Journal of chemical & engineering data

pubs.acs.org/jced

Article

Solubility and Activity Coefficients of Three Triazine-Type Compounds in Various Low Ionic Strength Aqueous Solutions

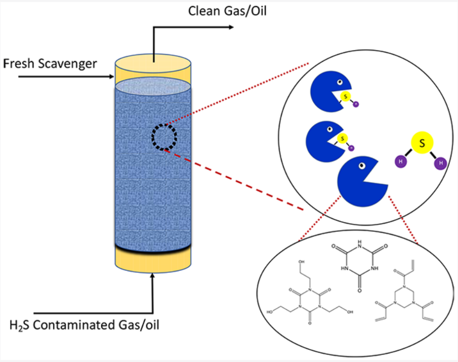
Linh Doan and Tracy J. Benson*

Cite This: *J. Chem. Eng. Data* 2020, 65, 2325−2331

Read Online

ACCESS | Metrics & More | Article Recommendations

ABSTRACT: In this work, three triazine-type compounds were studied for their solubilities in aqueous and saline environments. Using potentiometric titration and gravimetric methods, the solubility of cyanuric acid (CA), 1,3,5-triacryloylhexahydro-1,3,5-triazine (TR), and 1,3,5-tris(2-hydroxyethyl) cyanuric acid (IC) were studied in aqueous NaCl solutions (0 to 0.226 mol·kg^{-1}). From the solubility results, Setschenow coefficients, octanol/water partition coefficients, activity coefficients, and infinite dilution activity coefficients were determined. The trend of the solubility in pure water was IC ≫ TR > CA. The activity coefficient for CA increased continuously throughout the studied salt range. TR and IC, on the other hand, revealed a salting-in and salting-out effect based on the salt composition.



1. INTRODUCTION

Sour crudes, caused by high concentrations of hydrogen sulfide, can cause harm to the environment and to humans.[1−3] Hence, the removal of H_2S from oil and gas streams has been a priority, oftentimes a necessity, for upstream crude oil/gas facilities for decades. However, advances in production, namely horizontal drilling and hydraulic fracking, have produced oils and natural gases that contain higher concentrations of H_2S than previously seen.[1] These sour crudes must undergo separation and/or conversion processes to lower the H_2S content prior to entering the midstream pipelines and railcars. H_2S scavengers can be categorized into two types: the regenerative method (the absorption of H_2S is reversible, and the typical absorbent is often an alkanolamine due to the presence of hydroxyl and amino groups, which are very suitable for acid-gas scrubbing) and the nonregenerative method (where the absorption process is irreversible, and the scavengers can be oxidizing chemicals (i.e., calcium ferrite), metal carboxylates, chelates, aldehydes, ethers, and amines such as triazines).[1,4] Hence, one of the most widely used processes is the use of active scavengers that react with H_2S, thereby incorporating the sulfur into a hydrocarbon molecule. Triazine-based scavengers have become the most widely used of the reactive-type scavengers.[1] 1,3,5-Hexahydrotriazine is the most frequently used nonregenerative scavenger; however, its byproducts are deleterious (i.e., fouling) to downstream equipment (i.e., overhead condensers), which can make the H_2S extraction process expensive.[1,5]

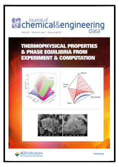
Due to the downstream fouling issues, molecular adaptations to the basic triazine molecule have been adopted in hopes that those byproducts would be benign to equipment fouling and corrosion. Varying the side chains has the impact of making the triazine scavenger more (or less) soluble in aqueous (or nonpolar crudes). In addition, these side chains could be tailored to be more active for sour gas, rather than sour liquid crude, streams. Side-chain modifications lead to uncertainty of the solubility, liquid/gas interactions, and possible byproduct complications. Because currently melamine has been studied extensively,[6−13] this work focuses on developing thermodynamic parameters (i.e., solubility, scavenger/ion interactions, etc.) and developing activity coefficient models for triazines containing ethanolic, amino, and acrolein substituents such as cyanuric acid (CA), 1,3,5-triacryloylhexahydro-1,3,5-triazine (TR), and tris(2-hydroxyethyl)isocyanuric acid (IC) (Figure 1), which are three out of 14 most commonly used nonregenerative H_2S scavengers.[1]

2. THEORETICAL CONSIDERATIONS

The theory of determining solubility has been shown in previous papers.[14,15] Proton association or dissociation can be seen in eq 1, where the H_2S scavenger is represented as A.

$$A^0 + H^+ = HA^+ \qquad (1)$$

Received: September 19, 2019
Accepted: April 16, 2020
Published: April 28, 2020



ACS Publications

https://dx.doi.org/10.1021/acs.jced.9b00898

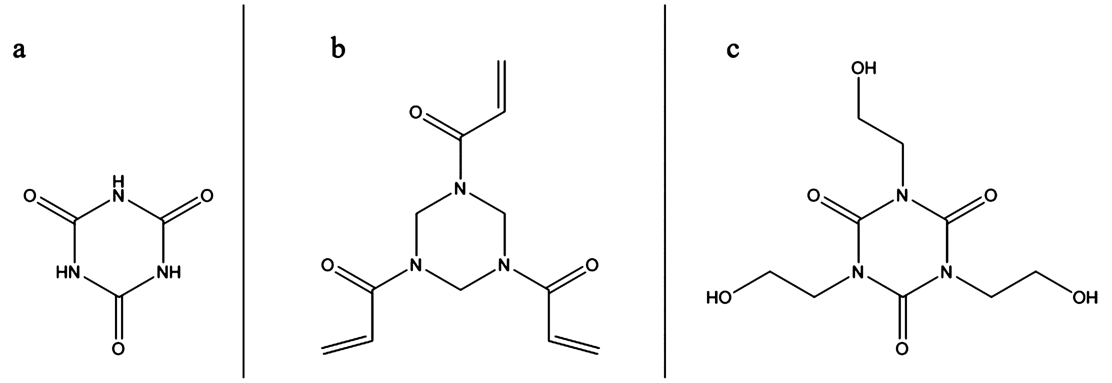


Figure 1. Scheme of (a) cyanuric acid, (b) 1,3,5-triacryloylhexahydro-1,3,5-triazine, and (c) tris(2-hydroxyethyl)isocyanuric acid.

