual solid, SEM analysis was performed on each stage of individual solid static Jar tests (Figure 8).

To verify the obtained results in the section of individual scales and use the computational model on an industrial scale, real formation water (Case study; Sarvak) was mixed with sea water in two distinct formation water/sea water mixing ratios of 0.5 and 0.75 (see Table 4 for the compositions of the candidate waters). Modeling was conducted at all the mixing ratios and the results were compared with the experimental data and commercial software results (Figure 9). Mixing ratios in the experimental studies were planned from 0.25 to 0.75 (0.25, 0.5, and 0.75). Since the main difference between commercial software and the developed model was in the scope of mixing ratios between 0.4 and 0.9, two points of experimental data (0.5 and 0.75) were selected to be compared with the available sources.

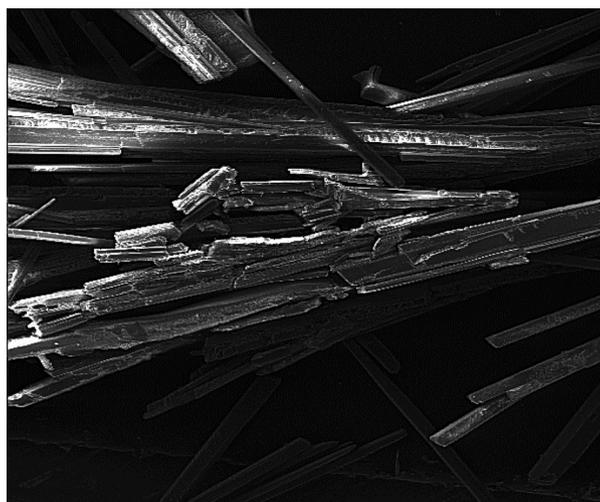


Figure 8: SEM image of anhydrate scale.

Table 4: Composition of Sarvak formation water and sea water for the prediction of scales formation in a real case.

Component	Formation Water Value (mg/L)	Sea Water Value (mg/L)	Component	Formation Water Value (mg/L)	Sea Water Value (mg/L)
Cations			Anions		
Sodium	66621	17600	Chloride	131165	12400
Potassium	2579	-	Sulfate	360	7000
Calcium	28750	-	Sulfite	48	-
Magnesium	5500	-	Bicarbonate	634	3921
Ferrous Iron	0.56	-	Nitrate	63.4	-
Barium	14.2	-	Nitrite	1.00E-02	-
Strontium	1390	-	Neutrals	-	-
Carbon dioxide, aq.	352	-	Hydrogen sulfide, aq.	62	-
Total Dissolved Solids, mg/L	237125	40921	Measured Density, g/cc	1.1441	1

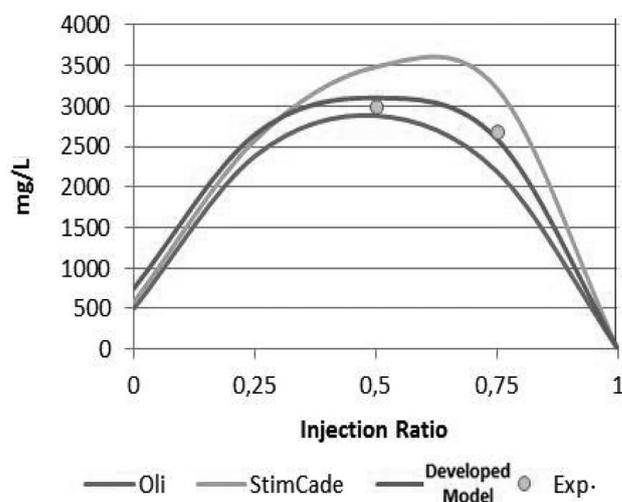


Figure 9: Comparison of total scales precipitation by different sources

To validate the developed model, a sensitivity analysis was performed on two main criteria, namely the sequence of scale precipitation and PRC. Two different patterns of precipitation at different PRC values were planned to check the dependency of model on the sequence of scale precipitation as well as PRC; A: ( $\text{CaCO}_3$ - $\text{SrSO}_4$ - $\text{CaSO}_4$ - $\text{BaSO}_4$ ) and B: the opposite sequence of A. Figures 10 and 11 show that the obtained results are independent of the sequence of scale precipitation and PRC in all scales, except  $\text{CaSO}_4$ .

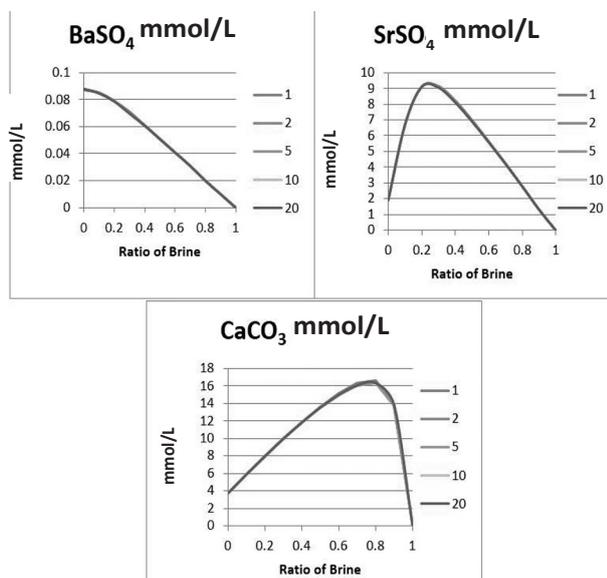


Figure 10: Sensitivity analysis on the sequence of scale precipitation and PRC in pattern A.

