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Thermodynamic description of aqueous solutions of silver nitrate: Experimental and modeling

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ABSTRACT

The water activity of silver nitrate solutions was measured at 298.2 K and 313.2 K using a humidity sensor instrument. Concentrations of silver nitrate up to almost saturation were included. The thermodynamic properties of the system were described by the Pitzer model, the specific interaction theory (SIT), and the Extended UNIQUAC model. The interaction parameters for the models were estimated using experimental freezing points, osmotic coefficients, and solubility for silver nitrate aqueous solutions collected from the open literature, as well as the water activity data measured in this work. Pitzer and SIT parameters were applied by introducing a temperature dependency, enabling these models to cover a more comprehensive temperature range and extrapolate the calculation to higher molalities. Both approaches represent the data satisfactorily up to moderate molalities. The Extended UNIQUAC model, with its built-in temperature dependence, provides the best thermodynamic description of this binary system. It has a very satisfactory solubility diagram and a good description of the osmotic and activity coefficients.

1. Introduction

The silver nitrate + water system is very relevant from both theoretical and industrial points of view. Experimental data on freezing point depression, water activity, osmotic coefficients, and solubility give an excellent opportunity to explore and test the capability of thermodynamic models to describe the behavior of binary systems in the whole composition range and cover different thermodynamic properties. This task is very demanding, and developing a comprehensive and accurate model that can be applied at higher concentrations for different properties is often very difficult. Long-range interactions dominate at high dilution, but short-range interactions play an important role when increasing the salt concentration. To describe these systems adequately, besides a Debye-Hückel-based term, short-range interactions must be accounted for. Many models in the open literature are available [1–8] aiming to model electrolyte solutions. Still, the majority fail in terms of predictions beyond the temperature and concentration ranges of the experimental data used for determining the model parameters, or the obtained parameters lack physical meaning. Industrial applications of aqueous silver nitrate span from the pigment industry [9] to pharmaceuticals [10], and hydrometallurgy [11]. Silver nitrate is, in fact, the least expensive salt of silver and the precursor for many other salts, offering several other advantages, such as being more stable and less hygroscopic.

For the thermodynamic description of very soluble electrolytes, several authors have extended the original form of the equations by adding higher-order terms with ionic strength dependence. That is the case with the Pitzer formalism, one of the most frequently used and successful models for electrolyte solutions with well-established parameters for many electrolytes. The model is well known for its ability to correlate properties of strong electrolyte solutions and describe different solution properties and phase equilibria, such as osmotic coefficients, activity coefficients, and solubility. Robinson and Stokes [12] calculated, at 298.15 K, the activity coefficients from measured osmotic coefficients up to a molality of 6 for several salts. Hamer and Wu [13] compiled the osmotic coefficient and the available vapor pressure data to propose a recommended set of osmotic and activity coefficients up to that same molality. In 1973, Pitzer and Mayorga [14] used the Pitzer model to determine the first set of Pitzer parameters for silver nitrate at 298.15 K and for concentrations up to 6 molal. In their work on electrolytes, May et al. [15] used a composition dependency on the Pitzer parameters, estimating four parameters to represent the osmotic

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Table 1
Source, CAS, and mass purity of the salts used.

Compound	Source	CAS	Mass purity (%)
$AgNO_3$	Alfa Aesar	7761–88–8	99.0
LiCl	Agros	7447–41–8	99.5
NaBr	Alfa Aesar	7647–15–6	99.0

coefficients in the aqueous silver nitrate system at 298.15 K up to 15 molal. In 2011, the modeling work by Partanen [7], using an extended three-parameter Hückel equation for the activity coefficients, agreed with the recommended values from the previous studies [12–15].

For this important system, however, the temperature effects have never been considered, which is very important, for instance, to properly represent silver nitrate's solubility in water. Moreover, there is a need to check the ability of other essential models in electrolyte thermodynamics to describe this system. To progress in this regard, this work presents new experimental water activity data for the water + silver nitrate systems at 298.2 K and 313.2 K, almost up to the saturation, and besides Pitzer, two other approaches, SIT and Extended UNIQUAC [16] models were applied, which are very briefly presented in the following paragraph.

The SIT (Specific Interaction) model was formulated first by Ciavatta [17] in 1980 based on the earlier works by Brønsted [18], Guggenheim [19–20], and Scatchard [21–22]. This approach is widely used in geochemistry and hydrometallurgy industries for its simplicity and the possibility of evaluating activity coefficients based only on the specific interaction of ions in the solution. An extended review by Grenthe et al. [23] is available for more details about this theory. On the other hand, the Extended UNIQUAC model [24] combines a Debye-Hückel long-rang term with a short-range one based on the well-known UNIQUAC model [16]. The water/ion, anion/cation, and anion/anion interaction parameters present a linear temperature dependence in this model. It, therefore, offers the possibility to model the entire concentration and a more comprehensive temperature range, which is particularly important for salts highly soluble in water, such as silver nitrate.