Table 1. Sample Table

chemical name	CAS#	source	initial mass fraction purity	purification method	final purity
CA[a]	108-80-5	Sigma-Aldrich	0.99	none	
TR[b]	959-52-4	Sigma-Aldrich	0.98	none	
IC[c]	839-90-7	Sigma-Aldrich	0.98	none	
sodium chloride	7647-14-5	Sigma-Aldrich	0.99	none	
hydrochloric acid	7647-01-0	Sigma-Aldrich	0.37	none	
potassium hydroxide	1310-58-3	Sigma-Aldrich	0.85	none	
water		in-house		[d]	18 MΩ

[a] CA = 1,3,5-triazine-2,4,6-triol (or cyanuric acid). [b] TR = 1,3,5-triacryloylhexahydro-1,3,5-triazine [c] IC = tris(2-hydroxyethyl)isocyanuric acid. [d] Distillation and deionization system tested by in-line conductivity meter.

The total solubility of scavengers can be described as

$$S^{T} = [A^{0}] + [HA^{+}] \tag{2}$$

where S^T is the total solubility and A^0 and HA^+ are the neutral and protonated species of scavengers, respectively. The literature[3,9,10] has shown that the solubility of neutral species depends on the salt concentration (for a 1:1 supporting electrolyte, $I \approx c_{MX}$ where c_{MX} is the NaCl concentration on a molar scale), which leads to the following Setschenow equations:[16−18]

$$\log \gamma = \log \frac{S_0^0}{S_0} = k_c c_{MX} \tag{3}$$

$$\log y = \log \frac{S_0^0}{S_0} = k_m m_{MX} \tag{4}$$

where γ and y are the activity coefficients expressed in molar (c) and molal (m) concentration scales, respectively. S_0^0 and S_0 are the solubilities of scavengers in DI water and NaCl solutions at different salt concentrations, respectively. The Setschenow coefficients are presented as k_c (molar concentration) and k_m (molal concentration). Eqs 3 and 4 are only valid when the activity coefficients are equal to unity and when $S^T < 0.05$ mol·L^{-1}. If the solubility is higher, the ligand self-interaction must be considered. This study, however, does not require this correction. The activity coefficient is used to quantitate the deviations from ideal behavior, which can be calculated by Raoult's law in a mixture of chemical substances. Hence, if the activity coefficient is less than 1.0 (i.e., negative deviation from Raoult's law), the molecules have a strong attractive force, and more energy is required to separate them. This also means that the actual vapor pressure of the mixture is less than that calculated by Raoult's law. On the other hand, when the activity coefficient is greater than 1.0 (i.e., positive deviation from Raoult's law), the molecules have a strong repelling force, and less energy would be required for separation. When the activity coefficient equals 1.0, the behavior of the chemical substance in the mixture is considered ideal.

3. EXPERIMENTAL SECTION

Information regarding chemicals used in this study are presented in Table 1. Potassium hydroxide and hydrochloric acid solutions were prepared from concentrated purchased solutions. An Orion 950 titrator was used to perform the potentiometric titrations. Initial experiments were performed using melamine (in similar salt concentration ranges), a well-studied system to validate the technique and instrument performance. Initially, the gravimetric method was used to determine the solubility of scavengers in water without salt (i.e., 18 MΩ DI water). The solubilities of TR and CA in saltwater were determined using the potentiometric method. For IC, the initial solubility measurements (i.e., without salt) yielded relatively large values.

To calibrate the titrator, a known mass of material was dissolved in a known concentration of salt solution. Then, 0.1 M HCl or KOH was used as the titrant. The titrator's parameter for molecular weight of the sample was set at 129.07. Calibration of the titrator was performed by adjusting the reaction ratio and the titrant until the instrument yielded the known concentration of cyanuric acid in the salt solution. The same trial and error method was used for all three absorbents. The same reaction ratio and titrant were then used when analyzing the absorbent concentrations in the various salt concentrations. The solubility of the known concentration of melamine (M), CA, and TR was used to determine the parameters of the potentiometric titrator. As shown in Table 2, the instrument provided solubilities similar to those of the known concentrations.

Table 2. Quality Assurance/Quality Control for Titrator

			solubility[a] ($g\ kg^{-1}$)	
analyte	V_{water} (mL)	mass of NaCl (g)	known conc.	avg. measured (titrator)
M[b]	700	5.86	1.033	1.214 ± 0.034
CA	580	10.03	1.033	0.987 ± 0.024
TR	500	3.79	1.105	1.136 ± 0.264

[a] Solubilities are given as g of solute per kg of water. [b] Melamine.

When the solubility of IC (with salt) was measured using the potentiometric method, the concentrations were outside the detection limits of the instrument. Therefore, the solubility of IC in saltwater was determined gravimetrically. The gravimetric method should not be applied for TR and CA because the solid contains NaCl and scavengers. Hence, the potentiometric titration method gives more accurate results when considering the low concentration values. However, the mass of IC when saturated in salt solution is much greater than the mass of NaCl in the solution. Therefore, the mass of NaCl becomes negligible. To decrease the error associated with the mass of the salt in the solution, the mass was subtracted according to the concentration of the salt in the original sample. All experiments were performed in triplicate, and the error bars indicate one standard deviation.

