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Thermodynamic Study of Water Activity of Single Strong Electrolytes

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Abstract

Today, due to the natural decline of oil exploitation, the use of methods of oil recovery, has made significant progress. However, these methods are accompanied by accumulation and deposition of mineral deposits in oil field installations. In the present study, aqueous solutions, strontium sulfate, barium sulfate, manganese sulfate and nickel sulfate are studied, in terms of EUNIQUAC model and genetic algorithms. Based on the findings of this article, as temperature increases, in order to increase the solubility of the system, the ionic strength decreases; but with increasing pressure, the solubility of barium sulfate increases. Meanwhile, in this article, to evaluate water activity, aqueous solutions of manganese sulfate and nickel sulfate is studied.

Keywords: Mineral Ions, Ionic Strength, Water Activity, EUNIQUAC Model.

1. Introduction

Today, managing improved oil recovery method using water injection, despite significant economic gains, can cause mineral deposits phenomenon because of the water injected impurities. Mineral deposits in oil fields (due to water injection), if the necessary conditions are aggregation and sedimentation. Sulfate mineral deposits such as calcium sulfate, barium sulfate, strontium sulfate, are of the most important common sediments in oil fields. In recent decades, the solubility and the formation of mineral deposits in the petroleum and industrial systems, have attracted more attention [1]. In order to investigate permeability properties of porous media in oilfields where mineral sediments were aggregated and precipitated, Moghadasi et al. [2] performed a research work based on empirical and theoretical studies. In this research, conditions were set for calcium ion-bearing water and sulfate ion and carbonate-rich water in porous media, considering the mixing effect of the two waters. Their research results pointed out the effectiveness of flow velocity on aggregation and precipitation of calcium carbonate precipitates with respect to mineral calcium sulfate sediments.

Bedrikawstki et al. [3] presented a numerical-experimental modelling of precipitation of mineral sediments containing barium sulfate compounds in some oilfields. They investigated the effect of porous medium in kinetics of the precipitation of barium sulfate-bearing mineral sediments. Their results indicated that, kinetic rate constant of barium sulfate-bearing sediments was proportional to flow velocity.

With the aim of investigating permeability ratio in porous medium considering the formation of mineral sediments in reservoir, Ahmad [4] performed an empirical and theoretical study. In his research, he focused on two ions, namely calcium and sulfate, in impure water existing in a carbonate reservoir. According to the study, increases in temperature and injection rate were associated with corresponding increase in the formation of calcium carbonate precipitates. In their research work, Fan et al. [5] evaluated kinetics of precipitation of calcium sulfate in the presence and absence of retainers. In this study, a semi-qualitative model based on pH conditions, saturation index, and temperature was developed for the prediction of kinetics of the formation of calcium sulfate.

Haghtalab et al. [6] presented a thermodynamic model to forecast the formation of sulfate precipitates, including barium sulfate, calcium sulfate, and strontium sulfate. In this model, they used eNRTL activity coefficient model.

Saturation index and the amount of mineral precipitates formed were among the outputs of this thermodynamic model. In another research, Safari et al [7] investigated solubility of strontium sulfate in an electrolyte system containing sodium, chloride, sulfate, and strontium under considered set of temperature and pressure conditions (temperature ranges from 2 to 253.5°C, and pressures from 1 to 568.51 atm). Among other research works on prediction of solubility of mineral materials in an electrolyte system, one can refer to the work by Wang et al [8].

2. The model and solution method

2-1.The Extended UNIQUAC activity coefficient equation

In this work, the model Extended UNIQUAC (as presented by Thomsen and Rasmussen [9]) was made use of to study the electrolytic system, temperature and concentration parameters of the study variables. This model is a combination of UNIQUAC model and Debye-Hückel term. With this method he required number of parameters are fewer, compared to similar models, such as Pitzr model. The UNIQUAC model was presented by Abrams and Prausnitz [10] for excess Gibbs free energy of a mixture and consists of two parts: first part is combinatorial that is related to the entropy of the system and is determined based on the size and shape of the molecules. The residual part is related to intermolecular forces involved in the enthalpy of mixing and depends on the intermolecular forces. UNIQUAC equation consists of adjustable parameters, which are expressed as follows for a liquid-solid (or liquid-vapor) equilibrium in the binary and multicomponent system [11-12]:

$$G^{\mathrm{E}} = G^{E}{}_{combinatorial} + G^{E}{}_{Residual} + G^{E}{}_{Debye-HucKel} \tag{1}$$

$$\frac{G^{E}{}_{combinatorial}}{\mathrm{RT}} = \sum x_i \, ln\left(\frac{\emptyset_i}{x_i}\right) - \frac{\mathrm{Z}}{2}\sum q_i x_i \, ln\left(\frac{\emptyset_i}{\theta_i}\right) \tag{2}$$