The model parameters were estimated based on the available experimental data of silver nitrate-water solutions [25–37] after consistency checking and the new experimental data presented in this work. The SIT model could only accurately reproduce the system's water activity and activity coefficient in the low to medium concentration range. At the same time, solubility cannot be therefore described accurately. On the other hand, the Pitzer model can reproduce experimental data for the osmotic coefficient up to 6 molal at different temperatures with high accuracy. As the silver nitrate solubility is higher than six molal at most temperatures, the Pitzer model could not also be used to calculate solubility in that system. Finally, the Extended UNIQUAC model was the most comprehensive since it accurately describes aqueous silver nitrate solutions in the complete temperature range and from the very dilute region up to the salt saturation.

2. Experimental

2.1. Chemicals

Table 1 presents the source, CAS, and purity of the substances used as given by the supplier. To avoid water contamination, NaBr was dried at 343.15 K and LiCl at 423.15 K in a drying stove for more than 2 days and cooled in a dehydrator with silica gel before use. Silver nitrate was kept in the dehydrator with silica gel. Double distilled water, passed through a reverse osmosis system and further treated with a Milli-Q plus 185 water purification equipment, was used in all experiments, presenting a water resistivity of 18.2 MΩ.cm.

Table 2
Water activity (a_w) and osmotic coefficients (ϕ), at 298.2 K and 313.2 K and p = 0.1 MPa, for the water + $AgNO_3$ system at different molalities (m).

T = 298.2 K			T = 313.2 K		
m / mol.kg^{-1}	a_w	ϕ	m / mol.kg^{-1}	a_w	ϕ
0.499	0.986	0.782	0.500	0.983	0.951
0.999	0.974	0.731	0.998	0.973	0.760
1.499	0.965	0.659	1.500	0.962	0.716
1.997	0.955	0.639	2.001	0.953	0.667
2.000	0.956	0.624	2.498	0.944	0.640
2.497	0.946	0.617	2.998	0.937	0.602
2.501	0.947	0.604	3.000	0.937	0.601
2.994	0.941	0.563	3.499	0.930	0.575
2.999	0.940	0.572	4.006	0.923	0.555
3.496	0.933	0.550	4.492	0.917	0.535
3.995	0.928	0.519	5.000	0.912	0.511
4.497	0.922	0.501	5.499	0.907	0.492
4.986	0.916	0.488	6.000	0.901	0.482
4.991	0.917	0.481	6.495	0.896	0.469
5.497	0.911	0.470	7.000	0.891	0.457
6.009	0.907	0.450	7.498	0.886	0.448
6.500	0.902	0.440	8.101	0.881	0.434
6.985	0.896	0.436	9.016	0.872	0.421
7.495	0.892	0.423	9.999	0.862	0.412
7.948	0.888	0.414	11.020	0.854	0.397
7.997	0.886	0.420	11.992	0.845	0.389
8.987	0.879	0.398	12.998	0.837	0.379
9.967	0.870	0.387	13.972	0.827	0.377
11.013	0.862	0.374	15.008	0.817	0.373
11.999	0.855	0.362	15.779	0.811	0.368

u (m) = 0.001, u (a_w) = 0.002, u (T) = 0.15 K, u_r (p) = 0.05; u is uncertainty.

2.2. Procedure

Approximately 80 g of silver nitrate aqueous solution was first prepared by weighing (Denver Instrument, USA) into a balloon flask the appropriate masses (±0.1 mg) of salt and water for the desired molality. The resulting solution was vigorously stirred to promote salt dissolution. This solution was used to prepare samples of about 10 cm^3 of silver nitrate solutions at other molalities, by dilution with water.

The measurement of water activities (a_w) was performed using a LabMaster-a_w hygrometer (Novasina, Switzerland), already used by us in a study of the manganese nitrate-water system [38] and aqueous solutions of ionic liquids [39]. Approximately 2–3 cm^3 samples were charged in the measuring dishes and placed in the air-tight equilibrium chamber for each measurement. The exchange of free water occurs until the partial pressure of water vapor reaches equilibrium, confirmed following the a_w variation with time. When a constant value is reached, which for these solutions usually requires one hour, the water activity is recorded. The uncertainty of the measurements is better than 0.002 a_w, enabling measurements under controlled chamber temperature conditions (±0.15 K).