4. RESULTS AND DISCUSSION

4.1. Development of Setschenow Coefficients and Activity Coefficients. As shown in Table 3, the solubility of CA in water decreased with increased salt concentrations. With no salt, the measured solubility was 2.12 ± 0.063 $g{\cdot}L^{-1}$, which is in good agreement with previously published results.[19,20] The solubility decreased with increased salt concentrations in the range of 0.073 to 0.226 $mol{\cdot}kg^{-1}$ of salt. The salt concentration range and increments were determined at a lower range (<0.3 $mol{\cdot}kg^{-1}$ of salt) since other literature sources[6,17,21−23] provided solubility data at a very high salt concentration. A graph of log γ versus [NaCl] (Figure 2) was shown to be nonlinear; therefore, eq 5 and a least-squares analysis were used to determine the Setschenow coefficient ($k_{c,m}$).

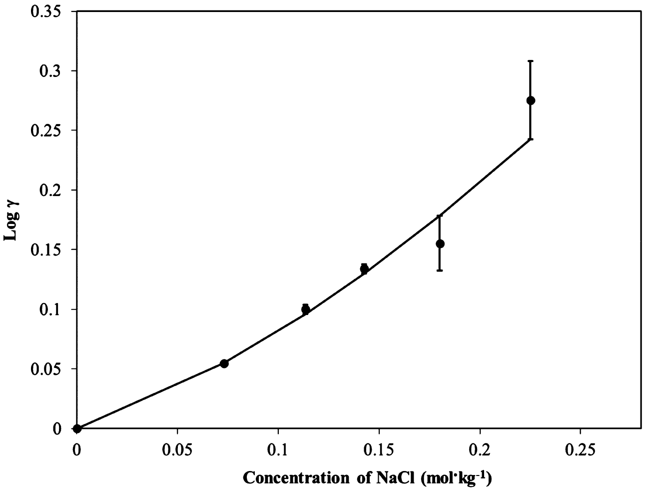


Figure 2. Log of the activity coefficient, γ, versus the salt content at T = 298.15 K for cyanuric acid. Error bars represent 1σ.

$$k_{c,m} = k_{c,m\infty} + \frac{k_{c,m0} - k_{c,m\infty}}{c,m_{MX} + 1} \tag{5}$$

where $k_{c,m\infty}$ and $k_{c,m0}$ are the relative values at $c,m_{MX} \to \infty$ and $c,m_{MX} \to 0$, respectively.

Similarly, the activity coefficient versus salt concentration graphs for TR and IC were used to determine the Setschenow coefficients (Figure 3). Both of these scavengers, however, exhibited a phenomenon known as "salting in" and "salting out", where a compound's affinity for a solvent is dependent upon the salt content. In this case, the scavengers could toggle between hydrophilic to hydrophobic. The calculated Setschenow coefficients are shown in Table 4. The positive and negative signs of the Setschenow coefficients represent the salting-out and salting-in effect, respectively.

As compared in Figures 1 and 3, the trend for the CA activity coefficients in NaCl is different than TR. However, the trend for TR is quite similar to IC. When the salt concentration is ~0.16 $mol{\cdot}kg^{-1}$, the log γ for CA and IC is ~0.15. When the salt concentration is <0.16 $mol{\cdot}kg^{-1}$, IC deviates from the ideal behavior less than CA. When the salt concentration is >0.16 $mol{\cdot}kg^{-1}$, IC deviates from ideal behavior more than CA. When the salt concentration is between 0.112 and 0.220 $mol{\cdot}kg^{-1}$, only TR deviates positively from the ideal behavior. However, for salt concentrations at 0.112 and 0.220 $mol{\cdot}kg^{-1}$, TR's activity coefficient is 1.04 and 1.02, respectively, which can be considered ideal. As the salt

Table 3. Average Experimental Solubilities (and Standard Uncertainties) of Tested Triazines in Water for Different NaCl Concentrations at Temperature T = 298.15 K and Pressure p = 0.1 MPa[a]

CA		TR		IC	
[NaCl][a] ($mol{\cdot}kg^{-1}$)	solubility[b] ($g{\cdot}kg^{-1}$)	[NaCl] ($mol{\cdot}kg^{-1}$)	solubility ($g{\cdot}kg^{-1}$)	[NaCl] ($mol{\cdot}kg^{-1}$)	solubility ($g{\cdot}kg^{-1}$)
0	2.121 ± 0.063	0	9.771 ± 0.030	0	961.5 ± 20.7
0.073	1.869 ± 0.101	0.025	7.189 ± 1.346	0.038	646.8 ± 26.0
0.114	1.685 ± 0.158	0.081	0.546 ± 0.213	0.091	555.6 ± 7.9
0.143	1.558 ± 0.032	0.112	0.888 ± 0.071	0.109	592.8 ± 22.3
0.181	1.483 ± 0.185	0.144	2.339 ± 0.352	0.149	663.8 ± 22.5
0.226	1.125 ± 0.143	0.184	2.009 ± 0.275	0.185	678.2 ± 21.0
		0.220	0.939 ± 0.205	0.222	661.8 ± 8.5

[a] Standard uncertainties u are $u(T)$ = 0.5 K and $u(m)$ = 0.0001 $mg{\cdot}kg^{-1}$. The relative standard uncertainty for pressure is $u_r(p)$ = 0.05. [b] Solubilities are given as g of solute kg^{-1} of water.

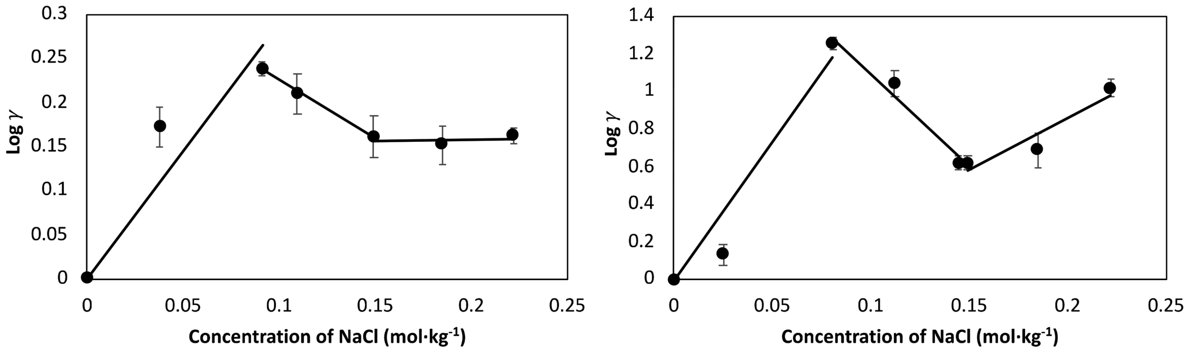


Figure 3. Log of the activity coefficient, γ, versus the salt content at T = 298.15 K for IC (left) and TR (right). Error bars represent 1σ.

