

Solubility of 2,6-Diaminopyridine in Toluene, *o*-Xylene, Ethylbenzene, Methanol, Ethanol, 2-Propanol, and Sodium Hydroxide Solutions

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Solubilities are reported for 2,6-diaminopyridine dissolved at atmospheric pressure in toluene, *o*-xylene, and ethylbenzene from (273.25 to 361.25) K, methanol, ethanol, and 2-propanol between (266.15 and 321.25) K, and sodium hydroxide solution containing mass fraction of 0.000 % to 17.308 % sodium hydroxide over the temperature range of (273.55 to 326.35) K. Results of these measurements are fitted with the combined binary solvent Margules equation. For the systems studied, the Margules equation is found to provide very reasonable mathematical representation, with average deviations between experimental values and calculated ones being on the order of ± 2.2 % or less. The coupler parameters of the Margules equation (A_{21} , A_{12}) and the enthalpy of fusion ($\Delta_{\text{fus}}H$) of 2,6-diaminopyridine in different systems are obtained by regressing the experimental data.

Introduction

2,6-Diaminopyridine (CAS Registry No. 141-86-6) is a white to gray crystal. It is useful in the chemical industry. For example, it is an intermediate in the manufacturing of the analgesic phenazopyridine hydrochloride^{1,2} and a novel antibacterial dihydropyridone carboxylic acid derivative.³ It can also be used in the production of polyamides.⁴ Additionally, a hair dye that is synthesized by using 2,6-diaminopyridine as a coupler agent shows a good dyeing capacity.^{5,6} 2,6-Diaminopyridine is mainly prepared by the Chichibabin reaction, and the pure product is always obtained by extraction or crystallization,⁷ but only a limited solubility data base exists for 2,6-diaminopyridine.

In this work, the solubilities of 2,6-diaminopyridine in toluene, *o*-xylene, ethylbenzene, methanol, ethanol, and 2-propanol are measured by the analytical method, and those in sodium hydroxide–water solution are measured by the synthetic method. The experimental solubility data are correlated by a three-parameter model based on the Margules equation.

Experimental Section

Materials. 2,6-Diaminopyridine prepared in the laboratory is recrystallized prior to use.⁷ Its mass fraction purity determined by HPLC is better than 99.8 %. Its melting point temperature range is (394.25 to 395.15) K, and the value reported in the literature⁸ is 394.65 K. Toluene, *o*-xylene, ethylbenzene, methanol, ethanol, and 2-propanol (purchased from Shanghai Chemistry Reagent Co., China) are in chromatographically pure grade. They are stored over molecular sieves to remove trace water shortly before use. The mass fraction purities of these solvents are higher than 99.9 % (GC). Karl Fischer titration gave a water mass fraction to be less than 0.02 %. Bidistilled water is used. Sodium hydroxide is of analytical reagent grade (purchased from Shanghai Chemistry Reagent Co.).

Experimental Method. The solubility of a solid in a solvent can be measured by the analytical method^{9,10} or the synthetic method.^{11,12} Both of these methods are widely used, and they are reliable. The advantage of the analytical method lies in the possibility of measuring a large number of samples simultaneously. The disadvantage is that it is tedious and time-consuming.¹³ In this work the analytical method is used to determine the solubility of 2,6-diaminopyridine in toluene, *o*-xylene, ethylbenzene, methanol, ethanol, and 2-propanol. The concentrations of 2,6-diaminopyridine in the above solvents are analyzed by HPLC. HPLC is not applied to a solution with a high pH, so the solubility of 2,6-diaminopyridine in sodium hydroxide solution is determined by the synthetic method. The solubility data are obtained by judging the disappearance of the particles suspended in the solution. The disappearance of the solid phase can be achieved either by adjusting the temperature or by adding a known mass of solvent.^{14,15}

Analytical Method Procedure. The apparatus and the procedure are similar to the case described in the literature.¹⁶ A mercury-in-glass thermometer is inserted through the hole in the stopper for the measurement of the temperature of the flask. The thermometer had a measurement range from (263.15 to 373.15) K with an uncertainty of ± 0.05 K. Mixtures are prepared by mass using a Mettler H542 balance. The balance had a range of measurement up to 160 g, with an uncertainty of ± 0.00001 g. The estimated error in the mole fraction is less than 0.00001.