The humidity sensor must be initially calibrated using the reusable humidity standards, with water activities from 0.113 to 0.970, usually included in the equipment. This calibration was tested, measuring the water activities in sodium or potassium chloride aqueous solutions, which are very well known, concluding that it was insufficient for accurate water activity measurements. Instead, a calibration curve for the new measurements with silver nitrate solutions was found each day to ensure high accuracy. To do so, at least four NaBr or LiCl aqueous solutions were prepared at distinct molalities (water activity in the range 0.800 and 0.990), chosen based on the expected values for the water activity to be measured in the aqueous silver nitrate solutions, but noting that each day the range can be narrower depending on the expected values of a_w in silver nitrate solutions in that same day. These measured a_w values for the NaBr and LiCl aqueous solutions at the working temperature (298.2 or 313.2 K) are then compared to those published for LiCl aqueous solutions by Hamer and Wu [14] and for NaBr aqueous solutions by Archer [40], at the same NaBr or LiCl

Table 3
Experimental data collected from the open literature (data retrieved from the original papers).

Temperature (K)	Molality	Data points	Reference
Osmotic coefficient			
298.15	0.1–13.5	33	Robinson and Tait [25]
Vapor pressure			
303.15–363.15	0.65–33.53	49	Campbell et al. [35]
293.15–298.15	1–15	28	Kangro et al. [26]
Freezing point depression and solubility			
266.84–272.65	Sat.	14	Raoult [28]
272.33–272.65	Sat.	3	Arrhenius [27]
383.15–471.15	Sat.	10	Benrath et al. [44]
272.70–273.11	Sat.	28	Roth-Greifswald [29]
267.55	Sat.	1	Rüdorff [32]
265.85–303.15	Sat.	2	Schreinemakers and de Baat [31]
267.15–273.35	Sat.	8	Guthrie [30]
265.58–468.05	Sat.	37	Campbell and Boyd [36]
283.15–308.15	Sat.	5	Bailey [34]
273.15–373.15	Sat.	7	Kazantsev [43]
266.15–453.15	Sat.	24	Etard [33]
Activity coefficient			
298.15	0.0002–0.01	6	Macinnes and Brown [37]

molalities. A calibration (straight) line relating the actual water activity [14,20] to the value measured in the equipment is then built. On each day of analysis, water activity measurements of the silver nitrate solutions and the standard solutions used for the external calibration curve are performed in an alternating sequence.

2.3. Experimental results

The experimental water activity, a_w, data measured in this work for the water + $AgNO_3$ system, at 298.2 and 313.2 K, and the corresponding osmotic coefficients (ϕ), are presented in Table 2.

3. Modeling

3.1. Compilation of experimental data

Before any modeling work, the first task was to develop an extensive literature review of the available experimental data to make the appropriate decisions concerning the modeling strategy. This evaluation concerns the nature of the data, accuracy, temperature, and concentration ranges. It is also essential to consider the final purpose of the model. The model could be focused on the water activity of the aqueous solution at lower concentration, the thermal properties and/or the solid-liquid phase diagram, for instance.

Concerning the osmotic coefficient data from the literature, the first data set was published by Robinson and Tait [25] at 298.15 K. Later, Kangro et al. [26] published vapor pressure measurements at 293.15 K and 298.15 K. Studies from Arrhenius [27], Raoult [28], Roth-Greifswald [29], and Guthrie [30] focused on the ice curve of silver nitrate. A comprehensive work on the properties of aqueous silver nitrate solutions was published by Campbell et al. [35–36]. In their work, the authors present vapor pressure data [35] and solubility data [36], from the ice curve to the fusion of the salt, combining several experimental techniques. As shown in Table 3, Etard [33], Bailey [34], Schreinemakers and de Baat [31], Rüdorff [32], and Kazanstev [43] measured the solubility in the water-silver nitrate system. Macinnes and Brown [37] reported the activity coefficient of silver nitrate solutions in water at low concentrations measured by the electromotive force method. Table 3 summarizes the experimental data collected.

3.2. Models and parameter estimation

In a previous work [38], we presented the features and the detailed description of the Pitzer and the SIT models. A detailed description of the extended UNIQUAC model can be found in the work by Thomsen et al. [24,42]. Therefore, only the main significant information concerning estimating the model parameters is presented here. The evaluation of the data used in the modeling was based on the agreement between each data set and the deviation of the experimental points from the general trend. Selection criteria include experimental accuracy, consistency, reproducibility, and coverage of various ionic strengths and temperatures.

After the data evaluation, the Pitzer and SIT parameters were estimated based on the freezing point [27–31], the osmotic coefficient data from Robinson and Tait [25], and the osmotic coefficient obtained from the water activities measured in this study. Data above the molality limit of Pitzer and SIT models (6 molal) were not used. The remaining data were left for comparison purposes and to test the extrapolation capacities of the Pitzer and the SIT models and the ability to describe the solid-liquid phase diagram.

