Solubility of 1-*H*-Tetrazole-1-acetic Acid in Different Solvents between 283 K and 323 K

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The solubility of 1-*H*-tetrazole-1-acetic acid in ethanol, water, methanol, 1-propanol, ethyl acetate, 2-propanol, acetone, and butyl acetate between 283 K and 323 K was measured using a laser monitoring observation technique. Results of these measurements were correlated with a semiempirical equation. For the eight solvents studied, the data are well fitted with a semiempirical equation.

Introduction

1-*H*-Tetrazole-1-acetic acid (CAS Registry No. 21732-17-2) is a white or almost white crystalline powder and a useful chemical for an intermediate for the synthesis of corrosion inhibitors, medicines, and agrochemicals. To determine the proper solvent and to design an optimized crystallization process, it is necessary to know its solubility in different solvents. In this paper, the solubility of 1-*H*-tetrazole-1-acetic acid in ethanol, water, methanol, 1-propanol, ethyl acetate, 2-propanol, acetone, and butyl acetate between 283 K and 323 K was measured using a laser monitoring observation technique at atmospheric pressure. The method employed in this work was classed as a synthetic method, which was much faster and more reliable than the analytical method.[1]

Experimental Section[1]

Materials. 1-*H*-Tetrazole-1-acetic acid with a mass purity of 0.991 was purchased from Zhe Jiang Lianhe Chemical Technology Co., Ltd. Other reagents are analytical research grade reagents from Shijiazhuang Chemical Reagent Co.

Apparatus and Procedure. The solubility of 1-*H*-tetrazole-1-acetic acid was measured using an apparatus similar to that described in the literature[2−6] and described briefly here. A 300 mL jacketed vessel was used to determine the solubility. The temperature in the vessel was maintained at the desired value by continuous forced water circulation from a water bath with a thermoelectric controller (type 501, China) (temperature uncertainty of ± 0.05 K). A mercury in-glass thermometer (uncertainty of ± 0.05 K) was used for the measurement of the temperature in the vessel. The dissolution of the solute was examined by the laser beam penetrating the vessel. To prevent the evaporation of the solvent, a condenser vessel was introduced. The masses of the samples and solvents were determined using an analytical balance (Sartorius CP124S, Germany) with an uncertainty of ± 0.1 mg.

Predetermined excess amounts of solvent and 1-*H*-tetrazole-1-acetic acid of known mass were placed in the inner chamber of the vessel. The contents of the vessel were stirred continuously at the required temperature. In the early stage of the experiment, the laser beam was decreased by the undissolved particles of 1-*H*-tetrazole-1-acetic acid in the solution. As the particles of the solute dissolved, the intensity of the laser beam increased gradually. When the solute dissolved completely, the solution was clear, and the laser intensity reached maximum. Then, additional solute of known mass (about 0.5 mg to 3 mg) was introduced into the vessel. This procedure was repeated until the penetrated laser intensity could not return a maximum, or in other words, the last addition of solute could not dissolve completely. The interval of addition was 90 min. The total amount of the solute consumed was recorded. The same solubility experiment was conducted three times, and the mean values were used to calculate the mole fraction solubility x_1 based on eq 1.

$$x_1 = \frac{m_1/M_1}{m_1/M_1 + m_2/M_2} \tag{1}$$

where m_1, m_2 represent the mass of the solute and solvent, respectively, and M_1, M_2 are the molecular weight of the solute and solvent, respectively.

Results and Discussion

The solubility data of 1-*H*-tetrazole-1-acetic acid in ethanol, water, methanol, 1-propanol, ethyl acetate, 2-propanol, acetone, and butyl acetate between 283 K and 323 K are presented in Table 1. The temperature dependence of 1-*H*-tetrazole-1-acetic acid solubility in pure solvents is described by the modified Apelblat equation which is a semiempirical equation[7−9]

$$\ln x_1 = A + \frac{B}{T/\mathrm{K}} + C \ln(T/\mathrm{K}) \tag{2}$$

where x_1 is the mole fraction solubility of 1-*H*-tetrazole-1-acetic acid; T is the absolute temperature; and A, B, and C are the dimensionless parameters. The calculated solubility values of 1-*H*-tetrazole-1-acetic acid are also given in Table 1. The values of parameters A, B, and C and the root-mean-square deviations (rmsd's) are listed in Table 2. The rmsd is defined as

$$\mathrm{rmsd} = \left[\frac{\sum_{j=1}^{N} (x_{1,j} - x_{1,j}^{\mathrm{calcd}})^2}{N-1}\right]^{1/2} \tag{3}$$

