Physicochemical Properties of Aqueous Hydroxylamine Sulfate and Aqueous (Hydroxylamine Sulfate + Ammonium Sulfate) Solutions at Different Temperatures

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The densities of an aqueous solution of pure hydroxylamine sulfate at T = (283.15, 288.15, 293.15, 298.15, 303.15, 308.15, and 313.15) K and a solution of hydroxylamine sulfate containing ammonium sulfate at T = (293.15, 298.15, 303.15, 308.15, and 313.15) K have been determined over a wide range of concentrations. The viscosities of binary and ternary mixtures of these compounds at T = (298.15, 303.15, and 308.15) K are also presented. The Masson-type equation and extended Jones−Dole equation in terms of ionic strength are used to correlate the density and viscosity, respectively, of aqueous solutions of hydroxylamine sulfate. The ideal mixing rule of Young has been successfully applied to predict the ternary densities and viscosities of aqueous hydroxylamine sulfate + ammonium sulfate based on relevant binary solution properties.

Introduction

Aqueous solutions of hydroxylamine sulfate in the presence of ammonium sulfate and sulfuric acid, produced by the Raschig process, are frequently used for polyamide manufacture.[1] Pure hydroxylamine sulfate is also necessary in industry for manufacture of various chemicals, including herbicides and pharmaceuticals. Over the past century, various technologies have been proposed to separate hydroxylamine sulfate from the aqueous solution of ammonium sulfate produced by the Raschig process.[1] The liquid ion-exchange extraction method has shown advantages because of its high efficiency and low cost and it can be performed in a continuous process.[2,3] In the process, the Raschig mixtures are first naturalized with aqueous ammonia and then extracted by a nonpolar solvent containing organophosphate extractant.[4] The industrial applications generally involve an organic phase containing organophosphate extractant and an aqueous phase containing hydroxylamine and ammonium salts. Moreover, the aqueous phase involves moderate to very high salt concentrations and different portions of hydroxylamine and ammonium salts in the extraction stages. Thus, reliable data and modeling on the physicochemical properties of the binary and ternary mixtures are needed over a wide range of concentrations and temperatures for simulation and optimization of the extraction process.

Previous works have been devoted to the measurement of densities and viscosities of the organophosphate extractant in different nonpolar solvents.[5,6] This work reports the properties of aqueous solutions of pure hydroxylamine sulfate and solutions of hydroxylamine sulfate + ammonium sulfate at different temperatures. The properties of aqueous hydroxylamine sulfate have been reported by Skvortsov et al.;[7] however data for the industrially important ternary mixtures of hydroxylamine sulfate + ammonium sulfate + H_2O have not been presented. For aqueous hydroxylamine sulfate solutions, the literature[7] is quite old and not easy to obtain. In this work the solution densities of aqueous hydroxylamine sulfate were determined by high-precision vibrating-tube densimetry. The densities and viscosities of binary mixtures were correlated by the Masson-type equation[8] and extended Jones−Dole equation[9] in terms of ionic strength, respectively. We found that the linear additivity rule of Young[10] can be successfully applied to predict the densities and viscosities of aqueous hydroxylamine sulfate + ammonium sulfate mixtures. The simple additivity rule assumes that the properties of a mixed solution at a given composition is additive in the properties of the individual binary solutions.

Experimental Section

Materials. Hydroxylamine sulfate with a minimum purity in mass fraction of 0.995 was obtained from UBE (Japan). Ammonium sulfate with a minimum purity in mass fraction of 0.999 was obtained from the Juhua Group Corp. Polyamide Fiber Factory (Quzhou, China). The salt was placed and dried in a vacuum desiccator and used without further purification. Double-distilled deionized water was used for the preparation of aqueous solutions.

Aqueous solutions were prepared from a known mass of salt and water in a stoppered glass bottle. The masses were recorded on a Sartorius Corp. BS 224S balance to a precision of $\pm 1 \cdot 10^{-4}$ g. The liquid samples were filtered by using syringes equipped with 0.45 μm membrane filters before density and viscosity measurements were recorded.