The objective function (*OF*) to be minimized is the squared difference between each experimental osmotic coefficient (ϕ_{Exp}) and that calculated (ϕ_{Model}) by the model, according to:

$$OF = \sum_i \left(\phi_{Model.i} - \phi_{Exp.i}\right)^2 \quad (1)$$

The osmotic coefficient is calculated by:

$$\phi = -\frac{lna_w}{\nu m M} \quad (2)$$

where ν is the sum of the stoichiometric coefficients (2 for 1:1 electrolytes), m is the molality, and M is the water molar mass in $kg.mol^{-1}$.

Eq. (1) does not include any weighting factors. However, some data points presenting random large deviations from the general trend were neglected in the parameter estimation. The freezing point data were converted to osmotic coefficients to ensure the representation of the system properties at low temperatures with good flexibility. To find the best set of parameters a series of tests, based on the temperature dependency expression of the type $p = a + b/T$ was made for all the parameters to be estimated. These are $\beta^{(0)}\beta^{(1)}$, and C^{ϕ} in the Pitzer model and $\varepsilon_{0_\gamma}(M.X)$ and $\varepsilon_{1_\gamma}(M.X)$in the SIT model.

The criterion of choice for the selected set of parameters was the value of the objective function combined with the capacity to represent the osmotic coefficient, freezing point, and the activity coefficient. A set with the lowest value of the objective function is not automatically the best set if the representation of the properties is not acceptable or the extrapolation capacity of the parameters is not good. The set of Pitzer (three) and SIT (four) parameters is presented in Table 4.

The modeling strategy used for the Pitzer and SIT models differs from that used for the Extended UNIQUAC model due to the nature of the models. Pitzer and SIT models' interaction parameters are salt-specific, while the Extended UNIQUAC parameters are ion-specific. The major limitation of the Pitzer and the SIT models is the concentration range

Table 4
Estimated Pitzer and SIT parameters (in the form $a + b/T$) for aqueous solutions of silver nitrate (temperature range from 260 K to 323 K, up to 6 molal).

$\beta^{(0)}$		$\beta^{(1)}$		$C^{(\varphi)}$		$\varepsilon_{0_\gamma}(M.X)$		$\varepsilon_{1_\gamma}(M.X)$	
a	b / K	a	b / K	a	b / K	a	b / K	a	b / K
0	-24.507	0	-62.507	0	1.3867	0.664	-268.641	-0.073	25.554

Table 5
UNIQUAC interaction parameters for describing the $Ag(NO)_3$ – H_2O system. The H_2O – NO_3^- and the NO_3^- – NO_3^- parameters are from Thomsen et al. [24] (applicable for the whole temperature and concentration ranges).

		$u_{0(K)}$	u_t^a
H_2O	NO_3^-	998.9202 [24]	9.3251 [24]
Ag^+	NO_3^-	952.6669	7.8833
Ag^+	H_2O	290.8728	-1.5949
NO_3^-	NO_3^-	2753.714 [24]	2.2866 [24]

limit imposed by the thermodynamic framework, and they do not explicitly account for the temperature influence on the properties. All the data in Table 3 were used to estimate the Extended UNIQUAC parameters. This includes the experimental solubility data.

An advantage of using the Extended UNIQUAC model is its capacity to model the system in the entire concentration range without any limitation regarding the model formalism. Another advantage is the ion-specific parameters, which allow the same parameters to be used in many other systems involving Ag^+ and NO_3^-. In the current work, the parameters for the nitrate ion were taken from earlier work [24], and only four parameters between $Ag^+$and water, or NO_3, have been estimated.

To include the solubility data in the parameter estimation, the solid-liquid equilibrium criterion based on the equality of the chemical potential of the solid and the sum of the chemical potentials of its constituents in water has been considered [24]. The chemical potential of an aqueous component is the sum of a standard state chemical potential and a term that describes its concentration dependence, being calculated using a thermodynamic model like the Extended UNIQUAC model. The standard state chemical potentials for the silver and nitrate ions and solid silver nitrate reported by NIST [41] were used in this work.

The estimation of the extended UNIQUAC model parameters was carried out using a least-squares minimization method using ESTIM software which was developed at the Technical University of Denmark; Thomsen [42] has presented this software, and the methodology followed in this work for modeling physical properties of several electrolyte systems. The objective function and fitting procedure used in this study are identical to the one used by Thomsen et al. [24]. The UNIQUAC volume and surface area parameters of ions and water can be found in Thomsen et al. [24], while the four UNIQUAC interaction parameters estimated are presented in Table 5.