Table 4. Setschenow coefficients of CA, TR, and IC

	k_c (mol·L^{-1})	k_m (mol·kg^{-1})	k_{c0}, k_{m0}	$k_{c\infty}$, $k_{m\infty}$
CA	0.86 ± 0.17	0.85 ± 0.17	0.56, 0.56	3.37, 3.35
TR	15.79 ± 1.71, −9.70 ± 4.59, 4.22 ± 0.36	15.75 ± 1.71, −9.67 ± 4.59, 4.21 ± 0.36		
IC	2.62 ± 0.067, −1.87 ± 0.62, 0.88 ± 0.15	2.60 ± 0.067, −1.87 ± 0.63, 0.87 ± 0.15		

concentration increased from 0 to 0.225 mol·kg^{-1}, TR deviates from ideal behavior much less than either IC or CA. When the salt concentration is ≥0.15 mol·kg^{-1}, the logarithm of the activity coefficient of IC has an average value of 0.158 ± 0.005.

According to Sugunan et al. (1995), the salting-out effect, which has positive Setschenow coefficients (i.e., solubility decreases as the salt concentration increases), occurs because the entry of the scavenger's molecule is obstructed by the small rigid, highly charged ions.[11] Moreover, the electronic repulsion[18,24] and enhancement of the hydrophobic effect[25−29] of these high-charge-density anions (Cl^-) can cause the salting-out effect. At the solute−water interface, to minimize the entropic penalty associated with the highly ordered structure, solute aggregation is caused by the hydrophobic effect in pure water.[30] On the other hand, Bockris et al. (1951)[31] and Long-McDevit (1952)[18] stated that the salting-in effect is caused by dispersion forces between the ions and a neutral solute and solvent molecules. As shown in Table 4, the value of the Setschenow coefficient, on a molar scale, is similar to that in molality because molality equals $n_{NaCl}/(\rho_{water} \times V_{water})$, which approximately equals n_{NaCl}/V_{water} = molarity. Moreover, the Setschenow coefficient trend is CA > IC > TR.

4.2. Pitzer's Parameters Calculation and Octanol/Water Partition Coefficient. Normally, solubilities of nonionic mixtures can be predicted using activity and osmotic coefficients. In ionic aqueous solutions, however, the use of activity coefficients can result in large errors when predicting solubility. Ionic interaction models can be used to calculate the properties of electrolyte components and better predict the solubilities of ionic mixtures. Pitzer and many other researchers[32−35] have used a single set of equations to describe the osmotic and activity coefficients of the components of single salts and mixtures with common ions. By using virial expansion expressions to account for the ionic strength, the dependence of the short-range forces in binary and ternary ion interactions can be modeled by Pitzer's model or the extended Debye−Hückel[36] method. Hence, the model is applicable to solutions of high ionic strength.[37] Moreover, the ion interaction parameters are expressed as explicit functions of ionic strength and need very few parameters to estimate the properties of both single and mixed electrolytes.[38−44]

This work obtained Pitzer's ion interaction parameters from activity coefficients that were obtained from solubility experimental data. Protonation constants are related to equilibrium by

$$H^+ + H_{i-1}L^{i-3} = H_iL^{i-2} \tag{6}$$

where L is the anion of the scavenger.

In addition, in salt, MX can be expressed by

$$\log_{10} K_i^{H} = \log_{10} (KT)_i^{H} + \log_{10} \gamma_H + \log_{10} \gamma_{H_{i-1}L} - \log_{10} \gamma_{H_iL} \tag{7}$$

where $(KT)_i^H$ is the protonation constant at infinite dilution and γ_i is the activity coefficient of the ith component. The Debye−Hückel,[36] specific ion interaction theory (SIT),[45−47] and Pitzer's equations[48,49] can be used to study the dependence of protonation constants on ionic strength. Bretti et al. (2008 and 2004)[17,50] used a modified version of the SIT equation, in which the ionic strength and the interaction coefficient, ε, has the relationship as shown in eq 8a.

$$\varepsilon = \varepsilon_\infty + \frac{\varepsilon_0 - \varepsilon_\infty}{I + 1} \tag{8a}$$

The ionic strength dependence of ε was proposed, as shown in eq 8b.[51]

$$\varepsilon = \varepsilon^{(0)} + \varepsilon^{(1)} \ln(1 + I) \tag{8b}$$

Pitzer's equations[48] express the activity coefficient of cation M or anion X, in the presence of electrolytes of 1−1, 1−2, and 2−1 charge type, by the following:

$$\ln_M \text{ and } \ln_X = f(I;\ \beta^{(0)};\ \beta^{(1)};\ C^{\phi};\ \theta;\ \psi) \tag{8c}$$

In addition, for neutral species

$$\ln_{MX}^{0} = 2\lambda I \tag{8d}$$

where I is the ionic strength (in molality); $\beta^{(0)}$, $\beta^{(1)}$, $\beta^{(1)}$, and C^{ϕ} represent the interaction parameters between two ions of opposite signs; θ are interaction parameters between two ions of the same sign (+ + , − −); and ψ are triple interaction parameters (+ − + , − + −).[52] When $I < 3$ mol·kg^{-1} of H_2O, θ and ψ are negligible. By combining eq 4 and 8d, Bretti et al. (2005, 2006, 2009, 2011, and 2013)[53−57] developed eq 9 that calculates Pitzer's interaction parameter of neutral species, λ.