In the upper equation, G^E is Gibbs free energy. In eq. 2, z is coordination number and its value is equal to 10; x_i is mole fraction; φ_i is volume fraction and θ_i is the surface area fraction of ions in the liquid-solid or liquid-vapor equilibrium system which is expressed as:

$$\emptyset_{\mathrm{i}} = \frac{\mathrm{x_i r_i}}{\sum_{\mathrm{l}} \mathrm{x_l r_l}} \tag{3}$$

$$\theta_{\mathrm{i}} = \frac{\mathrm{x_i q_i}}{\sum_{\mathrm{l}} \mathrm{x_l q_l}} \tag{4}$$

where r_i and q_i are the volume and surface area parameters for each ion. In addition, for the residual term, the following equation holds:

$$\frac{G^{E}{}_{Residual}}{RT} = -\sum_i q_i x_i \, ln\left(\sum_k \theta_k \varphi_{ki}\right) \tag{5}$$

$$\varphi_{ij} = \exp\left(-\frac{u_{ij} - u_{ii}}{T}\right) \tag{6}$$

u_{ii} is energy interaction between similar ions in an equilibrium system of solid-liquid and vapor-liquid. u_{ij} is energy interaction between different ions in an equilibrium system with each other. The energy interaction is a function of temperature and is defined as:

$$u_{ij} = u_{ij}{}^{\circ} + u_{ij}{}^{t}(T - 298.15) \tag{7}$$

u_{ij}^0 and u_{ij}^t are two adjustable parameters for energy interaction between the ions in the stable system.

The Debye-Hückel contribution (to the excess Gibbs energy) of the extended UNIQUAC model is given by the expression:

$$\frac{G^{E}{}_{Debye-HucKel}}{RT} = -x_w M_w \frac{4A}{b^3}\left[\ln\left(1 + bI^{\frac{1}{2}}\right) - bI^{\frac{1}{2}} + \frac{b^2 I}{2}\right] \tag{8}$$

where M_w is the molar mass of water , x_w is the mole fraction of water, A is a Debye-Hückel parameter, b is a constant equal to 1.5 (kg mol-1)½, and I is the ionic strength.

$$A = a_0 + a_1(T-273.15) + a_2(T-273.15)^2 \tag{9}$$

$$\mathrm{I} = 0.5 \sum \mathrm{m_i Z_i}^2 \tag{10}$$

In equation , zi is the charge of ion i and mi is the molality (mol (kg H2O)-1) of ion i. Considering the relationship between excess Gibbs free energy and basic equations of thermodynamics, by differentiation of equations 1 with respect to the mole fraction of different species, the activity coefficient for each ion is derived:

$$\ln \gamma_i = \left[\frac{\partial\left(\frac{nG^E}{RT}\right)}{\partial n_i}\right] \tag{11}$$

$$ln\gamma_i = ln\gamma_i{}^{Residual} + ln\gamma_i{}^{combinatorial} + ln\gamma_i{}^{\text{Debye–Hückel}} \tag{12}$$

To normalize equation 11 for electrolyte system, considering water presence (as a solvent) and its ions of solute, we have:

$$\gamma_i \rightarrow 1 \quad as \quad x_i \rightarrow 0 \tag{13}$$

$$\gamma^*{}_i = \frac{\gamma_i}{\gamma_i{}^{\infty}} \tag{14}$$

where in Eq. 14, for y_i^{∞} (activity coefficient of ion at infinite dilution), we have:

$$\gamma_i{}^{\infty} = \lim_{x_i \rightarrow 0} \gamma_i \tag{15}$$

And finally, we will have:

$$\begin{aligned} \ln\gamma_i{}^* = {} & \ln\left(\frac{\emptyset_i}{x_i}\right) + 1 - \frac{\emptyset_i}{x_i} - \frac{z}{2}.q_i\left[\ln\left(\frac{\emptyset_i}{\theta_i}\right) + 1 - \frac{\emptyset_i}{\theta_i}\right] - ln\left(\frac{r_i}{r_w}\right) + 1 - \frac{r_i}{r_w} - \frac{z}{2}.q_i\left[\ln\left(\frac{\mathrm{r_i.q_w}}{\mathrm{r_w q_i}}\right) + 1 - \frac{\mathrm{r_i.q_w}}{\mathrm{r_w q_i}}\right] \\ & + q_\mathrm{i}\left[1 - \ln\left(\sum_\mathrm{k} \theta_\mathrm{k}\,\varphi_\mathrm{lk}\right) - \sum_\mathrm{k}\frac{\theta_\mathrm{k}\varphi_\mathrm{ik}}{\sum\theta_\mathrm{l}\varphi_\mathrm{lk}}\right] - \mathrm{q_i}[1 - \ln(\varphi_\mathrm{wi}) - \varphi_\mathrm{iw}] - Z_i{}^2\frac{AI^{1/2}}{1 + bI^{1/2}} \end{aligned} \tag{16}$$

Finally, activity coefficient for water can be expressed as:

$$\begin{aligned} \ln\gamma_w{}^* = {} & \ln\left(\frac{\emptyset_w}{x_w}\right) + 1 - \frac{\emptyset_w}{x_w} - \frac{z}{2}.q_w\left[\ln\left(\frac{\emptyset_w}{\theta_w}\right) + 1 - \frac{\emptyset_w}{\theta_w}\right] + q_\mathrm{w}\left[1 - \ln\left(\sum_\mathrm{k}\theta_\mathrm{k}\,\varphi_\mathrm{kw}\right) - \sum_\mathrm{k}\frac{\theta_\mathrm{k}\varphi_\mathrm{wk}}{\sum\theta_\mathrm{l}\varphi_\mathrm{lk}}\right] \\ & - \mathrm{q_i}[1 - \ln(\varphi_\mathrm{wi}) - \varphi_\mathrm{iw}] + Mw\,.\frac{2A}{b^3}.\left[1 + bI^{0.5} - \frac{1}{1 + bI^{0.5}} - 2\ln(1 + bI^{0.5})\right] \end{aligned} \tag{17}$$

In the present paper, the extended UNIQUAC thermodynamic to account for pressure dependence [12]:

$$lnK_{ep} = lnK_{e,P0} + \alpha(P - P_0) + \beta(P - P_0)^2 \tag{18}$$

In Figure 1, the process of optimizing ion activity coefficient of electrolyte system by the Genetic Algorithm is shown. As shown in Figure 1, the temperature and initial concentration (based on molality units) are superimposed onto the model. According to the criteria defined, if error acceptability, optimization process stops the results will be Print Results