An excess amount of solute is added to the solvents in a specially designed sealed dual-wall flask. Between the outer and inner walls of the flask, ethylene glycol is circulated. The circulating ethylene glycol is controlled by a thermostat at a designed temperature within ± 0.05 K. To make it attain equilibrium, the solution is constantly stirred for 2 h with a magnet stirrer, and then the stirring is stopped to let the solution settle for 1 h. A clear liquid ((2 to 3) cm³) is taken into the sampling vial and weighed using the Mettler H542 balance.

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Table 1. Experimentally Determined Solubility Data of 2,6-Diaminopyridine (2) in Toluene (1), *o*-Xylene (1), and Ethylbenzene (1)

T	x_2	$(x_2 - x_{2cal})$	T	x_2	$(x_2 - x_{2cal})$
K	x_2	x_2	K	x_2	x_2
Toluene					
273.25	0.00149	-0.0334	321.25	0.00942	-0.0753
277.15	0.00191	0.0407	325.55	0.01242	0.0542
281.25	0.00214	-0.0231	329.45	0.01270	-0.0559
285.55	0.00270	0.0275	333.35	0.01619	0.0557
289.45	0.00304	-0.0143	337.25	0.01645	-0.0582
293.65	0.00375	0.0265	341.15	0.02072	0.0442
297.55	0.00417	-0.0204	345.65	0.02204	-0.0420
301.45	0.00510	0.0308	349.55	0.02768	0.0568
305.35	0.00558	-0.0258	353.45	0.02825	-0.0511
309.55	0.00704	0.0508	357.35	0.03550	0.0479
313.45	0.00733	-0.0497	361.25	0.03671	-0.0498
317.35	0.00919	0.0383			
<i>o</i> -Xylene					
273.35	0.00128	0.0354	321.25	0.00838	0.0340
277.15	0.00140	-0.0434	325.95	0.00937	-0.0192
282.25	0.00185	0.0169	329.45	0.01104	0.0225
285.55	0.00201	-0.0389	333.35	0.01208	-0.0230
289.45	0.00252	0.0276	336.95	0.01439	0.0268
293.65	0.00283	-0.0246	341.15	0.01534	-0.0568
297.55	0.00351	0.0375	346.25	0.01942	0.0012
302.25	0.00395	-0.0243	349.55	0.02264	0.0360
305.35	0.00469	0.0305	353.45	0.02401	-0.0481
309.55	0.00517	-0.0276	357.05	0.02922	0.0137
314.15	0.00640	0.0184	361.05	0.03403	0.0085
317.35	0.00695	-0.0140			
Ethylbenzene					
273.25	0.00022	0.0117	321.25	0.00150	0.0368
277.15	0.00026	0.0101	326.15	0.00164	-0.0411
282.25	0.00033	0.0082	329.45	0.00197	0.0257
285.55	0.00035	-0.0490	333.35	0.00212	-0.0342
289.45	0.00046	0.0489	336.95	0.00252	0.0178
293.05	0.00049	-0.0364	341.05	0.00288	0.0143
298.65	0.00066	0.0479	346.25	0.00330	-0.0253
302.25	0.00069	-0.0416	349.55	0.00385	0.0164
305.35	0.00086	0.0585	353.05	0.00430	0.0070
309.55	0.00090	-0.0562	357.05	0.00474	-0.0343
313.15	0.00113	0.0428	361.15	0.00577	0.0167
317.35	0.0012	-0.0441			

When the temperature is higher than room temperature, more solvent weighed with the balance is added into the sampling vial to avoid recrystallization of solute, which will tend to a 2-fold increase in the error of the mole fraction of solute (<0.00002). By repeating the above procedure at different temperatures, a series of samples are obtained. The analyses are carried out by HPLC at 254 nm.¹⁷ The related calibration curve is prepared by using the standard solution in the appropriate concentration range. On the basis of the calibration curve and diluted multiple, the real concentrations of samples at different temperatures can be obtained.