Experimental Measurements. The densities of the binary and ternary mixtures were measured with an Anton Paar DMA 4500 density meter whose measurement cell temperature was controlled automatically within ± 0.01 K of the selected value. It was calibrated at atmospheric pressure with double-distilled deionized water and dry air. Densities, both of water and of dry air, at the various working temperatures were given by the

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10.1021/je8008649 CCC: $40.75

Table 1. Density of Hydroxylamine Sulfate in Water from $T = (283.15 \text{ to } 313.15)$ K

		ρ/g·cm^{-3}						
$m(SO_4^{2-})$/mol·kg^{-1}	I/mol·kg^{-1}	$T = 283.15$ K	$T = 288.15$ K	$T = 293.15$ K	$T = 298.15$ K	$T = 303.15$ K	$T = 308.15$ K	$T = 313.15$ K
0.0075	0.0224	1.00057	0.99995	0.99903	0.99789	0.99646	0.99484	0.99300
0.0380	0.1141	1.00384	1.00318	1.00223	1.00103	0.99959	0.99795	0.99609
0.0702	0.2107	1.00727	1.00656	1.00558	1.00434	1.00288	1.00117	0.99912
0.2199	0.6596	1.02247	1.02157	1.02043	1.01905	1.01748	1.01570	1.01304
0.3709	1.1127	1.03687	1.03581	1.03452	1.03305	1.03138	1.02951	1.02724
0.4314	1.2942	1.04264	1.04122	1.03997	1.03836	1.03673	1.03477	1.03285
0.5334	1.6002	1.05159	1.05039	1.04898	1.04739	1.04561	1.04369	1.04160
0.8069	2.4206	1.07532	1.07390	1.07196	1.07020	1.06830	1.06630	1.06415
0.9684	2.9051	1.08831	1.08680	1.08512	1.08329	1.08132	1.07923	1.07698
1.1647	3.4940	1.10323	1.10158	1.09981	1.09789	1.09585	1.09370	1.09142
1.4781	4.4343	1.12668	1.12491	1.12300	1.12096	1.11882	1.11650	1.11363
1.8505	5.5516	1.15194	1.14999	1.14792	1.14549	1.14234	1.14116	1.13836
2.4613	7.3840	1.18898	1.18687	1.18469	1.18241	1.18007	1.17764	1.17514
2.9977	8.9932	1.21805	1.21584	1.21355	1.21121	1.20879	1.20632	1.20399

Table 2. Polynomial Coefficients of Equation 5 for Densities of Aqueous Hydroxylamine Sulfate Solution

a_1	a_2	b_1	b_2	c_1	c_2	100 ARD	rmsd
0.012324	7.237611	0.002835	−1.979776	−0.000479	0.098221	0.014	0.00021

manufacturer in the instruction manual. The reproducibility of the density measurement was within ± $5 \cdot 10^{-5}$ g·cm^{-3}.

The viscosities of the solutions were determined by using a Ubbelohde viscometer with capillaries (0.3 to 0.4) mm or (0.5 to 0.6) mm in diameter according to the viscosity of the samples. It was filled with experimental liquid and placed vertically in a DF-02 transparent-walled water bath (Fangao Scientific Co., Ltd., Nanjing, China), which was maintained constant to ± 0.01 K at the selected temperature. To minimize the evaporation loss during heating and measurement, the viscometer's limb was closed using a rubber stopper with a small plastic tube in the center which was open to the atmosphere. An electronic digital stopwatch with a precision of ± 0.01 s was used for flow-time measurements. The viscosity of the solutions can be expressed as

$$\frac{\eta}{\rho} = k_1 t - k_2/t \tag{1}$$

where ρ is the density of the liquid, t is the flow time, and k_1, and k_2 are the viscometer constants. The k_1 and k_2 parameters for both Ubbelohde viscometers were obtained by the measurements on double-distilled deionized water at (288.15, 293.15, 298.15, 303.15, and 308.15) K. It is convenient to get $k_1 = 0.003486$ and $k_2 = -0.268447$ for the (0.3 to 0.4) mm viscometer and $k_1 = 0.009773$ and $k_2 = 0.810429$ for the (0.5 to 0.6) mm viscometer by the linear regression of $\eta t/\rho$ with t^2. Measurements were repeated at least four to five times for each solution and temperature. The reproducibility of the viscosity estimates was found to be within ± 0.1 %.