4. Results and discussion

In Fig. 1, the calculated, experimental, and recommended values by Hamer and Wu [13] for the osmotic coefficients at 298.15 K are plotted for comparison. Using the estimated Pitzer parameters it is possible to calculate the osmotic coefficient of silver nitrate solutions with good accuracy up to 6 molal. The SIT model produces values higher than the experimental data at intermediate silver nitrate molalities and generally lower at higher molalities.

The results obtained with both models show that care must be taken when extrapolating the value of thermodynamic properties in electrolyte systems. The excellent agreement between the available experimental data lead to confirm the higher flexibility of the Pitzer model to express the temperature than the SIT model. The Extended UNIQUAC representation of the osmotic coefficient is in excellent agreement with the available experimental data and our measured values. The root mean square errors, considering the number of estimated parameters in each model, were calculated for the osmotic coefficients, showing to be 4.9 %, 2.8 % and 1.6 % for the SIT, Piter and Extended UNIQUAC models, respectively.

$$RMSE\ (\%) = \sqrt{\frac{\sum_{i=1}^{n}\left(\phi_{Model,i} - \phi_{Exp,i}\right)^2}{n-k}} * 100 \qquad (3)$$

where i is the specific point among n experimental data points, k is the number of estimated parameters, and Model and Exp, respectively, are calculated by the model and experimental.

The discrepancy observed in Fig. 1 for the representation of the osmotic coefficient at 298.15 K, for Pitzer and SIT models, is connected to the selected strategy to cover a broader temperature range, from freezing point data up to 323.15 K. Naturally, in this most general approach, the fit at 298.15 K is not comparable to the expected accuracy of these models when estimating the parameters at a temperature only.

In Fig. 2, the calculated molal mean ionic activity coefficients of silver nitrate at 298.15 K are plotted together with those published by Hamer and Wu [13]. The Pitzer model could represent the activity

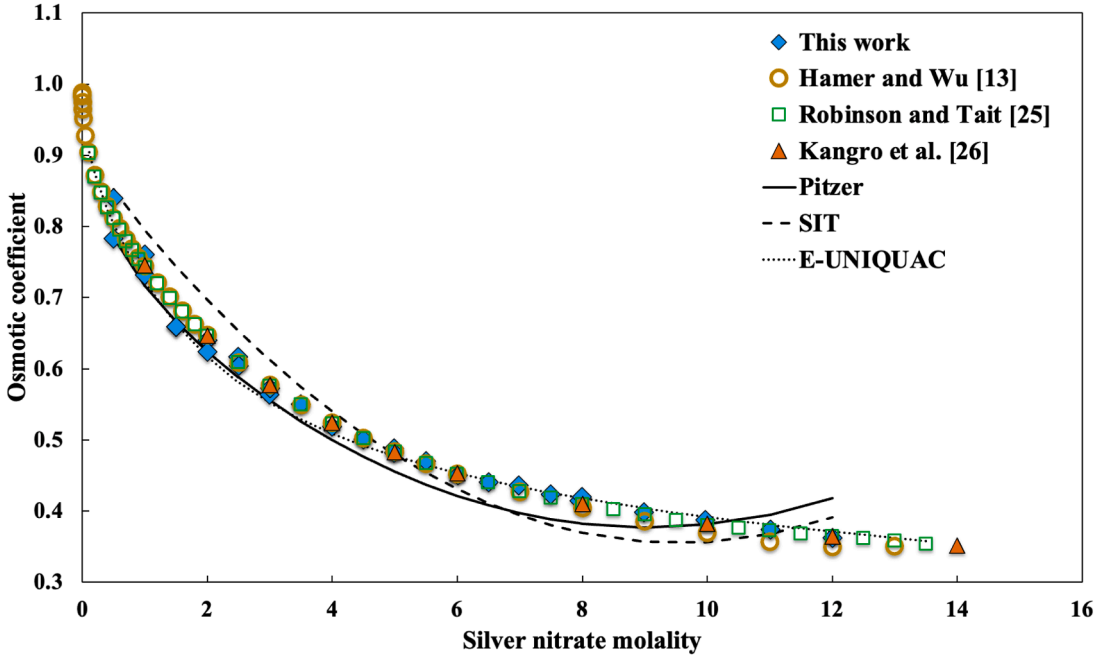

Fig. 1. Osmotic coefficient of silver nitrate aqueous solutions at 298.2 K: Experimental data and the calculated by the Pitzer, SIT and Extended UNIQUAC models.