Table 5. K_{OW} of Scavengers (Calculated in Eq 10) and Infinite Dilution Activity Coefficients of Scavengers

	λ	K_{OW}	$\gamma^{\infty,w}$	$\gamma^{\infty,o}$	$\gamma^{\infty,op}$
CA	1.07	18.98	0.9627	0.0074	0.0076
TR	2.52, 2.29, 1.87	394.98, 245.72, 105.17	0.7464	0.0003	0.0005
IC	1.63, 1.46, 1.08	64.02, 44.99, 19.48	0.8882	0.0028	0.0031

The calculated Pitzer's interaction parameters (for a 1:1 electrolyte) are presented in Table 4.

$$\lambda = \frac{(\ln 10)k_m}{2} \tag{9}$$

The octanol−water partition coefficient, K_{OW}, is the ratio of a chemical's concentration in the octanol phase relative to its concentration in the aqueous phase at equilibrium. It is a physical property and can be used to measure a component's hydrophobic or hydrophilic properties.[58,59] Hence, K_{OW} is mainly used to assess the potential environmental behavior, which includes physical and biological phenomena.[42,43] Moreover, K_{OW} can be used to describe the soil adsorption and toxicology of chemicals.[60] Ni and Yalkowsky (2003)[61] and Bretti et al.(2005)[54] proposed an empirical equation for NaCl to calculate K_{OW} using k_c (Table 5). K_{OW} represents the tendency of the species to partition itself between the organic phase and an aqueous phase. Hence, chemicals with low K_{OW} values may be considered relatively hydrophilic.

$$k_c = 0.039K_{OW} + 0.117 \tag{10}$$

As shown in Table 5, CA is the most hydrophilic when comparing to TR and IC. On the other hand, TR is very hydrophobic due to the very large values of K_{OW}. Moreover, Tse and co-workers (1994) showed the correlation of K_{OW} and infinite dilution activity coefficients (Table 4) through Eqs 11, 12, and 13.[46] Since TR and IC have three different values of K_{OW}, an average K_{OW} was used.

$$K_{OW} = 0.1508\frac{\gamma^{\infty,w}}{\gamma^{\infty,op}} \tag{11}$$

$$\pi = 0.1508\frac{\gamma^{\infty,w}}{\gamma^{\infty,o}} \tag{12}$$

$$\log K_{OW} = 0.10 + 0.91 \log \pi \tag{13}$$

where $\gamma^{\infty,w}$, $\gamma^{\infty,op}$, and $\gamma^{\infty,o}$ are the infinite dilution activity coefficient in the pure water, octanol phase, and pure octanol, respectively. The factor of 0.1508 is the ratio of the total molar concentration of the 1-octanol-rich phase (8.378 mol·L^{-1}) to that of the water-rich phase (55.56 mol·L^{-1}).

The linear relationship of K_{OW} and π was determined by using literature data.[62] However, Tse et al. (1994) showed that the model works for volatile hydrocarbons, aromatics, and halogenated aliphatic substances such as CCl_4, $CHCl_3$, pentane, cyclohexane, and heptane. Since H_2S scavengers are triazine-based and are not as similar to substances reported in the literature, further experiments are required to validate the model. The models provided here are good first-round approximations.

5. CONCLUSIONS

The solubility of cyanuric acid (CA), 1,3,5-triacryloylhexahydro-1,3,5-triazine (TR), and 1,3,5-tris(2-hydroxyethyl) cyanuric acid (CI) in NaCl/H_2O mixtures (0 to 0.226 mol·kg^{-1}) at T = 298.15 K was determined. Overall, the solubility in the salt solution had a trend of IC ≫ CA > TR, except for when the salt concentration was 0.14−0.18 mol·kg^{-1}. In this salt concentration range, the solubility trend was IC ≫ TR > CA. As the salt concentration increased, the solubility of CA decreased. TR and IC, on the other hand, exhibited the phenomenon known as salting in and salting out, which was based on the salt concentration.

Since the activity coefficient of the neutral species is roughly correlated with the solubility of the scavengers, it also exhibited the same trend as solubility. Moreover, deviations from the ideal behavior trend were TR < IC < CA and TR < CA < IC when the concentration of NaCl was <0.16 mol·kg^{-1} and the concentration of NaCl was >0.16 mol·kg^{-1}, respectively. From the Setschenow coefficients, Pitzer's interaction parameters of neutral species, octanol−water partition coefficients, and infinite dilution activity coefficients of the scavengers were also calculated. The Setschenow coefficients trend was CA < IC < TR. From the calculated values, the octanol−water partition coefficient trend is CA < IC ≪ TR. The trend for the octanol−water partition coefficient and the infinite dilution activity coefficients trend is CA < IC < TR due to proportionality.

■ AUTHOR INFORMATION

Corresponding Author

Tracy J. Benson − *Center for Midstream Management and Science, Dan F. Smith Department of Chemical Engineering, Lamar University, Beaumont, Texas 77705, United States;* orcid.org/0000-0001-6347-8325; Phone: 1+ (409)880-7536; Email: tracy.benson@lamar.edu

Author

Linh Doan − *Center for Midstream Management and Science, Dan F. Smith Department of Chemical Engineering, Lamar University, Beaumont, Texas 77705, United States*

Complete contact information is available at: https://pubs.acs.org/10.1021/acs.jced.9b00898

Notes

The authors declare no competing financial interest.

■ ACKNOWLEDGMENTS

This work was funded in part by the Texas Hazardous Waste Research Center and the Lamar University Center for Midstream Management and Science. The authors would like to especially thank Professor Jerry Lin (Civil & Environmental Engineering at Lamar University) for use of the potentiometer and Professor Richard Lumpkin (Chemistry and Biochemistry at Lamar University) for his insightful contributions on acid/base chemistries.