Synthetic Method Procedure. A laser monitoring system with a laser generator, a photoelectric transformer, and a recorder are used to determine the dissolving temperature of a solid-liquid mixture of known composition. Besides the monitoring system, the apparatus is the same as above. The procedure is similar to the case described in the literature.¹⁸ The solution of known composition is prepared by using a Mettler H542 balance. The contents of the vessel are heated very slowly at a rate of about $0.6 \text{ K}\cdot\text{h}^{-1}$. The dissolution of solute is examined by the laser beam penetrating the vessel. As soon as the solute disappeared, the temperature is recorded. More solute is added into the flask, and another solution known composition is obtained. Repeating the procedure above, a corresponding dissolving temperature can be recorded again. The total amount

Table 2. Experimentally Determined Solubility Data of 2,6-Diaminopyridine (2) in Methanol (1), Ethanol (1), and 2-Propanol (1)

T	x_2	$(x_2 - x_{2cal})$	T	x_2	$(x_2 - x_{2cal})$
K	x_2	x_2	K	x_2	x_2
Methanol					
266.15	0.05984	0.0154	293.35	0.13135	-0.0250
269.95	0.06600	-0.0192	297.15	0.14964	0.0096
274.15	0.07786	0.0068	301.15	0.16484	0.0095
278.05	0.08883	0.0154	304.55	0.17823	0.0084
281.15	0.09331	-0.0295	307.95	0.18758	-0.0173
285.95	0.11146	0.0100	311.15	0.20739	0.0133
290.15	0.12519	0.0112	317.85	0.23324	-0.0094
Ethanol					
266.95	0.07515	0.0048	297.15	0.20355	0.0139
270.35	0.08533	0.0013	301.15	0.22557	0.0131
274.15	0.09924	0.0124	304.55	0.23644	-0.0235
277.45	0.10832	-0.0165	307.95	0.26611	0.0153
281.15	0.12261	-0.0176	312.05	0.28172	-0.0188
286.35	0.14793	0.0045	316.25	0.31865	0.0163
290.15	0.16553	0.0027	321.25	0.34208	-0.0111
293.55	0.18231	0.0019			
2-Propanol					
266.25	0.02221	-0.0231	297.25	0.07325	-0.0339
269.95	0.02796	0.0511	301.15	0.08868	0.0189
274.15	0.03024	-0.0418	305.15	0.0976	-0.0253
277.55	0.03793	0.0485	308.25	0.11392	0.0225
281.15	0.04009	-0.0366	311.15	0.12011	-0.0234
285.65	0.05116	0.0347	315.85	0.14734	0.0234
290.15	0.05648	-0.0351	319.25	0.15935	-0.010
293.65	0.06799	0.0221			

of solute added is recorded, and the solubility expressed in mole fraction is calculated.

Solubility Model. For a solid-liquid system, the effect of pressure and heat capacity difference on the solubility can be neglected, so the solubility model is simplified as¹⁹

$$\ln \gamma_2 x_2 = \frac{\Delta_{\text{fus}} H}{R} \left(\frac{1}{T_m} - \frac{1}{T} \right) \quad (1)$$

where γ_2 is the liquid-phase activity coefficient of solute, x_2 is the mole fraction of solute, $\Delta_{\text{fus}} H$ is the enthalpy of fusion of solute, and T_m is the melting point temperature of solute. In this work, the Margules equation is used to describe the relationship between the activity coefficient γ_2 and the mole fraction x_2 ¹⁹

$$\ln \gamma_2 = (1 - x_2)^2 [A_{21} + 2(A_{12} - A_{21})x_2] \quad (2)$$

where A_{21} and A_{12} are the coupler parameters of Margules equation and A_{21} is related to the infinite-dilution activity coefficient (γ_2^∞) of solute by the expression as

$$A_{21} = \ln \gamma_2^\infty$$

By incorporating eqs 1 and 2, the linear equation can be obtained:

$$\frac{1}{T} = -\frac{R \ln x_2}{\Delta_{\text{fus}} H} - \frac{R A_{21} (1 - x_2)^2}{\Delta_{\text{fus}} H} - \frac{2R(A_{12} - A_{21})}{\Delta_{\text{fus}} H} (1 - x_2)^2 x_2 + \frac{1}{T_m} \quad (3)$$

The linear least-squares method is used to regress the experimental solubility data with the above equation to estimate the parameters A_{21} , A_{12} , and $\Delta_{\text{fus}} H$.