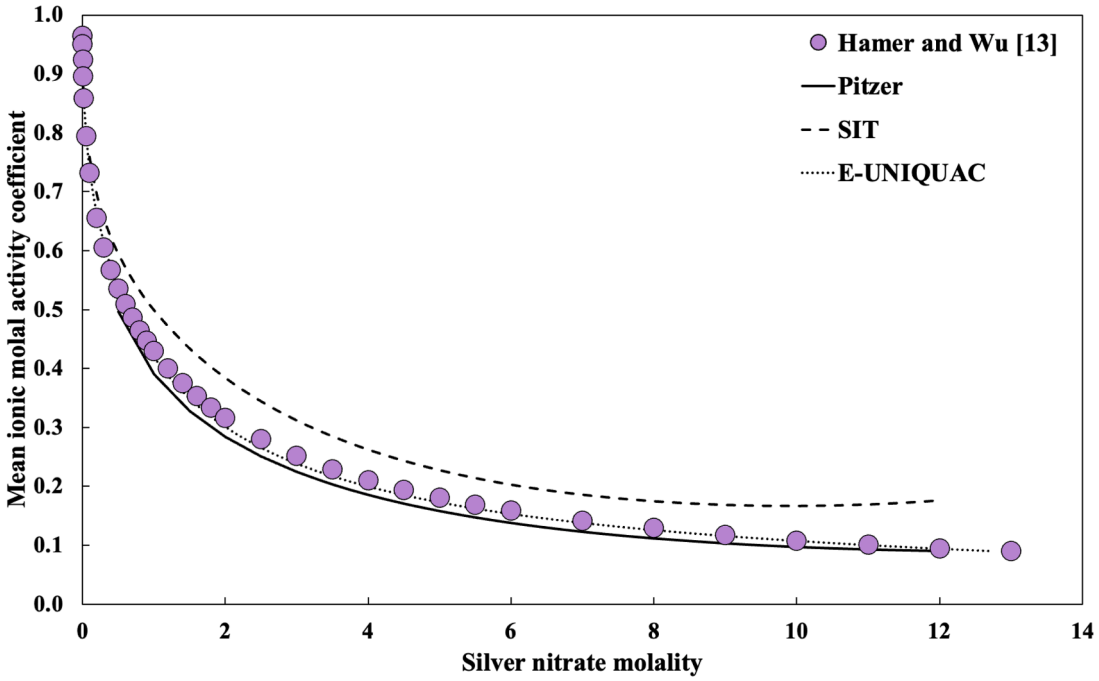

Fig. 2. The mean ionic molal activity coefficient of silver nitrate in aqueous solutions at 298.15 K; the recommended values by Hamer and Wu [13] plotted together with values calculated by Pitzer, SIT and Extended UNIQUAC models.

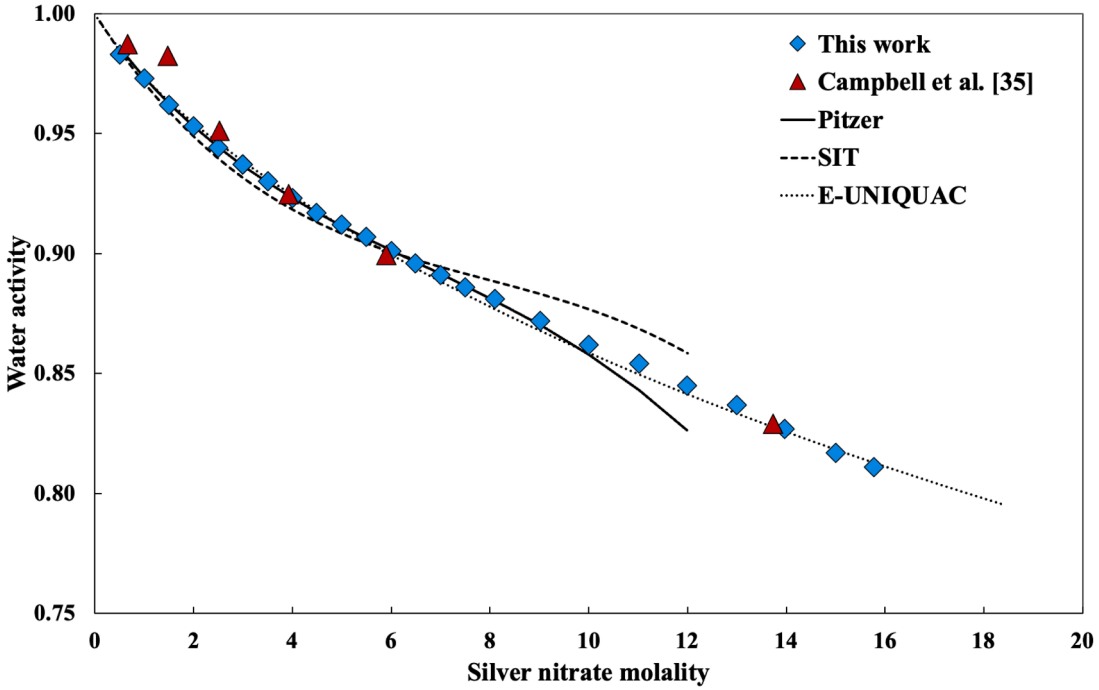

Fig. 3. Experimental water activities of silver nitrate solutions at 313.2 K plotted together with values calculated with the Pitzer, SIT, and Extended UNIQUAC models.