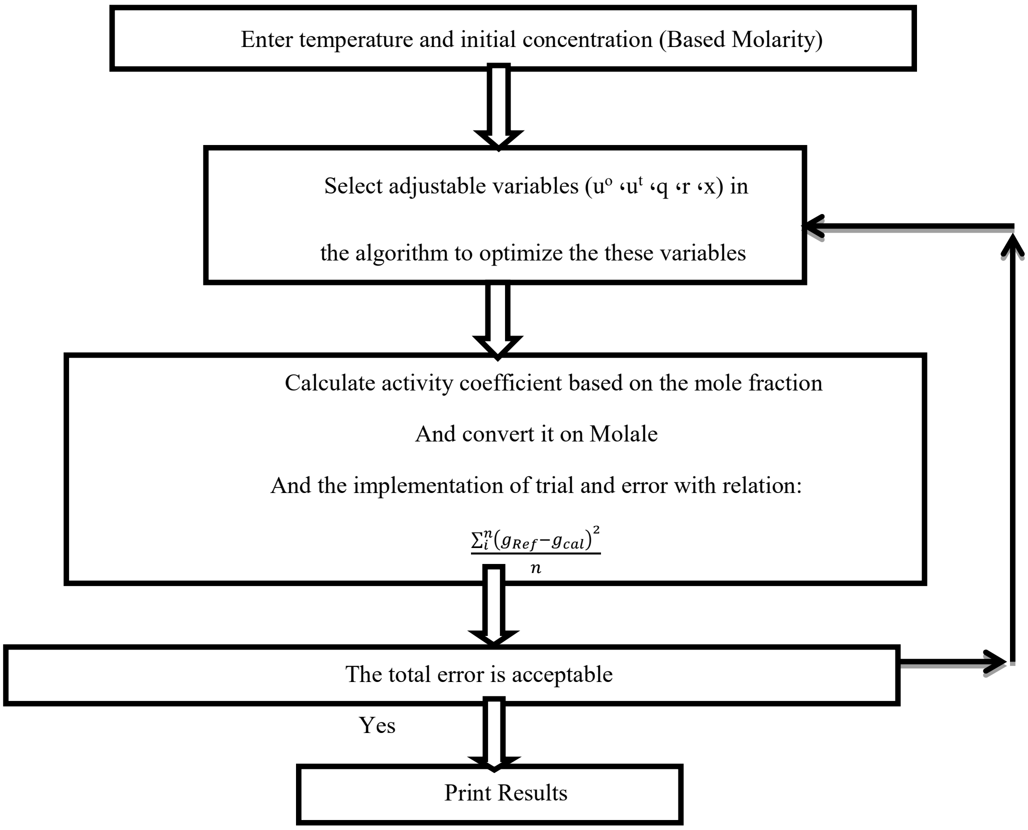


Fig. 1. The process of optimizing EUNIQUAC model based on Genetic Algorithm

In the present study, the stable constant (according to the survey predicted deposits) used the following equation [13]:

$$ln\, K_{SP}(T,P_0) = ln\, K_{SP0} - \left(\frac{\Delta H_0}{R}\right)\left(\frac{1}{T}-\frac{1}{T_0}\right) + \frac{\Delta Cp_0}{R}\,\left[\, ln\left(\frac{T}{T_0}\right) + \frac{T_0}{T} - 1\,\right] \qquad (19)$$

We have to thermodynamic equilibrium constant (at standard conditions):

$$lnK_{sp0} = -\frac{\Delta G_0}{RT_0} \qquad (20)$$

The value of the equation 19 and 20 is given in Table 1.

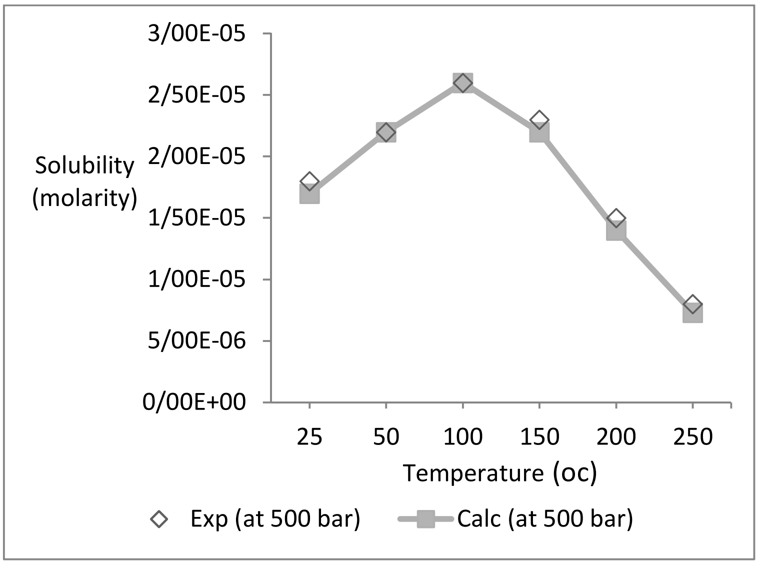
Table 1.parameter values relationships, 19 and 20 to calculate the equilibrium constant mineral deposition [13]

Mineral salt	ΔH_0 (J/mol)	ΔCp_0 (J/mol K)	ΔG_0 (J/mol)
$BaSO_4$	28158.32	-373.85	56944.24
$SrSO_4$	-5313.68	-320.79	36902.88

3. Results and discussion

In this research, the described flowchart (Fig. 1) provided very good results after running the application for $SrSO_4$+H_2O, $BaSO_4$+H_2O, $MnSO_4$+H_2O and $NiSO_4$+H_2O. Accordingly, the total error based on defined error equation for $SrSO_4$+H_2O, $BaSO_4$+H_2O, $MnSO_4$+H_2O and $NiSO_4$+H_2O are 8.09e-22, 0.0012, 0.017 and 0.011, respectively. The optimized value for parameters r, q, u^0, u^t, α and β are given in Tables 5 to 17. Figures 2 illustrates a comparison between Solubility values ($BaSO_4$) predicted by the model and the experimental data. In Table 2 a comparison between Solubility values ($SrSO_4$) predicted by model and experimental data are tabulated.

