



# Prediction mineral scale formation in oil reservoirs during water injection



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## ABSTRACT

Mineral scale formation during incompatible water injection into aquifer of oil reservoirs is one of most challenging problem in IOR method. Thermodynamic modeling of incompatible mixing of formation and injection brines before water injection operation could prevent the scale formation in oil reservoirs and injection and produced wells. Using the solid–liquid equilibrium, ENRTL activity coefficient model, mass balance and electroneutrality equations a new method for prediction of different mineral scale formation is presented. Using the experimental mean activity coefficient and solubility data, the binary interaction parameters of ENRTL activity coefficient for studied salts are optimized. Also, the correlation of the solubility product of the main sulfate salts such as  $\text{CaSO}_4$ ,  $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ ,  $\text{BaSO}_4$ ,  $\text{SrSO}_4$  in vast range of pressures are obtained through fitting the solubility data. Once scaling index becomes greater than zero, the precipitation amount of salt(s) is obtained by mass balance and electroneutrality equations. The present model is compared to previous models for different case studies. The results verify the prediction of new model for determination of scale type and amounts.

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## 1. Introduction

Water injection into oil reservoir aquifer as improving oil recovery (IOR) method has been used for many years. Incompatible mixing between injection and formation waters yields scale formation in oil fields. Removing the precipitated scale in produced well, injection well and perforations are expensive and sometimes impossible. Thus, it seems prevention of the scale formation is better than treatment of reservoir. One of the efficient ways is using a thermodynamic method for prediction of scale formation potential in oil fields. An appropriate thermodynamic model should determine the type of the precipitated scale salt and maximum amount of precipitant in the mixed injection and formation waters [1]. Moreover, supersaturation and scaling index of each salt in the different brines or mixing of brines would be obtained using the thermodynamic model at specified temperature and pressure. In recent years many researches have been focused on this area. Vetter et al. [2] presented a model for prediction of simultaneous precipitation of barium, strontium and calcium sulfates. In this model, they supposed that barium sulfate as most insoluble salt

precipitates first and strontium sulfate tends to precipitate afterward and finally calcium sulfate precipitates in the brine. Yuan and Todd [1] developed a predictive model for calculation of sulfate salt precipitation due to incompatible mixing and pressure and temperature changes for oil reservoirs. The Pitzer activity coefficient model [3] was used to consider the non-ideality of solution at reservoir conditions. Moreover, the simultaneous precipitation and scaling tendency of sulfate salts including barium, strontium and calcium sulfates was considered. An equilibrium model was provided for prediction of sulfate and carbonate salts formation at reservoir temperature and pressure by Haarberg et al. [4]. The initial brine composed of  $\text{Ca}^{2+}$ ,  $\text{Sr}^{2+}$ ,  $\text{Ba}^{2+}$ ,  $\text{CO}_3^{2-}$  and  $\text{SO}_4^{2-}$  where  $\text{CO}_2$  and common organic acid could dissolve in so that the aqueous solution is in equilibrium with oil and gas phases. Furthermore, the equilibrium precipitation reactions, species mass balance and electroneutrality of brine were recognized in this model. The Gibbs energy of solution was calculated using UNIQUAC [5], Brønsted–Guggenheim [6] and Debye–Hückel [7] models as short range, medium range and long range term, respectively. By minimizing the Gibbs energy of the solution, the equilibrium calculation of the solid–liquid was carried out. Then using the initial and equilibrium concentration of species, the precipitation amount in the solution was obtained. Yeboah et al. [8] developed a predictive model that calculates the scale formation potential and distribution in oil fields using the thermodynamic and kinetic data. The input data for modeling is

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AAD	average absolute deviation
$A_0$	Debye–Hückel constant
$G$	Gibbs free energy (J)
$I$	ionic strength
$K_{sp}$	solubility product
$N_p$	number of experimental data
O.F.	objective function
$P$	pressure (bar)
$R$	gases global constant (kJ/kmol K)
$T$	temperature (K)
$X$	local mole fraction
$g$	Gibbs free energy per mol (J/mol)
$m$	molality (mol/kg solvent)
$s$	solubility
$x$	mole fraction
$z$	charge number of ionic species

#### Greek symbols

$\alpha$	adjustable parameter
$\beta$	adjustable parameter
$\gamma$	activity coefficient
$\lambda$	interaction energy parameter
$\nu$	stoichiometric coefficient
$\rho$	closest approach parameter
$\tau$	ENRTL binary Interaction energy parameter

#### Superscripts

E	excess
cal	calculated
exp	experimental
$n$	hydration number
*	unsymmetrical convention
$\infty$	infinite dilution