coefficient at 298.15 K with good accuracy, but a small deviation was noticed with the SIT model. This confirms our earlier statement about the ability of the temperature-dependent parameters of this model to represent the data. The mean ionic molal activity coefficients calculated by the Extended UNIQUAC model are in good agreement with the experimental data.

Since new experimental data at 313.2 K is presented in this work, we have tested the prediction capacity of the Pitzer and SIT models over six molal [15,17], which is the limit of the use of the framework of these molality-based models. As shown in Fig. 3, the Pitzer model could represent the water activity until ten molal $AgNO_3$. A progressive deviation appears at higher concentrations. The SIT model was able to reproduce the experimental data, but a deviation was noticed starting from 6 molal. The Extended UNIQUAC model can reproduce data in the entire concentration range.

As shown in Fig. 4, the Extended UNIQUAC model provides good

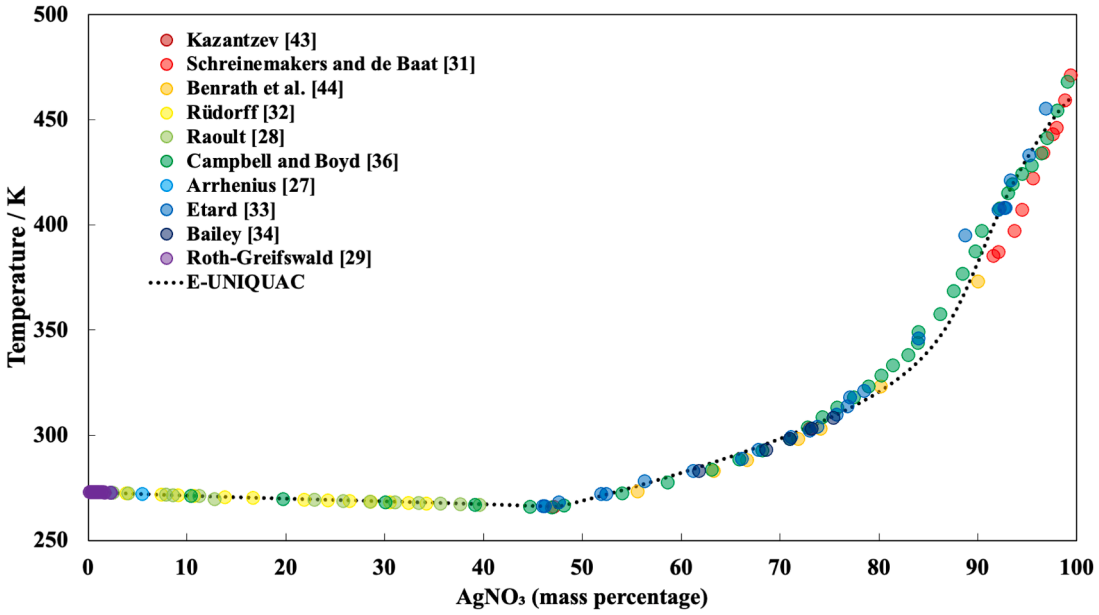

Fig. 4. Solubility of silver nitrate in water calculated with the Extended UNIQUAC model plotted together with the available experimental data.