Results and Discussion

Densities for $(NH_3OH)_2SO_4 + H_2O$ Solutions. The densities of $(NH_3OH)_2SO_4 + H_2O$ solutions are listed in Table 1. The experimental data can be described by the Masson-type equation[11,12] in terms of ionic strength in the form

$$\rho = \rho_{H_2O} + aI + bI^{3/2} + cI^2 \tag{2}$$

with ionic strength in molality I defined as

$$I = \frac{1}{2}\sum_i m_i z_i^2 \tag{3}$$

where m_i is the molality of ion i in moles per kilogram of pure solvent H_2O and z_i represents the ion charges. The temperature dependence of parameters a, b, and c in eq 2 will facilitate the density interpretation at other temperatures that are not determined by experiment. Thus, the following equation for the temperature dependence of the three parameters is used

$$P = P_1 + \frac{P_2}{T} \tag{4}$$

where P represents the parameters in eq 2, T is the absolute temperature in Kelvin, and P_1 and P_2 are constants. Thus, the temperature and concentration dependence of the density equation can be written as

$$\rho = \rho_{H_2O} + \left(a_1 + \frac{a_2}{T}\right)I + \left(b_1 + \frac{b_2}{T}\right)I^{3/2} + \left(c_1 + \frac{c_2}{T}\right)I^2 \tag{5}$$

The values of the six parameters in eq 5 were estimated by the Levenberg−Marquardt algorithm with the help of SPSS software

Table 3. Densities of Aqueous Hydroxylamine Sulfate + Ammonium Sulfate Mixture at Different Temperatures

		ρ/g·cm^{-3}									
		T = 293.15 K		T = 298.15 K		T = 303.15 K		T = 308.15 K		T = 313.15 K	
$m(NH_3OH^+)$/mol·kg^{-1}	$m(NH_4^+)$/mol·kg^{-1}	exp	calcd[a]	exp	calcd[a]	exp	calcd[a]	exp	calcd[a]	exp	calcd[a]
0.0000	9.3100	1.21766	1.21644	1.21579	1.21517	1.21389	1.21366	1.21194	1.21193	1.20997	1.21001
0.5325	8.7906	1.22288	1.22040	1.22098	1.21908	1.21904	1.21752	1.21705	1.21575	1.21504	1.21378
1.0693	8.2826	1.22681	1.22459	1.22487	1.22323	1.22289	1.22163	1.22088	1.21982	1.21881	1.21780
1.3383	8.0292	1.22841	1.22671	1.22646	1.22532	1.22446	1.22370	1.22243	1.22186	1.22033	1.21983
1.6073	7.7681	1.23061	1.22871	1.22868	1.22730	1.22674	1.22565	1.22475	1.22380	1.22276	1.22174
2.1536	7.2655	1.23487	1.23314	1.23287	1.23168	1.23082	1.22999	1.22873	1.22809	1.22660	1.22600
0.2535	7.3433	1.19303	1.19229	1.19106	1.19087	1.18901	1.18922	1.18683	1.18735	1.18455	1.18528
1.0792	6.5382	1.20033	1.19917	1.19771	1.19766	1.19625	1.19592	1.19358	1.19396	1.19173	1.19181
1.7861	5.8686	1.20652	1.20535	1.20446	1.20376	1.20234	1.20194	1.20009	1.19992	1.19780	1.19770
3.1504	0.2265	1.13488	1.13509	1.13267	1.13302	1.12977	1.13075	1.12664	1.12828	1.12339	1.12564
2.4698	0.8970	1.12793	1.12816	1.12566	1.12622	1.12265	1.12407	1.11880	1.12172	1.11563	1.11919
1.7931	1.5684	1.12116	1.12137	1.11914	1.11956	1.11585	1.11753	1.11296	1.11530	1.10967	1.11288
0.8918	2.4525	1.11167	1.11207	1.10980	1.11044	1.10776	1.10857	1.10549	1.10650	1.10304	1.10422
1.2916	0.8607	1.08424	1.08433	1.08247	1.08263	1.08055	1.08072	1.07845	1.07860	1.07585	1.07629
0.6971	1.4479	1.07783	1.07780	1.07610	1.07625	1.07423	1.07447	1.07169	1.07247	1.06919	1.07029
	rmsd		0.00135		0.00095		0.00090		0.00103		0.00128
	100 ARD		0.09		0.06		0.07		0.08		0.10