■ REFERENCES

(1) Agbroko, O. W.; Piler, K.; Benson, T. J. A Comprehensive Review of H $_2$ S Scavenger Technologies from Oil and Gas Streams. *ChemBioEng Rev.* **2017**, *4* (6), 339−359.

(2) Hudgins, C. M.; McGlasson, R. L.; Mehdizadeh, P.; Rosborough, W. M. Hydrogen Sulfide Cracking of Carbon and Alloy Steels. *CORROSION* **1966**, *22* (8), 238−251.

(3) Buller, J.; Carpenter, J. F. H2S Scavengers for Non-Aqueous Systems. In *SPE International Symposium on Oilfield Chemistry*; Society of Petroleum Engineers: The Woodlands, Texas, 2005..

(4) Kohl, A. L.; Nielsen, R. *Gas Purification*, 5th ed.; Gulf Pub: Houston, Texas, 1997.

(5) Galán Sánchez, L. M.; Meindersma, G. W.; de Haan, A. B. Solvent Properties of Functionalized Ionic Liquids for CO_2 Absorption. *Chem. Eng. Res. Des.* **2007**, *85* (1), 31−39.

(6) Bretti, C.; De Stefano, C.; Lando, G.; Sammartano, S. Thermodynamic Properties of Melamine (2,4,6-Triamino-1,3,5-Triazine) in Aqueous Solution. Effect of Ionic Medium, Ionic Strength and Temperature on the Solubility and Acid-Base Properties. *Fluid Phase Equilib.* **2013**, *355*, 104−113.

(7) Chapman, R. P.; Averell, P. R.; Harris, R. R. Solubility of Melamine in Water. *Ind. Eng. Chem.* **1943**, *35* (2), 137−138.

(8) Fan, K.; Niu, L.; Li, J.; Feng, R.; Qu, R.; Liu, T.; Song, J. Application of Solubility Theory in Bi-Component Hydrogels of Melamine with Di(2-Ethylhexyl) Phosphoric Acid. *Soft Matter* **2013**, *9* (11), 3057.

(9) Sharmin, E.; Imo, L.; Ashraf, S. M.; Ahmad, S. Acrylic-Melamine Modified DGEBA-Epoxy Coatings and Their Anticorrosive Behavior. *Prog. Org. Coat.* **2004**, *50* (1), 47−54.

(10) Zanetti, M.; Pizzi, A.; Beaujean, M.; Pasch, H.; Rode, K.; Dalet, P. Acetals-Induced Strength Increase of Melamine-Urea-Formaldehyde (MUF) Polycondensation Adhesives. II. Solubility and Colloidal State Disruption. *J. Appl. Polym. Sci.* **2002**, *86* (8), 1855−1862.

(11) Zanetti, M.; Pizzi, A. Low Addition of Melamine Salts for Improved Melamine-Urea-Formaldehyde Adhesive Water Resistance. *J. Appl. Polym. Sci.* **2003**, *88* (2), 287−292.

(12) Bachhuber, K.; Frösch, D. Melamine Resins, a New Class of Water-Soluble Embedding Media for Electron Microscopy. *J. Microsc.* **1983**, *130* (1), 1−9.

(13) Bann, B.; Miller, S. A. Melamine And Derivatives Of Melamine. *Chem. Rev.* **1958**, *58* (1), 131−172.

(14) Bretti, C.; Cigala, R. M.; Crea, F.; Foti, C.; Sammartano, S. Solubility and Activity Coefficients of Acidic and Basic Non-Electrolytes in Aqueous Salt Solutions. *Fluid Phase Equilib.* **2008**, *263* (1), 43−54.

(15) Bretti, C.; Crea, F.; De Stefano, C.; Sammartano, S. Solubility and Activity Coefficients of 2,2′-Bipyridyl, 1,10-Phenanthroline and 2,2′,6′,2″-Terpyridine in NaCl(Aq) at Different Ionic Strengths and T = 298.15K. *Fluid Phase Equilib.* **2008**, *272* (1−2), 47−52.

(16) Setschenow, J. Z. Über die Konstitution der Salzlösungen auf Grund ihres Verhaltens zu Kohlensäure. *Z. Phys. Chem.* **1889**, *4* (1), 117−125.

(17) Bretti, C.; De Stefano, C.; Millero, F. J.; Sammartano, S. Modeling of Protonation Constants of Linear Aliphatic Dicarboxylates Containing -S-Groups in Aqueous Chloride Salt Solutions, at Different Ionic Strengths, Using the SIT and Pitzer Equations and Empirical Relationships. *J. Solution Chem.* **2008**, *37* (6), 763−784.

(18) Long, F. A.; McDevit, W. F. Activity Coefficients of Nonelectrolyte Solutes in Aqueous Salt Solutions. *Chem. Rev.* **1952**, *51* (1), 119−169.

(19) He, L.; Liu, Y.; Lin, M.; Awika, J.; Ledoux, D. R.; Li, H.; Mustapha, A. A New Approach to Measure Melamine, Cyanuric Acid, and Melamine Cyanurate Using Surface Enhanced Raman Spectroscopy Coupled with Gold Nanosubstrates. *Sens. & Instrumen. Food Qual.* **2008**, *2* (1), 66−71.

(20) Seifer, G. B. Cyanuric Acid and Cyanurates. *Russ. J. Coord. Chem.* **2002**, *28* (5), 301−324.

(21) Bretti, C.; Giacalone, A.; Gianguzza, A.; Milea, D.; Sammartano, S. Modeling S-Carboxymethyl-l-Cysteine Protonation and Activity Coefficients in Sodium and Tetramethylammonium Chloride Aqueous Solutions by SIT and Pitzer Equations. *Fluid Phase Equilib.* **2007**, *252* (1−2), 119−129.