#### Subscripts

LR	long range
PDH	Pitzer–Debye–Hückel
SR	short range
$a$	anion
$c$	cation
$i,j,k$	any compound
CA	salt
$m$	solvent
$x$	mole fraction basis
O	reference state
$\pm$	mean

the composition of the produced water and thermochemical data for determination of scale thermodynamic potential. When scale formation was predicted by using software, the distribution of the salt along the produced well is calculated based on the kinetic and well data. Scaling index (SI) for precipitation of barium, strontium and calcium sulfate and calcium carbonate salts were performed by Oddo and Thomson [9]. The proposed SI was a polynomial relation which was a function of temperature, pressure and ionic strength. For the calcium carbonate SI, CO<sub>2</sub> concentration in brine was also included. Thomas et al. [10] developed a professional model for prediction of scale precipitation and formation damage in oil fields during production and rock–fluid interactions. Moghadasi et al. [11] represented a model based on the experimental data and semi-empirical relations which could describe scale precipitation in one of the Iranian oil field during water injection for pressure maintenance. The model was based on the incompatible mixing in aquifer

of reservoir and considered the effect of temperature, pressure and pH of solution. MacKay [12] developed a model for calculation of scale formation in produced well. Using the geometry of reservoir, geometry of well and precipitation reaction rate, the position relevant to maximum scale precipitation and composition of final brine in produced well were obtained. Sheikholeslami [13] defined scaling potential index (SPI) as the difference of between the measured pH and saturated pH of the brine for studying of the calcium carbonate scale formation. In this model, the saturated pH was calculated using the equilibrium constants, calcium carbonate solubility product and Pitzer [3] activity coefficient. When SI is greater than zero, the calcium carbonate precipitates. Garcia et al. [14] developed an accurate thermodynamic model for calculation of the sulfate and carbonate salts solubility under hydrothermal conditions, i.e. up to 300 °C and 1000 bar. They studied the binary, ternary and quaternary systems of ions including Ba<sup>2+</sup>, Sr<sup>2+</sup> and Ca<sup>2+</sup> species. The main concept of that model was relayed on vapor–liquid–solid equilibria for carbonate salts such as CaCO<sub>3</sub> and solid–liquid equilibria for sulfate salts such as CaSO<sub>4</sub>. The activity coefficient of ion and molecular species were calculated through using the extended UNIQUAC model [15]. Bin Merdhan et al. [16] developed a simple kinetic model for precipitation of barium sulfate in porous medium using the solubility product, temperature, pressure and brine composition. A summary of the last works in modeling of thermodynamic scale formation in porous and non-porous medium is given in Table 1.

In this work, the scaling index (SI) of each salt in the present brines or mixed brines is calculated using the E-NRTL activity coefficient and solubility product relation. The solubility product is calculated at given temperature and pressure by backbone of thermodynamic relations and experimental solubility data. Moreover, the parameters of the E-NRTL function are optimized for several important salts particularly the sparingly soluble salts such as BaSO<sub>4</sub>, SrSO<sub>4</sub>, CaSO<sub>4</sub> and CaSO<sub>4</sub>·2H<sub>2</sub>O for wide range of temperatures and salt molalities. The calculated solubility of the mentioned salts by E-NRTL model is agreed well with the experimental data. When SI becomes greater than zero for each salt(s), using the mass balance of species in brine and electroneutrality condition the amount of precipitation salt(s) would be obtained. In salt precipitation models, the iterative or sequence algorithm is often used that it starts from low soluble salt, i.e. BaSO<sub>4</sub>, to high soluble salt, i.e. CaSO<sub>4</sub>·2H<sub>2</sub>O. Thus, this method does not usually consider the competition of common ions such as SO<sub>4</sub><sup>2-</sup>, Cl<sup>-</sup>, etc. Using the present modeling, all solid–liquid equilibrium equations are solved simultaneously so that the common ion effect could be taken into account in the present model. Furthermore, the model is tested for different brines especially combination of Forties brine and North Sea water so that their results are compared with those of the previous models.

## 2. Solubility product relation versus temperature

For investigation of the solid–liquid equilibrium and calculation of mineral salt solubility at variable temperature and pressure, solubility product is given as [17]:

$$K_{sp}(T, P) = v_C^{\nu_C} v_A^{\nu_A} (m\gamma_{\pm})^{\nu} a_w^n \quad (1)$$

where  $m$ ,  $\gamma_{\pm}$ ,  $a$ ,  $\nu$  and  $n$  refer to salt solubility, mean activity coefficient, activity, stoichiometric coefficient and hydration number, respectively. The subscripts C, A and w indicate cation, anion and water, respectively.

On the other hand, solubility product of mineral salts at different pressure and temperature could be shown as [14]:

$$\ln K_{sp}(T, P) = \ln K_{sp}(T, P_0) + \alpha(P - P_0) + \beta(P - P_0)^2 \quad (2)$$

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