[a] Calclulated by eq 8.

under the 0.95 confidence interval by minimizing the average relative deviation (ARD) as

$$\mathrm{ARD} = \frac{1}{N}\sum_{i=1}^{N}\frac{|\rho_{i,\mathrm{calcd}} - \rho_{i,\mathrm{exp}}|}{\rho_{i,\mathrm{exp}}} \qquad (6)$$

where N is the number of experimental points, $\rho_{i,\mathrm{exp}}$ represents the experimental densities, and $\rho_{i,\mathrm{calcd}}$ represents the densities calculated. The parameters a_1, a_2, b_1, b_2, c_1, and c_2 and root-mean-square deviations (rmsd) defined below are listed in Table 2.

$$\mathrm{rmsd} = \left\{\sum_{i}^{N}(\rho_{\mathrm{exp}} - \rho_{\mathrm{calcd}})^2/N\right\}^{1/2} \qquad (7)$$

From the data listed in Table 2, it can be seen that the results show good agreement with the experimental values, which indicates that eq 5 can be applied to represent the densities within the concentration and temperature range.

Densities for $(NH_3OH)_2SO_4 + (NH_4)_2SO_4 + H_2O$ Solutions. The densities of ternary mixtures for $(NH_3OH)_2SO_4$ + $(NH_4)_2SO_4$ + H_2O are collected in Table 3. In the industrial extraction stage, different portions of hydroxylamine and ammonium salts exist in the aqueous phase. The densities reported in this work will be beneficial for the research of mass transfer during extraction, the simulation of the extraction process, and the design of extraction equipment.

The simple additivity rule has been tested for its ability in the prediction of properties of the title system. The widely used Young's rule[10,12] can be applied as

$$\rho(I_{\mathrm{T}}) = \frac{I_{(NH_3OH)_2SO_4}}{I_{\mathrm{T}}}\rho_{(NH_3OH)_2SO_4}(I_{\mathrm{T}}) + \frac{I_{(NH_4)_2SO_4}}{I_{\mathrm{T}}}\rho_{(NH_4)_2SO_4}(I_{\mathrm{T}}) \qquad (8)$$

where $\rho(I_{\mathrm{T}})$ is the density of the ternary mixture using total ionic strength, I_{T}, in molality units, $I_{(NH_3OH)_2SO_4}$ and $I_{(NH_4)_2SO_4}$ are the ionic strength contributions of $(NH_3OH)_2SO_4$ and $(NH_4)_2SO_4$ in solution, respectively, and $\rho_{(NH_3OH)_2SO_4}(I_{\mathrm{T}})$ and $\rho_{(NH_4)_2SO_4}(I_{\mathrm{T}})$ are the densities of the binary solutions at total ionic strength, I_{T}, respectively. In the present case, the density of aqueous $(NH_4)_2SO_4$ solution, $\rho_{(NH_4)_2SO_4}(I_{\mathrm{T}})$, is calculated from the equation and parameters provided by Tans.[13]

Table 4. Viscosity of Hydroxylamine Sulfate in Water from T = (298.15 to 308.15) K

$m(SO_4^{2-})$		η/mPa·s		
mol·kg^{-1}	I/mol·kg^{-1}	T = 298.15 K	T = 303.15 K	T = 308.15 K
0.0204	0.0611	0.897	0.806	0.727
0.0473	0.1418	0.909	0.814	0.736
0.0702	0.2107	0.914	0.821	0.741
0.2315	0.6944	0.968	0.869	0.787
0.3709	1.1127	1.019	0.912	0.823
0.4314	1.2942	1.035	0.927	0.838
0.5334	1.6002	1.069	0.961	0.867
0.8069	2.4206	1.167	1.053	0.950
1.0436	3.1308	1.256	1.128	1.020
1.3236	3.9709	1.369	1.237	1.112
1.5892	4.7676	1.483	1.333	1.201
1.9145	5.7435	1.631	1.463	1.319
2.0882	6.2645	1.732	1.542	1.386
2.8469	8.5406	2.123	1.887	1.702