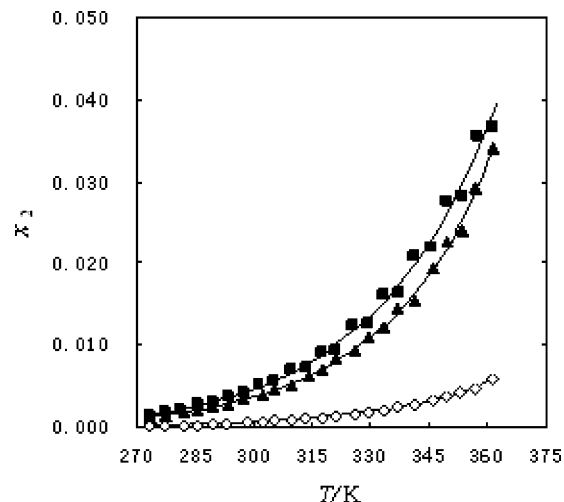
Table 3. Experimentally Determined Solubility Data of 2,6-Diaminopyridine (2) in Sodium Hydroxide (w) + Water ($1 - w$) Solutions with $w =$ Mass Fraction

T K	x_2	$(x_2 - x_{2cal})$ x_2	T K	m_2	$(x_2 - x_{2cal})$ x_2
$w = 0.000\%$					
273.65	0.01030	0.0309	300.95	0.03169	-0.0346
277.15	0.01114	-0.0437	304.35	0.03930	0.0230
280.55	0.01378	0.0226	307.75	0.0439	-0.0316
283.95	0.01509	-0.0332	311.15	0.05521	0.0220
287.15	0.01849	0.0325	314.55	0.06408	-0.0215
290.55	0.02131	0.0280	317.95	0.08305	0.0168
294.15	0.02312	-0.0476	321.35	0.10912	0.0114
297.55	0.02903	0.0309	324.75	0.16531	-0.0211
$w = 2.814\%$					
273.65	0.00733	0.0169	301.05	0.02373	-0.0102
277.15	0.0083	-0.0144	304.35	0.02809	0.0085
280.45	0.00962	-0.0121	307.75	0.03297	0.0091
283.95	0.01146	0.0101	310.65	0.03735	-0.008
287.15	0.01316	0.0093	314.55	0.04644	0.0064
290.55	0.01499	-0.0081	317.95	0.05585	-0.0057
294.15	0.01743	-0.014	321.55	0.07205	0.0044
298.15	0.02131	0.0114	324.75	0.09553	-0.0056
$w = 5.914\%$					
273.65	0.00542	-0.0650	301.05	0.01909	-0.0028
277.15	0.00695	0.0290	304.35	0.02205	-0.0064
280.35	0.00795	0.0224	307.75	0.02568	-0.0102
283.95	0.00926	0.0176	310.85	0.02966	-0.0137
287.15	0.0106	0.0138	314.55	0.03558	-0.0179
290.55	0.01223	0.0097	317.85	0.04295	-0.0098
294.15	0.01423	0.0053	321.15	0.05284	-0.0059
297.35	0.0163	0.0019	326.15	0.08494	0.0454
$w = 9.217\%$					
273.55	0.00354	0.0081	300.85	0.01072	0.0059
277.15	0.00412	0.0073	304.35	0.01221	-0.0026
280.35	0.00467	-0.0009	307.75	0.01409	0.0051
283.95	0.00525	-0.0325	311.15	0.01609	0.001
287.15	0.00621	0.0059	314.55	0.01858	0.0057
290.55	0.00708	0.0002	317.85	0.02106	-0.0074
294.15	0.00832	0.0178	321.85	0.02506	-0.008
297.35	0.00917	-0.0118	326.15	0.03103	0.0052
$w = 12.455\%$					
273.55	0.00176	0.0091	300.85	0.00506	0.0122
277.15	0.00200	-0.0119	304.35	0.00562	-0.0101
280.25	0.00231	0.0072	307.75	0.00648	0.0100
283.95	0.00261	-0.0165	311.15	0.00717	-0.0103
287.15	0.00304	0.0124	314.55	0.00826	0.0106
290.55	0.00337	-0.0123	317.85	0.00911	-0.0078
294.15	0.00395	0.0100	321.55	0.01058	0.0104
297.85	0.00442	-0.0134	326.35	0.01235	-0.0073
$w = 17.308\%$					
273.75	0.00054	0.0205	301.05	0.00147	0.01520
277.15	0.00058	-0.0431	304.35	0.00166	0.0243
279.85	0.00069	0.0264	307.75	0.00178	-0.0198
283.65	0.00079	0.0176	311.45	0.00209	0.0185
287.15	0.00086	-0.0279	314.55	0.00221	-0.0270
290.55	0.00102	0.0189	318.05	0.00259	0.0188
294.15	0.00112	-0.0164	321.65	0.00278	-0.0256
297.25	0.00125	-0.0156	325.05	0.00321	0.0107

