

Diffusivity, Density, Viscosity and Refractive Index of Nickel Ammonium Sulfate Aqueous Solutions

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Differential diffusivities and refractive indices of the system nickel ammonium sulfate in water have been measured at 25° C. at various concentrations up to near saturation. In addition, the densities and viscosities of these solutions have been measured over the temperature range 10° to 35° C. and over the range up to near saturation. Solubilities also have been checked over the range 10° to 49° C.

SEVERAL basic physical properties of nickel ammonium sulfate aqueous solutions [stable phase $\text{Ni}(\text{NH}_4)_2(\text{SO}_4)_2 \cdot 6\text{H}_2\text{O}$] have been measured as the initial part of an investigation into the precipitation characteristics and crystal growth mechanism of this substance. In all this work, deionized water and salt which was recrystallized several times were used.

Diffusion coefficients for aqueous solutions of nickel ammonium sulfate have been measured at 25° C. with a Zeiss diffusion interferometer. The experimental procedure has been described (3), and the usefulness and limitations of the method have been discussed (4). The estimated accuracy of measurement is about $\pm 2\%$. By using sufficiently small concentration differences between the two solutions under examination ($\Delta c = c_1 - c_2$), diffusivity can be assumed to vary linearly with concentration. The differential diffusivity can thus be directly determined at the mean concentration \bar{c} , of the concentration range Δc .

The differential diffusion coefficients for nickel ammonium sulfate in aqueous solution at 25° C. are listed in Table I. These data are typical of many electrolytes in water;

Table I. Diffusivity of Nickel Ammonium Sulfate in Aqueous Solution at 25° C.

Solution Concn., G. of Hydrate/100 G. of Water				Diffusion Coefficient Sq. Cm./ Sec. $\times 10^{-6}$
c_1	c_2	Δc	\bar{c}	
1.004	0	1.004	0.50	10.3
2.007	0	2.007	1.00	9.20
3.002	1.004	1.998	2.00	8.60
4.012	2.007	2.005	3.01	8.07
4.761	2.744	2.017	3.75	7.82
6.253	4.266	1.987	5.26	7.57
7.007	5.000	2.007	6.00	7.41
7.481	5.470	2.011	6.48	7.40
9.202	7.208	1.994	8.21	7.25
10.05	8.059	1.988	9.05	7.19

Table II. Refractive Indices of Nickel Ammonium Sulfate Aqueous Solutions at 25° C.

Solution Concn., G. of Hydrate/ 100 G. of Water	Refractive Index
0.000	1.3327
1.004	1.3339
2.007	1.3352
3.002	1.3366
4.012	1.3378
5.000	1.3390
5.997	1.3402
7.007	1.3412
7.973	1.3429
8.974	1.3439
9.987	1.3451

Table III. Densities and Viscosities of Nickel Ammonium Sulfate Aqueous Solutions

Temp., ° C.	Concn., G. of Hydrate/100 G. of Water	Density, d_4^t G./Ml.	Viscosity, cP
10	2.156	1.013	1.367
	3.646	1.022	1.409
	5.396	1.032	1.460
15	2.156	1.013	1.195
	3.646	1.022	1.228
	5.396	1.032	1.271
20	6.876	1.040	1.302
	2.156	1.012	1.045
	3.646	1.020	1.078
25	5.396	1.030	1.121
	6.876	1.038	1.145
	8.753	1.048	1.194
30	2.156	1.010	0.936
	3.646	1.019	0.970
	5.396	1.029	1.004
35	6.876	1.037	1.033
	8.753	1.047	1.070
	9.993	1.053	1.097
40	2.156	1.009	0.837
	3.646	1.017	0.864
	5.396	1.027	0.895
45	6.876	1.035	0.920
	8.753	1.045	0.954
	9.993	1.052	0.980
50	11.46	1.059	1.007
	2.156	1.006	0.759
	3.646	1.015	0.781
55	5.396	1.025	0.810
	6.876	1.033	0.832
	8.753	1.043	0.864
60	9.993	1.050	0.886
	11.46	1.057	0.913
	12.89	1.065	0.939

Table IV. Solubility of Nickel Ammonium Sulfate in Water

Temp., ° C.	Equilibrium Saturation Concn., G. of Anhydrous Salt/100 G. of Water			
	Ref. (5)	Ref. (2)	Ref. (6)	Present work
0	0.99	1.0
10	3.85	4.0	3.2	5.19
15	6.10
20	6.10	6.5	5.9	6.86
25	7.04	7.6	...	7.52
30	8.45	9.0	8.3	8.44
37.5	10.2
40	10.72	12.0	11.5	10.9
45	11.8
47	12.1
49	12.5
50	12.96	14.5	14.4	...

the diffusivity decreases very rapidly as concentration increases in the dilute region, but becomes much less dependent on concentration in the concentrated region.