If the system follows Young's rule, the mixing in terms of density can be considered as ideal. For comparison, the calculated results by eq 8 are shown in Table 3 combined with the ARD and rmsd. It can be seen that this ideal mixing model results in average absolute deviations between experimental and calculated densities within 0.1 %. The results indicate that Young's rule is suitable and reliable to extrapolate densities of the aqueous $(NH_3OH)_2SO_4$ + $(NH_4)_2SO_4$ system. Similar results have been obtained for the aqueous H_2SO_4 + $FeSO_4$[11] and NaOH + $NaAl(OH)_4$[12,14,15] systems.

Viscosities for $(NH_3OH)_2SO_4 + H_2O$ Solutions. The viscosities of aqueous $(NH_3OH)_2SO_4$ + H_2O solutions were measured at T = (293.15, 303.15, and 308.15) K. The experimental viscosities are listed in Table 4. For electrolyte solution, the extended Jones–Dole equation[9] in terms of ionic strength, I, was used to express the relation between the viscosity and electrolyte concentration.

$$\eta_r = \frac{\eta}{\eta_0} = 1 + AI^{1/2} + BI + CI^2 \qquad (9)$$

When a linear dependence of temperature is used, the above equation can be written as

$$\eta_r = \frac{\eta}{\eta_0} = 1 + (A_1 + A_2T)I^{1/2} + (B_1 + B_2T)I + (C_1 + C_2T)I^2 \qquad (10)$$

In the above equations, η_0 is the viscosity of pure water at the selected temperature and A_i, B_i, and C_i are temperature-

Table 5. Coefficients of Equation *10* for Viscosity Fits of Hydroxylamine Sulfate in Water

A_1	A_2	B_1	B_2	C_1	C_2	100 ARD	rmsd
−0.115593	0.000435	0.060594	0.000148	0.025103	−0.000064	0.15	0.003

independent constants, which are estimated under 0.95 confidence intervals from fitting of the experimental data. The values of these parameters are reported in Table 5 combined with the AAD and rmsd of calculation. These results show that eq 10 can be satisfactorily used for correlation of the experimental viscosity data over a wide range of concentrations.

In the Jones−Dole equation, the viscosity B coefficient is a valuable tool to provide information concerning the solvation of solutes and their effects on the structure of the solvent in the near environment of solute molecules or ions.[16,17] Table 5 indicates that values of the B (calculated by B_1 and B_2) of hydroxylamine sulfate in water at different temperatures are positive, suggesting the presence of strong ion−solvent interactions.[18] The result also indicates that these interactions are strengthened with a rise in temperature. A number of studies[17,18] report that $\mathrm{d}B/\mathrm{d}T$, B_2, is a better criterion for determining the structure-making/breaking nature of any solute rather than the B coefficient. The positive B_2 value indicates the structure-breaking tendency of hydroxylamine sulfate in the studied solvent systems.

Viscosities for $(NH_3OH)_2SO_4 + (NH_4)_2SO_4 + H_2O$ Solutions. The viscosities of ternary mixtures of $(NH_3OH)_2SO_4$ + $(NH_4)_2SO_4 + H_2O$ are shown in Table 6. The results show that the replacing of NH_4^+ by NH_3OH^+ at the same ionic strength makes the solution become more viscous. This may be attributed to the association of the hydroxylamine ion to form dimers and trimers.[19] The hydroxylamine ion has a hydroxyl group with self-association and H-bonding properties. The network of self-association forces binds the hydroxylamine ion together with itself or water molecules. Extra energy is required to break this interaction.