A sodium hydroxide solution with a constant concentration can be regarded as a unitary solvent, so the solubilities for all systems of this work can be regressed by eq 3.

Results and Discussion

The solubilities of 2,6-diaminopyridine in toluene, *o*-xylene, ethylbenzene, methanol, ethanol, and 2-propanol are measured by the analytical method in the temperature range of (266.15 to 361.25) K, and the data are shown in Tables 1 and 2. The synthetic method is applied to measure the solubilities of 2,6-diaminopyridine in sodium hydroxide solutions containing mass fractions of 0.000 % to 17.308 % sodium hydroxide over the

**Figure 1.** Solubility of 2,6-diaminopyridine (2) in aromatic hydrocarbons: ■, toluene; ▲, *o*-xylene; ◇, ethylbenzene; ○, calculated values.**Table 4. Margules Equation Fitting Parameters of 2,6-Diaminopyridine in Toluene, *o*-Xylene, Ethylbenzene, Methanol, Ethanol, and 2-Propanol**

solvent	$\frac{\Delta_{fus}H}{\text{kJ}\cdot\text{mol}^{-1}}$	A_{21}	A_{12}	$10^3\sigma$	R^2
toluene	27.77	2.727	2.068	0.837	0.9979
<i>o</i> -xylene	27.40	3.006	-0.087	0.390	0.9990
ethylbenzene	27.75	4.685	-17.16	0.058	0.9985
methanol	25.01	-1.052	-0.255	1.997	0.9986
ethanol	25.43	-1.203	-1.860	3.099	0.9990
2-propanol	24.28	0.245	-0.167	2.045	0.9971

Table 5. Margules Equation Fitting Parameters of 2,6-Diaminopyridine in Sodium Hydroxide (w) + Water ($1 - w$) Solutions with $w =$ Mass Fraction

100 w	$\frac{\Delta_{fus}H}{\text{kJ}\cdot\text{mol}^{-1}}$	A_{21}	A_{12}	$10^3\sigma$	R^2
0.000	24.77	1.363	-2.012	1.284	0.9981
2.814	25.75	1.540	-2.311	4.018	0.9997
5.194	26.04	1.716	-2.690	1.002	0.9988
9.217	25.22	2.300	-2.824	0.105	0.9996
12.455	24.91	3.023	-3.154	0.061	0.9996
17.308	24.38	4.283	-10.74	0.035	0.9982

temperature range of (273.55 to 326.35) K, and the data are shown in Table 3.

Equation 3 is used to regress the experimental data in Tables 1, 2, and 3, and the parameters ($\Delta_{fus}H$, A_{21} , A_{12}) in eq 3 are obtained and shown in Tables 4 and 5, respectively. The values of the root-mean-square deviations (rmsd) and the correlation coefficients R^2 are also listed in Tables 4 and 5. The rmsd of the mole fraction is defined as

$$\sigma = \sqrt{\frac{\sum_{i=1}^N (x_{2cali} - x_{2i})^2}{N}}$$

where N is the number of experimental points, x_{2cali} represents the solubility calculated from eq 3, and x_{2i} represents the experimental solubility value. The comparison between experimental data and calculated ones are shown in Tables 1, 2, and 3 and Figures 1, 2, 3, and 4. The average deviation between experimental values and calculated ones is 2.2 %. It can be seen that the solubility model can be used to describe the variation with temperature of the solubility of solute.