Refractive indices of nickel ammonium sulfate solutions were measured at 25°C. with an Abbé refractometer. The instrument was capable of measuring refractive index with a precision of ± 0.00005 (Table II).

Densities were determined by the density bottle method, as described by Findlay (1), over the temperature range 10° to 35°C. The precision of these measurements (1) is estimated to be $\pm 0.1\%$.

Viscosities of the solutions were measured with an Ostwald viscometer, as described by Findlay (1), over the temperature range 10° to 35°C. The estimated precision is $\pm 0.3\%$. The density and viscosity results are recorded in Table III.

The solubility of nickel ammonium sulfate in water was measured over the temperature range 10° to 49°C. by approaching equilibrium from the under- and oversaturated states. The solutions contained in stoppered flasks were immersed in a thermostatically controlled bath ($\pm 0.1^\circ\text{C}$.) and shaken at intervals over 48 hours. Samples were withdrawn through a filter and their concentrations determined by measuring the density at a temperature about 5° above the saturation temperature.

The results (Table IV) do not agree very well with those recorded in the handbooks (2, 5) or with the early work

of Tobler (6). It is, of course, difficult to explain differences between results from various sources without knowledge of the techniques used, but certain comments can be made. Two of the references (2, 5) are data handbooks, one of which (2) quotes no literature source. Neither gives any experimental detail. The work of Tobler (6) makes no mention of the purity of the salt or solvent, and saturation was approached only from the undersaturated state. The present investigation has shown that up to 48 hours is needed to approach equilibrium.

The detailed measurements of solubility in the range 37.5° to 49°C. were made in an attempt to detect any possible phase change. None was found in this region.

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Vapor-Liquid Equilibria of the Methane-Toluene System at Low Temperatures and High Pressures

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The system methane-toluene exhibits limited liquid-liquid miscibility at temperatures below the critical for methane. A quadruple point involving the two immiscible liquids, a solid phase, and a gas phase has been located visually at -149.7°F . and 358 p.s.i.a. Experimental data shown on the pressure-temperature projections permit one to classify the system as one with a Type K critical point and one with qualitatively similar behavior in all respects to the methane-heptane system. Experimental techniques were developed to sample and analyze with good precision the equilibrium gas phases containing as little as 0.001 mole % toluene. Thus, toluene K values as low as 2×10^{-5} can be reported with confidence. Binary K values for methane and toluene are reported at intervals of 20° from 0° to -100°F . for pressures from 100 to 3500 p.s.i.a. The results are reported numerically and illustrated graphically.

STUDIES on the low temperature phase behavior of the methane-*n*-paraffin binaries have appeared since the review of the vapor-liquid equilibria of light hydrocarbons by Price and Kobayashi (9). The equilibrium ratios and the low temperature phase behavior of the methane-*n*-hexane (8), methane-*n*-heptane (2, 6), methane-*n*-octane (7), methane-*n*-nonane (11), and methane-*n*-decane (12) systems have been investigated in the last few years. There are very few reports made on the study of methane-aromatic and methane-naphthenic systems up to the present time. Elbishlawi and Spencer (5) reported vapor-liquid equi-

librium ratios of methane-toluene and methane-benzene systems at 150°F. in 1951. Sabbina and Velikovski (10) reported some results for the methane-toluene system from 40° to 150°C. in 1956. Clark (4) studied methane-cyclohexane and methane-methylcyclohexane at 150°F. in 1949. The methane-cyclohexane system has also been reported by Stepanov and Vybornov (13) at temperatures from 32° to 140°F.

The objectives of this study were to: obtain vapor-liquid equilibria data on the methane-toluene system which could be related to the methane-*n*-heptane (2) and methane-methylcyclohexane systems (3) recently studied, all systems with a different solvent type of the same carbon number; and provide basic data on binary systems which would

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