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10.1021/je700200b CCC: $37.00
Published on Web 08/03/2007

Table 1. Mole Fraction Solubility of 1-*H*-Tetrazole-1-acetic Acid in Pure Solvents

T/K	$100x_1$	$100(x_1 - x_1^{calcd})$	T/K	$100x_1$	$100(x_1 - x_1^{calcd})$
		Ethanol			
283.27	5.173	−0.047	308.17	12.91	−0.01
288.17	6.205	−0.001	313.23	15.65	0.01
293.46	7.504	−0.001	318.21	18.93	0.02
298.22	8.914	−0.015	323.26	22.91	−0.05
303.31	10.76	−0.02			
		Water			
283.17	0.7291	0.0143	308.30	5.49	0.205
288.31	1.089	0.024	313.17	8.257	0.374
293.28	1.597	0.022	318.22	12.19	0.22
298.24	2.337	−0.003	323.16	17.26	−0.8
303.19	3.483	−0.004			
		Methanol			
288.18	14.55	0.61	308.24	23.33	−0.29
293.29	15.55	−0.10	313.34	27.64	−0.16
298.23	17.32	−0.40	318.22	32.55	−0.25
303.22	19.91	−0.42	323.19	38.34	−0.82
		1-Propanol			
288.27	3.119	−0.204	308.33	6.801	−0.011
293.21	3.862	−0.009	313.18	8.389	−0.008
298.23	4.636	0.038	318.22	10.59	0.02
303.29	5.573	0.016	323.32	13.54	0.04
		Ethyl Acetate			
288.27	1.208	−0.036	308.22	1.934	0.000
293.28	1.377	−0.004	313.26	2.177	−0.006
298.19	1.544	0.007	318.16	2.460	−0.004
303.31	1.731	0.006	323.21	2.810	0.010
		2-Propanol			
288.17	2.819	0.032	308.21	7.936	0.006
293.22	3.745	0.002	313.29	9.846	0.003
298.26	4.913	−0.002	318.24	11.95	0.000
303.20	6.295	0.003	323.27	14.32	0.01
		Acetone			
288.23	9.645	0.025	308.18	13.63	−0.01
293.32	10.54	0.01	313.26	14.86	0.00
298.24	11.48	−0.01	318.27	16.16	0.00
303.31	12.54	0.00	323.23	17.52	−0.01
		Butyl Acetate			
288.22	0.4435	0.0278	308.22	0.7733	0.0001
293.30	0.4834	0.0034	313.30	0.9309	0.0064
298.33	0.5523	−0.0066	318.24	1.113	0.005
303.18	0.6465	−0.0062	323.28	1.327	−0.014

Table 2. Parameters of Equation 2 for Solubility of 1-*H*-Tetrazole-1-acetic Acid in Pure Solvents

solvent	A	B	C	10^4 rmsd
ethanol	−195.49	5819	30.461	2.72
water	−405.88	11838	63.608	33.01
methanol	−386.89	15151	58.681	44.20
1-propanol	−512.19	19953	77.609	7.55
ethyl acetate	−185.12	6379.8	28.002	1.49
2-propanol	288.64	−16919	−41.23	1.30
acetone	−39.014	318.121	6.2801	1.21
butyl acetate	−331.59	12178	50.118	1.27

where N is the number of experimental points; $x_{1,j}^{calcd}$ represents the solubility calculated from eq 2; and $x_{1,j}$ represents the experimental solubility values.

From data listed in Table 1 and Table 2, we can draw the following conclusions: (1) The solubility of 1-*H*-tetrazole-1-acetic acid increases with temperature in the eight solvents. The solubility of 1-*H*-tetrazole-1-acetic acid in butyl acetate is the lowest, and the solubility is largest in methanol. (2) The solubility of 1-*H*-tetrazole-1-acetic acid in butyl acetate is the lowest in eight solvents, so butyl acetate may be used for dilution to increase the yield of the product in the crystallization process of 1-*H*-tetrazole-1-acetic acid. (3) From Table 1 and Table 2, we can see that the experimental solubility and correlation equation in this work can be used as essential data and models in the purification process of 1-*H*-tetrazole-1-acetic acid. The calculated solubility shows good agreement with the experimental values.

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Received for review April 16, 2007. Accepted June 13, 2007.

JE700200B