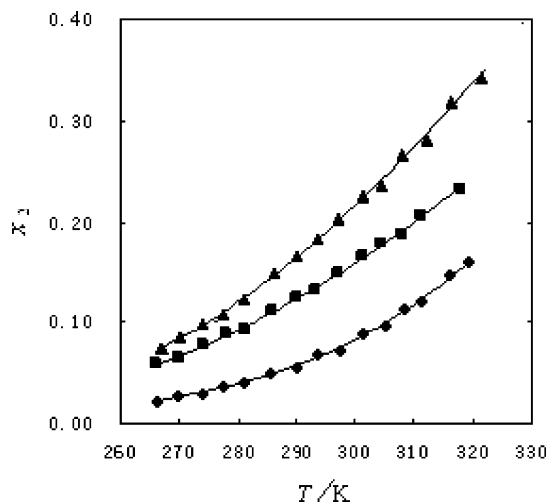


Figure 2. Solubility of 2,6-diaminopyridine (2) in alcohols: ■, methanol; ▲, ethanol; ◆, 2-propanol; —, calculated values.

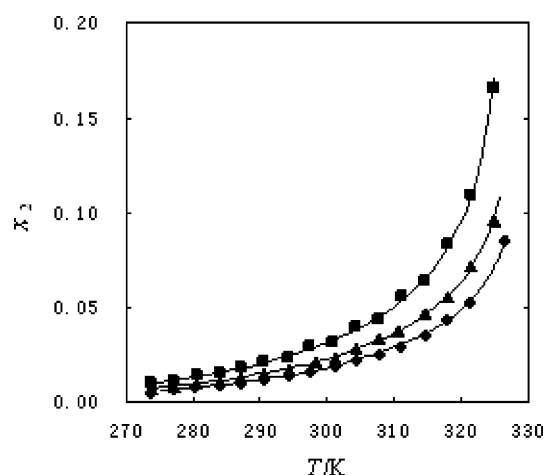


Figure 3. Solubility of 2,6-diaminopyridine (2) in sodium hydroxide solutions with w = mass fraction: ■, $w = 0.000\%$; ▲, $w = 2.814\%$; ◆, $w = 5.194\%$; —, calculated values.

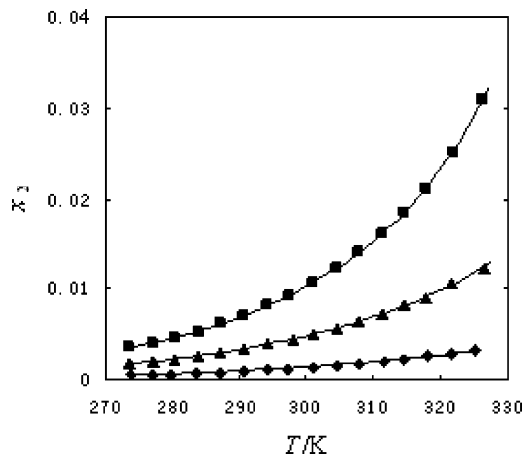


Figure 4. Solubility of 2,6-diaminopyridine (2) in sodium hydroxide solutions with w = mass fraction: ■, $w = 9.217\%$; ▲, $w = 12.455\%$; ◆, $w = 17.308\%$; —, calculated values.

According to the data shown in Tables 4 and 5, it is clear that for every system the greater the solubility of 2,6-diaminopyridine is, the smaller is the γ_2^∞ , which is accord with the thermodynamics.