The ideal mixing rule of Young[10] has also been used to predict the experimental viscosity data for the studied ternary mixture. It has been shown that Young's rule expresses the properties of a ternary mixture as the ionic strength-weighted sum of relevant binary solution properties. Thus, Young's rule for viscosity of an aqueous hydroxylamine sulfate + ammonium sulfate mixture, $\eta(I_T)$, can be written as

$$\eta(I_\mathrm{T}) = \frac{I_{(\mathrm{NH_3OH})_2\mathrm{SO_4}}}{I_\mathrm{T}} \eta_{(\mathrm{NH_3OH})_2\mathrm{SO_4}}(I_\mathrm{T}) + \frac{I_{(\mathrm{NH_4})_2\mathrm{SO_4}}}{I_\mathrm{T}} \eta_{(\mathrm{NH_4})_2\mathrm{SO_4}}(I_\mathrm{T}) \tag{11}$$

where I_T is the toal ionic strength in molality units, $I_{(NH_3OH)_2SO_4}$ and $I_{(NH_4)_2SO_4}$are the ionic strength contributions of $(NH_3OH)_2SO_4$ and $(NH_4)_2SO_4$ in solution $\rho_{(NH_3OH)_2SO_4}(I_T)$, respectively, and $\eta_{(NH_3OH)_2SO_4}(I_T)$ and $\eta_{(NH_4)_2SO_4}(I_T)$ are the viscosities of the binary solutions at total ionic strength, I_T, respectively. The viscosity of the aqueous $(NH_4)_2SO_4$ solution was calculated from the literature,[20] which was determined over a wide range of concentrations and temperatures.

The calculated values are listed in Table 6 for comparison. The obtained standard deviations for η at T = (298.15, 303.15, and 308.15) K are 0.015, 0.012, and 0.023, respectively. The average relative deviation between the experimental and calculated viscosity data is less than 1 %. These results show that the mixing in terms of viscosity can also be considered as ideal and eq 11 can be satisfactorily used for the prediction of ternary viscosities for the title system.

Table 6. Viscosities of Aqueous Hydroxylamine Sulfate + Ammonium Sulfate Mixture at Different Temperatures

$m(NH_3OH^+)$	$m(NH_4^+)$	η/mPa·s					
		T = 298.15 K		T = 303.15 K		T = 308.15 K	
mol·kg^{-1}	mol·kg^{-1}	exp	calcd[a]	exp	calcd[a]	exp	calcd[a]
0.0000	9.3100	2.114	2.109	1.944	1.938	1.785	1.781
0.5325	8.7906	2.219	2.181	2.011	1.995	1.824	1.831
1.0693	8.2826	2.291	2.256	2.051	2.055	1.866	1.884
1.3383	8.0292	2.290	2.294	2.071	2.085	1.879	1.911
1.6073	7.7681	2.319	2.331	2.090	2.114	1.887	1.937
2.1536	7.2655	2.410	2.411	2.157	2.178	1.939	1.994
0.2535	7.3433	1.858	1.847	1.679	1.689	1.531	1.547
1.0792	6.5382	1.929	1.948	1.758	1.770	1.598	1.619
1.7861	5.8686	2.029	2.038	1.825	1.843	1.665	1.683
3.1504	0.2265	1.504	1.510	1.350	1.356	1.223	1.222
2.4698	0.8970	1.442	1.448	1.295	1.303	1.169	1.177
1.7931	1.5684	1.387	1.387	1.248	1.251	1.138	1.133
0.8918	2.4525	1.304	1.304	1.179	1.181	1.069	1.073
1.2916	0.8607	1.200	1.199	1.081	1.081	0.981	0.978
0.6971	1.4479	1.150	1.149	1.041	1.038	0.942	0.942
	rmsd		0.015		0.012		0.023
	100 ARD		0.49		0.55		0.94

[a] Calculated by eq 11.

Conclusions

The densities and viscosities of the industrially important $(NH_4)_2SO_4 + H_2O$ and $(NH_3OH)_2SO_4 + (NH_4)_2SO_4 + H_2O$ systems are presented over a wide range of concentrations and temperatures. The Masson-type equation and Jones−Dole equation in terms of ionic strength were used to correlate the experimental binary mixture data. This work demonstrates that the ideal mixing rule of Young in terms of ionic strength can be successfully applied to predict the properties of the ternary $(NH_3OH)_2SO_4 + (NH_4)_2SO_4 + H_2O$ system.

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Received for review November 15, 2008. Accepted March 27, 2009. This work was supported by the National Key Technology R&D Program of China (Project No. 2007BAI34B07) and the Commission of Science and Technology of Zhejiang Province (Project No. 2008C01006-1).

JE8008649