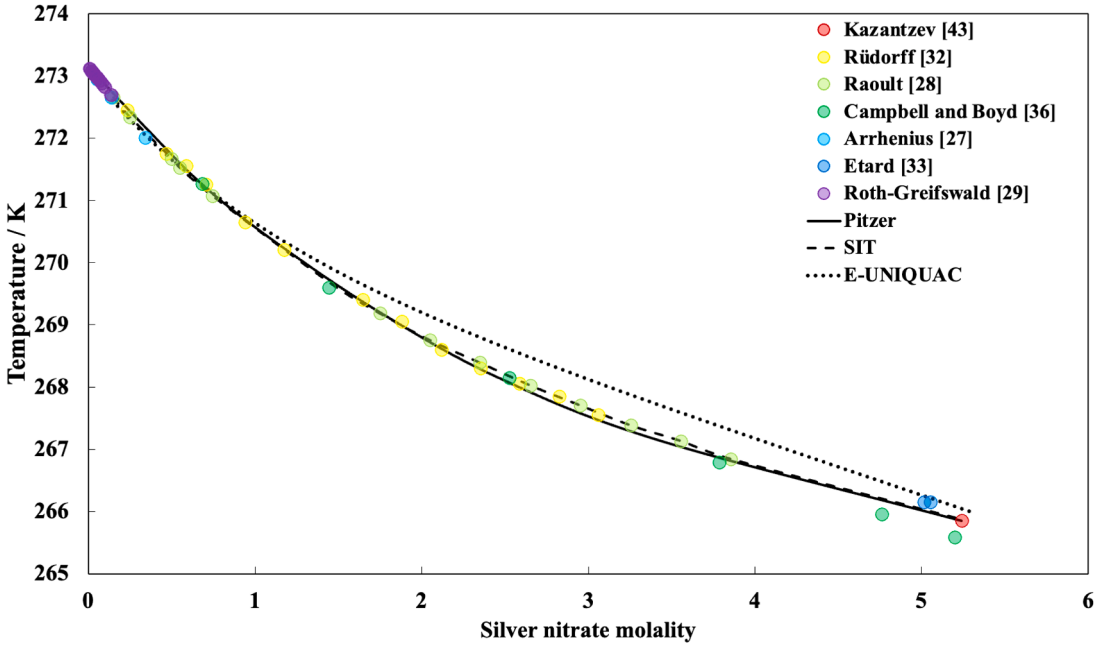

Fig. 5. The freezing point branch of the solubility curve calculated with the Pitzer, SIT and Extended UNIQUAC models.

results in modeling solid-liquid equilibrium in the silver nitrate-water system in the entire temperature and concentration range. The Pitzer and SIT models could not be applied to describe the solubility, not only due to the difficulties related to the temperature extrapolations (over 471 K) but also because the molality can attain very high values (several hundreds). However, The Pitzer and the SIT models could be used for solid-liquid equilibrium representation of the freezing point branch, where the concentration is below six molal, and the temperatures within the values used for the parameter estimation. Fig. 5 shows a very good representation by the Pitzer and SIT models, while E-UNIQUAC presents a slightly higher freezing temperature than the experimental data at higher molalities. However, the much larger temperature and the whole composition range achieved using this model, using only four interaction parameters, completely supports its use for describing such type of electrolyte systems.

As the global analysis of the modelling results, one can ascertain that the prediction capacity of Pitzer and SIT models is limited due to the modelling approach framework, applying temperature dependency with a reduced number of parameters (three for Pitzer and four for SIT), turning the model simpler, it does not allow a rigorous representation of all the properties in the whole temperature range, making it more challenging to extrapolate over the molality range (six molal) usually applied for these models and 1:1 electrolytes. On the other hand, the inclusion of a temperature dependence introduces, as expected, some discrepancies at 298.15 K, compared with the direct modelling of parameters using experimental data only at 298.15 K. The modelling of this work aims to show that based on a reduced number of model parameters, and even with concentration limitations, the models could

represent the osmotic coefficient, freezing point, vapour pressure and activity coefficient with satisfactory accuracy. For the E-UNIQUAC, an additional objective was also to develop a model that can represent the solubility diagram in the whole composition range to have a comprehensive view of the water-silver nitrate system, which was well succeeded. Summing up, the final accuracy of the models is based on a delicate balance between the diversity of properties it represents, temperature and concentration ranges, and the number of model parameters.

5. Conclusions

Water activity data of silver nitrate solutions were measured in this work at 298.2 and 313.2 K. The available experimental data (water activity, osmotic and mean ionic molal activity coefficients, and solubility) from the literature were checked for consistency. The Pitzer and SIT models can satisfactorily represent the water activities and the osmotic and mean activity coefficients in the aqueous nitrate solutions up to the six molal concentrations. The Pitzer model is more accurate, while the SIT model generally overestimates those properties at lower molalities. Extrapolations for higher molalities are not precise enough using these approaches and could not be used to describe the solubility of silver nitrate in water. On the other hand, the Extended UNIQUAC model can represent the whole solubility diagram, being only less precise than the other two models in the representation of the freezing point depression.

CRediT authorship contribution statement

Mouad Arrad: Writing – original draft, Software, Investigation. **Mehriban Aliyeva:** Writing – original draft, Methodology, Investigation. **Mónia A.R. Martins:** Writing – original draft, Methodology, Investigation. **Kaj Thomsen:** Writing – review & editing, Validation, Supervision, Data curation, Conceptualization. **Simão P. Pinho:** Writing – review & editing, Validation, Supervision, Resources, Methodology, Funding acquisition, Data curation, Conceptualization.

Declaration of competing interest

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Simao Pinho reports financial support was provided by Foundation for Science and Technology. If there are other authors, they declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Data availability

Data will be made available on request.

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