(22) Bretti, C.; Crea, F.; De Stefano, C.; Sammartano, S.; Vianelli, G. Some Thermodynamic Properties of Dl-Tyrosine and Dl-Tryptophan. Effect of the Ionic Medium, Ionic Strength and Temperature on the Solubility and Acid-Base Properties. *Fluid Phase Equilib.* **2012**, *314*, 185−197.

(23) Bretti, C.; Cigala, R. M.; Crea, F.; Lando, G.; Sammartano, S. Thermodynamics of Proton Binding and Weak (Cl-, Na+ and K+) Species Formation, and Activity Coefficients of 1,2-Dimethyl-3-Hydroxypyridin-4-One (Deferiprone). *J. Chem. Thermodyn.* **2014**, *77*, 98−106.

(24) Johnson, N. O.; Light, T. P.; MacDonald, G.; Zhang, Y. Anion-Caffeine Interactions Studied by ^{13}C and ^{1}H NMR and ATR-FTIR Spectroscopy. *J. Phys. Chem. B* **2017**, *121* (7), 1649−1659.

(25) Zangi, R.; Hagen, M.; Berne, B. J. Effect of Ions on the Hydrophobic Interaction between Two Plates. *J. Am. Chem. Soc.* **2007**, *129* (15), 4678−4686.

(26) Graziano, G. Hydrophobic Interaction of Two Large Plates: An Analysis of Salting-in/Salting-out Effects. *Chem. Phys. Lett.* **2010**, *491* (1−3), 54−58.

(27) Thomas, A. S.; Elcock, A. H. Molecular Dynamics Simulations of Hydrophobic Associations in Aqueous Salt Solutions Indicate a Connection between Water Hydrogen Bonding and the Hofmeister Effect. *J. Am. Chem. Soc.* **2007**, *129* (48), 14887−14898.

(28) Shimizu, S. Caffeine Dimerization: Effects of Sugar, Salts, and Water Structure. *Food Funct.* **2015**, *6* (10), 3228−3235.

(29) Breslow, R. Hydrophobic Effects on Simple Organic Reactions in Water. *Acc. Chem. Res.* **1991**, *24* (6), 159−164.

(30) Blokzijl, W.; Engberts, J. B. F. N. Hydrophobic Effects. Opinions and Facts. *Angew. Chem., Int. Ed. Engl.* **1993**, *32* (11), 1545−1579.

(31) Bockris, J. O.; Bowler-Reed, J.; Kitchener, J. A. The Salting-in Effect. *Trans. Faraday Soc.* **1951**, *47*, 184.

(32) Ma, X.-C.; Li, X.-P.; He, X.-F.; Sang, S.-H.; Lei, N.-F.; Nie, Z. Thermodynamic Study of the NaCl-$CuCl_2$-H_2O Ternary System at 298.15 K by the Electromotive Force Method. *J. Chem. Eng. Data* **2019**, *64* (1), 90−97.

(33) Dou, Z.; Li, S.; Zhai, Q.; Jiang, Y.; Hu, M. Potentiometric Investigation of the Thermodynamic Properties of Mixed Electrolyte Systems at 298.2 K: CsF + CsBr + H_2O and CsF + $CsNO_3$ + H_2O. *J. Chem. Eng. Data* **2018**, *63* (10), 3801−3808.

(34) Temoltzi-Avila, J.; Iglesias-Silva, G. A.; Ramos-Estrada, M. Comparison Among Pitzer Model and Solvation Models. Calculation of Osmotic and Activity Coefficients and Dilution Enthalpy for Single-Electrolyte Aqueous Solutions. *Ind. Eng. Chem. Res.* **2018**, *57* (31), 10684−10700.

(35) Liang, Z.; Cheng, X.; Zhang, L.; Yang, Z.; Ran, J.; Ding, L. Study of Main Solutes on Evaporation and Crystallization Processes of the Desulfurization Wastewater Droplet. *Energy Fuels* **2018**, *32* (5), 6119−6129.

(36) Daniele, P. G.; De Robertis, A.; De Stefano, C.; Sammartano, S.; Rigano, C. On the Possibility of Determining the Thermodynamic Parameters for the Formation of Weak Complexes Using a Simple Model for the Dependence on Ionic Strength of Activity Coefficients: Na+, K+, and Ca2+ Complexes of Low Molecular Weight Ligands in Aqueous Solution. *J. Chem. Soc., Dalton Trans.* **1985**, No. 11, 2353.

(37) Kim, H. T.; Frederick, W. J. Evaluation of Pitzer Ion Interaction Parameters of Aqueous Electrolytes at 25. Degree. C. 1. Single Salt Parameters. *J. Chem. Eng. Data* **1988**, *33* (2), 177−184.

(38) Lach, A.; André, L.; Guignot, S.; Christov, C.; Henocq, P.; Lassin, A. A Pitzer Parametrization To Predict Solution Properties and Salt Solubility in the H-Na-K-Ca-Mg-NO_3-H_2O System at 298.15 K. *J. Chem. Eng. Data* **2018**, *63* (3), 787−800.

(39) Wang, D.; Wen, X.-H.; Zhang, W.-Y.; Sang, S.-H. Thermodynamic Study of the $NaNO_3$-$Cd(NO_3)_2$-H_2O Ternary System at 298.15 K by the Potential Difference Method. *J. Chem. Eng. Data* **2017**, *62* (4), 1232−1239.

(40) Wang, D.; Yang, Y.-Y.; Zhang, X.-P.; Sang, S.-H. Mean Activity Coefficients of NaCl in NaCl-$CdCl_2$-H_2O Ternary System at

298.15 K by Potential Difference Method. *J. Chem. Eng. Data* **2016**, *61* (9), 3027−3033.

(41) Ren, Y.; Liu, C.; Cao, J.; Mu, W.; Ding, X. Liquid-Liquid Equilibrium Data of Extraction with Tri- *n* -Butyl Phosphate on Phosphoric Acid Containing Sodium Sulfate or Potassium Sulfate. *J. Chem. Eng. Data* **2016**, *61* (5), 1735−1742.