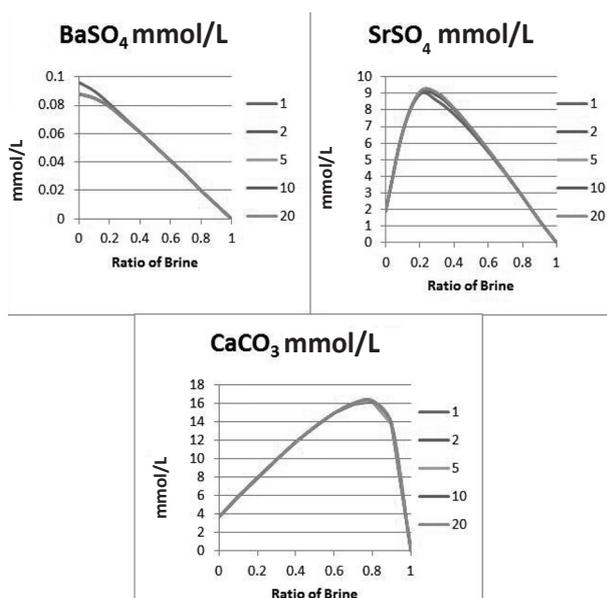


Figure 11: Sensitivity analysis on the sequence of scale precipitation and PRC in pattern B.

In case of  $\text{CaSO}_4$ , a significant change was observed in both aspects of changes in PRC values and the sequence of scale precipitation (Figure 12). The origin of this sensitive behavior in  $\text{CaSO}_4$  is related to the contribution of both calcium and sulfate ions in carbonated and sulfated scale reactions. However, it was approved that this dependency would be diminished as the PRC value approached its optimum value (10%) and would be omitted approximately for lower values of PRC (Figure 13).

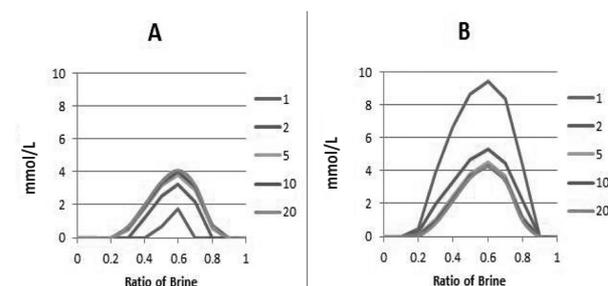


Figure 12: Sensitivity analysis on the sequence of scale precipitation and PRC in  $\text{CaSO}_4$  Scale; A: Carbonate/sulfate; B: Sulfate/carbonate.

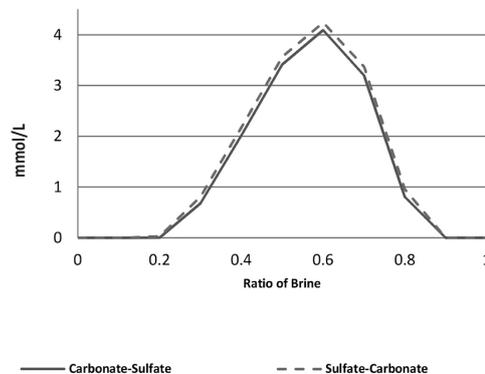


Figure 13: Independency of scale precipitation amount from the sequence of scale precipitation in PRC=5%.

As observed in Figures 10 to 13, the developed model is independent of the sequence of scale precipitation as well as PRC values in each step.

## CONCLUSIONS

The results of model developed for predicting the formation of mineral scales in multicomponent aqueous systems caused by mixing incompatible waters (sea water and formation water) and by changing thermodynamic conditions are consistent with experimental results for

CaSO<sub>4</sub> and with the previous scale prediction models and commercial software (OLI Scale Chem, StimCade). Briefly, the following conclusions can be drawn according to the results of the developed model:

- The main advantageous of the present model is related to applying a procedure to the sequence and value of precipitants taken part in mineral scale deposition reactions at each stage of modeling.
- At the optimum value of precipitant (10%) in scale deposition reactions and by defining the best temperature and pressure dependent coefficients, a relative error of less than 0.06% was observed compared to commercial software with 36% overestimation and 22% underestimation in the prediction of CaSO<sub>4</sub> scale deposited amount.
- The origin of sensitive behavior in CaSO<sub>4</sub> scale prediction is related to the contribution of both calcium and sulfate ions in carbonated and sulfated scale reactions (ions competition).
- The sensitivity analysis of our developed model shows that the scale prediction results are independent of the sequence of scale precipitation and PRC in all scales, except CaSO<sub>4</sub>. In the case of CaSO<sub>4</sub>, scale precipitation curves approach each other in PRC values lower than 10% for both defined reaction sequences, and the amount of CaSO<sub>4</sub> scale become constant.

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