The solubility curves of 2,6-diaminopyridine in toluene, *o*-xylene, and ethylbenzene, and also in methanol, ethanol, and

Table 6. Interpolating Prediction of Solubility of 2,6-Diaminopyridine in Sodium Hydroxide (w) + Water ($1 - w$) Solutions by Empirical Model with w = Mass Fraction

100 w	T		x_2	$x_{2\text{pred}}$	$(x_2 - x_{2\text{pred}})$
	K				
1.015	288.15		0.01629	0.01684	-0.0338
2.023	288.15		0.01534	0.01593	-0.0385
2.023	300.15		0.02610	0.02690	-0.0307
3.059	288.15		0.01446	0.01480	-0.0235
3.059	300.15		0.02438	0.02497	-0.0242
7.025	300.15		0.01592	0.01518	0.0465
7.025	314.55		0.03017	0.02855	0.0537
13.067	325.05		0.00975	0.01029	-0.0554
15.061	300.15		0.00237	0.00249	-0.0506

2-propanol, are shown in Figures 1 and 2, respectively. It is seen that the solubilities of 2,6-diaminopyridine in aromatic hydrocarbons decreased in the sequence toluene, *o*-xylene, ethylbenzene and those in alcohols decreased in the sequence ethanol, methanol, 2-propanol. On the other hand, the solubilities of 2,6-diaminopyridine in the alcohols are much larger than those in the aromatic hydrocarbons.

The solubility curves of 2,6-diaminopyridine in sodium hydroxide solutions with mass fraction of (0.000 %, 2.814 %, 5.194 %) and (9.217 %, 12.455 %, 17.308 %) are shown in Figures 3 and 4, respectively. It is obvious that the solubility of 2,6-diaminopyridine increases with increase in temperature and decreases sharply with the increase of the mass fraction of sodium hydroxide. Furthermore, the smaller the mass fraction of sodium hydroxide is, the more remarkable is the influence of temperature. For instance, at the mass fraction of 2.814 % the solubility increases 190 % when the temperature changes from (307.75 to 324.75) K; however, at the mass fraction of 17.308 % the solubility increases 80 % when the temperature changes from (307.75 to 325.05) K.

The enthalpies of fusion ($\Delta_{\text{fus}}H$) of 2,6-diaminopyridine regressed from the data of sodium hydroxide solutions with different mass fraction are consistent, but the coupler parameters of the Margules equation (A_{21} , A_{12}) are a function of the mass fraction (w). If A_{21} and A_{12} are expressed as a function of w by polynomials, eqs 4 and 5 can be obtained as follows:

$$A_{21} = 80.575w^2 + 2.9617w + 1.3673 \quad (4)$$

$$A_{12} = -67764w^4 + 16189w^3 - 1124.9w^2 + 10.688w - 2.0167 \quad (5)$$

Equations 4 and 5 can be used to estimate A_{21} and A_{12} for the sodium hydroxide solution at any mass fraction from 0.000 % to 17.308 %.

The enthalpy of fusion ($\Delta_{\text{fus}}H$) of 2,6-diaminopyridine in sodium hydroxide solution is calculated as

$$\Delta_{\text{fus}}H = \Delta_{\text{fus}}H_m = \frac{\sum_1^6 \Delta_{\text{fus}}H_i}{6} \quad (6)$$

where $\Delta_{\text{fus}}H_i$ (shown in Table 5) is the enthalpy of fusion of 2,6-diaminopyridine regressed in different mass fractions of sodium hydroxide solution.

By substituting eq 4 to eq 6 for A_{21} , A_{12} , and $\Delta_{\text{fus}}H$ in eq 3, respectively, the solubilities of 2,6-diaminopyridine in sodium hydroxide solution can be predicted at any temperature from (273.55 to 326.35) K. Some solubility data predicted by

interpolation are listed and compared with experimental results in Table 6. It can be seen that the precision of the interpolation calculation for the Margules equation model can meet the demand of the application to some degree.

Conclusions

Solubilities are reported for 2,6-diaminopyridine in toluene, *o*-xylene, and ethylbenzene from (273.25 to 361.25) K, in methanol, ethanol, and 2-propanol between (266.15 and 321.25) K, and in sodium hydroxide solution containing mass fraction of 0.000 % to 17.308 % sodium hydroxide over the temperature range of (273.55 to 326.35) K. The Margules equation is used to fit the experiment data, and the average deviations between experimental values and calculated ones are on the order of ± 2.2 % or less. Especially, the parameters (A_{21} , A_{12}) for sodium hydroxide solution are expressed as a function of w by polynomials, and the solubility of 2,6-diaminopyridine in the solution at any mass fraction can be predicted by interpolation.

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