Based on Figure 2a, the solubility of barium sulfate, decreased with increasing temperature. However, with increasing pressure (from 10 to 1000 bar), the solubility of barium sulfate, associated with the increase (Fig2.b). According to Equation 10, with increasing temperature and decreasing the solubility, ionic strength decreased. With increasing pressure, ionic strength (due to the increased solubility signified an increasing trend. According to Table 2, with increasing pressure from 100 to 500 with constant temperature of 25 ° C, the solubility of strontium sulfate was associated with an increase. However, with an increase in system temperature, from 25 to 100 ° C, the solubility of strontium sulfate, decreased considerably. As a result, at the pressure of 100 bar and temperature of 100 ° C, the solubility of strontium sulfate is 0.0007, whereas at pressure of 100 bar and temperature of 100 ° C, the solubility of strontium sulfate is 0.00038. Also according to Table 2, with increasing pressure, ionic strength aqueous solution of strontium sulfate and water tended to increase; but with increasing temperature a decreasing trend appeared.



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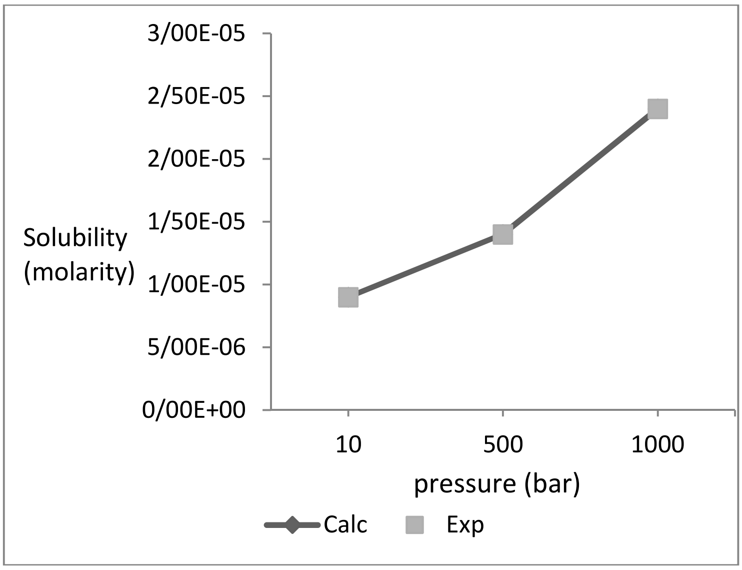

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Fig. 2. Comparison between Solubility values ($BaSO_4$) predicted by the model versus the experimental data [14]

Tables 3 and 4 illustrate a comparison between Water Activity values predicted by the model and the experimental data for $MnSO_4$ + water system and $NiSO_4$ + water system, respectively. According to Tables 3 and 4, with increasing temperature, water activity became associated with an increase. Meanwhile, by increasing the solubility, water activity decreased.

Table3.Comparison between Water Activity values predicted by model and experimental data [16-17] (MnSO4 and water system)

ionic strength	Pressure (bar) & (100 oc)	ionic strength	Pressure (bar) & (25 oc)	Calc	Exp	Pressure (bar) & (100oc)	Calc	Exp	Pressure (bar) (25 oc)
7.6E-4	1.00E+02	1.40E3	100	3.80E-4	3.80E-4	100	7.00E-04	7.00E-04	100
7.8E-4	2.00E+02	1.50E-3	200	3.90E-4	3.90E-4	200	7.50E-04	7.50E-04	200
8.2E-4	3.00E+02	1.60E-3	300	4.10E-4	4.10E-4	300	8.00E-04	8.00E-04	300
8.6E-4	4.00E+02	1.68E-3	400	4.30E-4	4.30E-4	400	8.40E-04	8.40E-04	400
8.9E-4	5.00E+02	1.90E-3	500	4.45E-4	4.45E-4	500	9.50E-05	9.50E-05	500

Table 4.Comparison between values predicted by model and experimental data [16-17] (NiSO4 and water system)

	at 383 k			at 323.15 k		ionic strength	aw at 383.15 k	Iionic strength	aw at 323.15 k
molality	Exp	Calc	molality	Exp	Calc				
3.5	0.925	0.9414	3.2	0.915	0.92	5.25	0.925	4.804	0.915
3.2	0.929	0.9438	2.8	0.925	0.9272	4.804	0.929	4.203	0.925
3	0.938	0.9456	2.4	0.94	0.9351	4.504	0.938	3.603	0.94
2.6	0.955	0.9496	2.1	0.955	0.9413	3.9032	0.955	3.152	0.955
2.4	0.962	0.95179	1.8	0.965	0.948	3.603	0.962	2.7022	0.965