(42) Zhang, X.; Ning, P.; Cao, H.; Zhang, Y. Measurement and Modeling for Molybdenum Extraction from the Na_2MoO_4-H_2SO_4-H_2O System by Primary Amine N1923. *Ind. Eng. Chem. Res.* **2016**, *55* (5), 1427−1438.

(43) Zhou, M.-F.; Sang, S.-H.; Liu, Q.-Z.; Wang, D.; Fu, C. Mean Activity Coefficients of NaCl in NaCl + $SrCl_2$ + H_2O Ternary System at 298.15 K Determined by Potential Difference Measurements. *J. Chem. Eng. Data* **2015**, *60* (11), 3209−3214.

(44) Zhou, M.-F.; Sang, S.-H.; Zhang, J.-J.; Hu, J.-X.; Zhong, S.-Y. Studies on Mean Activity Coefficients of NaBr in NaBr-$SrBr_2$-H_2O Ternary System at 298.15 K by EMF Method. *J. Chem. Eng. Data* **2014**, *59* (11), 3779−3784.

(45) Guggenheim, E. A.; Turgeon, J. C. Specific Interaction of Ions. *Trans. Faraday Soc.* **1955**, *51*, 747.

(46) Scatchard, G. Concentrated Solutions of Strong Electrolytes. *Chem. Rev.* **1936**, *19* (3), 309−327.

(47) Brönsted, J. N. Studies on Solubility. IV. The Principle of the Specific Interaction of Ions. *J. Am. Chem. Soc.* **1922**, *44* (5), 877−898.

(48) Pitzer, K. S. *Activity Coefficients in Electrolyte Solutions*; Taylor & Francis Group: Boca Raton, Florida, 2018.

(49) Millero, F. J. *The Physical Chemistry of Natural Waters*; Wiley-Interscience Series in Geochemistry; Wiley-Interscience: New York, 2001.

(50) Bretti, C.; Foti, C.; Sammartano, S. A New Approach in the Use of SIT in Determining the Dependence on Ionic Strength of Activity Coefficients. Application to Some Chloride Salts of Interest in the Speciation of Natural Fluids. *Chem. Speciation Bioavailability* **2004**, *16* (3), 105−110.

(51) Ciavatta, L. The Specific Interaction Theory in Evaluating Ionic Equilibria. *Ann. Chim. Rome* **1980**, *70*, 551−562.

(52) Majlesi, K.; Bretti, C.; De Stefano, C.; Lando, G.; Majlesi, K.; Sammartano, S. Thermodynamic Study on the Interaction of Nicotinic Acid with H+, Na+, Ca2+ and Mg2+ at Different Temperatures and Ionic Strengths. *J. Solution Chem.* **2019**, *48*, 1671.

(53) Bretti, C.; Crea, F.; Foti, C.; Sammartano, S. Solubility and Activity Coefficients of Acidic and Basic Nonelectrolytes in Aqueous Salt Solutions. 2. Solubility and Activity Coefficients of Suberic, Azelaic, and Sebacic Acids in NaCl(Aq), $(CH_3)_4NCl$(Aq), and $(C_2H_5)_4NI$(Aq) at Different Ionic Strengths and at t = 25 °C. *J. Chem. Eng. Data* **2006**, *51* (5), 1660−1667.

(54) Bretti, C.; Crea, F.; Foti, C.; Sammartano, S. Solubility and Activity Coefficients of Acidic and Basic Nonelectrolytes in Aqueous Salt Solutions. 1. Solubility and Activity Coefficients of *o* -Phthalic Acid and L -Cystine in NaCl(Aq), $(CH_3)_4NCl$(Aq), and $(C_2H_5)_4NI$(Aq) at Different Ionic Strengths and at t = 25 °C. *J. Chem. Eng. Data* **2005**, *50* (5), 1761−1767.

(55) Bretti, C.; De Stefano, C.; Foti, C.; Giuffrè, O.; Sammartano, S. Thermodynamic Protonation Parameters of Some Sulfur-Containing Anions in NaClaq and (CH3)4NClaq at T = 25 °C. *J. Solution Chem.* **2009**, *38* (10), 1225−1245.

(56) Bretti, C.; De Stefano, C.; Foti, C.; Sammartano, S. Acid-Base Properties, Solubility, Activity Coefficients and Na+ Ion Pair Formation of Complexons in NaCl(Aq) at Different Ionic Strengths. *J. Solution Chem.* **2013**, *42* (7), 1452−1471.

(57) Bretti, C.; De Stefano, C.; Manfredi, G.; Sammartano, S. Solubility, Activity Coefficients and Acid-Base Properties of Three Naphthol Derivatives in NaCl(Aq) at Different Ionic Strengths and at T = 298.15K. *J. Mol. Liq.* **2011**, *158* (1), 50−56.

(58) Meylan, W. M.; Howard, P. H. Atom/Fragment Contribution Method for Estimating Octanol-Water Partition Coefficients. *J. Pharm. Sci.* **1995**, *84* (1), 83−92.

(59) Banerjee, S.; Yalkowsky, S. H.; Valvani, C. Water Solubility and Octanol/Water Partition Coefficients of Organics. Limitations of the Solubility-Partition Coefficient Correlation. *Environ. Sci. Technol.* **1980**, *14* (10), 1227−1229.

(60) Sangster, J. Octanol-Water Partition Coefficients of Simple Organic Compounds. *J. Phys. Chem. Ref. Data* **1989**, *18* (3), 1111−1229.

(61) Ni, N.; Yalkowsky, S. H. Prediction of Setschenow Constants. *Int. J. Pharm.* **2003**, *254* (2), 167−172.

(62) Tse, G.; Sandler, S. I. Determination of Infinite Dilution Activity Coefficients and 1-Octanol/Water Partition Coefficients of Volatile Organic Pollutants. *J. Chem. Eng. Data* **1994**, *39* (2), 354−357.

https://dx.doi.org/10.1021/acs.jced.9b00898