Table 5. r and q parameter optimized for $SrSo_4$ and water system

Components	r	q
H2O	0.0256	20.931
Sr	16.627	7.26
SO_4	5.695	0.162

Table 6. u^o parameters for calculating UNIQUAC interaction energy parameters ($SrSO_4$ and water system)

Components	H2O	Sr	SO_4
H2O	0	0	
Sr	3575.04	-	
SO_4	735.82	674.47	-97.32

Table 7. u^t parameterfor calculating UNIQUAC interaction energy parameters ($SrSO_4$ and water system)

Components	H_2O	Sr	SO_4
H2O	0		
Sr	13.713	0	
SO_4	-28.41	34.113	11.0596

Table 8. r and q parameter optimized for $BaSo_4$ and water system

Components	r	q
H2O	10.05	12.989
Ba	10.966	8.86
SO_4	4.9155	4.097

Table 9. u^o parameters for calculating UNIQUAC interaction energy parameters ($BaSO_4$ and water system)

Components	H2O	Ba	SO_4
H2O	0		
Ba	-210.45	0	
SO_4	-992.888	-745.302	727.6

Table 10. u^t parameterfor calculating UNIQUAC interaction energy parameters ($BaSO_4$ and water system)

Components	H_2O	Ba	SO_4
H2O	0		
Ba	20.78	0	
SO_4	14.359	-16.95	10.6653

Table 11. r and q parameter optimized for $MnSo_4$ and water system

Components	r	q
H2O	12.36	9.295
Mn	13.83	6.81
SO_4	4.612	3.683

Table 12. u^o parameters for calculating UNIQUAC interaction energy parameters ($MnSO_4$ and water system)

Components	H2O	Mn	SO_4
H2O	0		
Mn	431.53	0	
SO_4	-2132.4	-739.67	-2069.114

Table 13. u^t parameterfor calculating UNIQUAC interaction energy parameters ($MnSO_4$ and water system)

Components	H_2O	Mn	SO_4
H2O	0		
Mn	47.43	0	
SO_4	33.189	-3.396	-8.312

Table 14. r and q parameter optimized for $NiSo_4$ and water system

Components	r	q
H2O	3.896	3.547
Ni	11.285	8.3264
SO_4	6.465	3.1399

Table 15. u° Parameters for calculating UNIQUAC interaction energy parameters ($MnSO_4$ and water system)

Components	H2O	Ni	SO_4
H2O	0		
Ni	-1191.11	0	
SO_4	878.45	62.64	-1408.3

Table 16. u^t parameterfor calculating UNIQUAC interaction energy parameters ($MnSO_4$ and water system)

Components	H_2O	Ni	SO_4
H2O	0		
Ni	39.8	0	
SO_4	39.16	16.53	1.29

Table 17. Optimized values of variables (A, α and β)

System	a_0	a_1	a_2	α	β
$SrSO_4$+H2O	1.3712	0.001335	0.00001164	-0.001597	-0.009817
$BaSO_4$+H2o	1.652	0.00015	0.000064	-0.00472	-0.005859
$MnSO_4$+H2O	1.258	-0.07052	-0.00605	-	-
$NiSO_4$+H2O	1.29	-0.0522	-0.00155	-	-

5. Conclusion

Mineral deposits in oil fields are among the serious costly problems anywhere in the O oil industry. Inorganic ions in the formation water from reservoir to production wellhead equipment despite conditions such as temperature, pressure, alkalinity and reducing or increasing the ionic strength of the solution, will tend to accumulate. In this study, aqueous solutions, strontium sulfate + water, water + barium sulfate, manganese sulfate+ water and nickel sulfate + water, were studied, according to EUNIQUAC model and genetic algorithms. In this research, the described flowchart provided very good results after running the application for SrSO4+H2O, BaSO4+H2O, MnSO4+H2O and NiSO4+H2O. Therefore, the total error based on defined error equation for SrSO4+H2O, BaSO4+H2O, MnSO4+H2O and NiSO4+H2O is 8.09e-22, 0.0012, 0.017 and 0.011, respectively. Based on this paper, the solubility of barium sulfate, decreased with increasing temperature. However, with increasing pressure (from 10 to 1000 bar), the solubility of barium sulfate was associated with the increase. Also by increasing temperature and decreasing the solubility, ionic strength was decreased. With increasing pressure, ionic strength (due to the increased solubility), characterized an increasing trend. Also according to this study, with increasing pressure, ionic strength aqueous solution of strontium sulfate and water increased; but with increasing temperature decreasing became visible; and with increasing temperature, water activity was associated with an increase. Also, by increasing the solubility, water activity decreased.

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