Measurement and Correlation of Solubility of Xylitol in Binary Ethanol + Acetone Solvent Mixtures with the Combined Nearly Ideal Binary Solvent/ Redlich−Kister Equation

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The solubilities of xylitol in binary ethanol + acetone solvent mixtures were measured using a laser technique with the temperature range from 293 K to 323 K at atmospheric pressure. Results of these measurements were correlated by the combined nearly ideal binary solvent (CNIBS)/Redlich−Kister equation. The experimental solubility and correlation equation in this work can be used as essential data and as a model in the purification process of xylitol.

1. Introduction

Xylitol, a kind of white crystal, is an important sweetener which is one kind of sugar substitute. The main branches of xylitol use are food production, perfumery, pharmaceutics, and chemistry.[1−3] In industrial manufacture, xylitol is crystallized from solution as the final step. So, crystallization is a key step because, in many respects, it determines the yield and quality of the target product. To determine proper solvents and to design an optimized production process, it is necessary to know the solubility of xylitol in different solvents. However, from a review of the literature, it was found that no experimental solubility data in binary ethanol + acetone solvent mixtures were available. In this paper, the solubility of xylitol in binary ethanol + acetone solvent mixtures was experimentally determined in the temperature range from 293 K to 323 K. The method employed in this work was classified as a synthetic method, which was much faster and more credible than the analytical method.[4]

2. Experimental Section

Materials. A crystal of xylitol, with a melting/decomposition point of 94.5 (± 0.5) °C, was prepared by recrystallization from aqueous solution. Its purity is higher than 99.9 mass %. It was dried in a vacuum at 50 °C for 48 h and stored in a desiccator. All solvents used for experiments were of analytical reagent grade. It was dehydrated with molecular sieves, and its purity was higher than 99.8 mass % checked by gas chromatography.

Apparatus and Procedures. The solubility of xylitol was measured using an apparatus similar to that described in the literature[5] and described briefly here. A 500 mL jacketed vessel was used to determine the solubility, and the temperature was controlled to be constant (fluctuates within 0.05 K) through a thermostat water bath. 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 weighted using an analytical balance (Sartorius CP124S, Germany) with an uncertainty of ± 0.0001 g.

The solubility of xylitol was determined by using a laser technique.[4−9] During experiments, the fluid in the glass vessel was monitored by a laser beam. Predetermined excess amounts of solvent and xylitol of known mass were placed in the inner chamber of the vessel. The contents of the vessel were stirred continuously at a required temperature. In the early stage of the experiment, the laser beam was blocked by the undissolved particles of xylitol in the solution, so the intensity of the laser beam penetrating the vessel was lower. Along with the dissolution of the particles of the solute, the intensity of the laser beam increased gradually. When the solute dissolved completely, the solution was clear and transparent, and the laser intensity reached maximum. Then additional solute of known mass {about (0.5 to 3) mg} was introduced into the vessel. This procedure was repeated until the penetrated laser intensity could not return to maximum or, in other words, the last addition of solute could not dissolve completely. The interval of addition was 60 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_A) based on eq 1. The composition of the solvent mixture (x_B^0) was defined by eq 2

$$x_A = \frac{m_1/M_1}{m_1/M_1 + m_2/M_2 + m_3/M_3} \quad (1)$$

$$x_B^0 = \frac{m_2/M_2}{m_2/M_2 + m_3/M_3} \quad (2)$$

where m_1, m_2, and m_3 represent the mass of the solute, ethanol, and acetone, respectively, and M_1, M_2, and M_3 are the molecular weights of the solute, ethanol, and acetone, respectively.

3. Results and Discussion

The solubility data of xylitol in binary ethanol + acetone solvent mixtures in the temperature range from 293 K to 323 K are presented in Table 1. The solubility data in binary ethanol + acetone solvent mixtures are described by the combined nearly ideal binary solvent (CNIBS)/Redlich−Kister model.

Acree and co-workers[10−12] suggested the CNIBS/Redlich−Kister model

$$\ln x_A = x_B^0 \ln(x_A)_B + x_C^0 \ln(x_A)_C + x_B^0 x_C^0 \sum_{i=0}^{N} S_i (x_B^0 - x_C^0)^i \quad (3)$$

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Table 1. Experimental Solubility (x_1) and Calculated Values of Xylitol of Equation 5

x_2^0	$10^3 x_1^{exptl}$	$10^3 x_1^{calcd}$	RE = $[(x_1^{exptl} - x_1^{calcd})/x_1^{exptl}]\cdot 100$	x_2^0	$10^3 x_1^{exptl}$	$10^3 x_1^{calcd}$	RE = $[(x_1^{exptl} - x_1^{calcd})/x_1^{exptl}]\cdot 100$
			$T = 293.15$ K				
1.0000	1.716	1.691	1.46	0.4567	0.8820	0.8873	−0.60
0.9190	1.707	1.756	−2.87	0.3508	0.6753	0.6728	0.37
0.8345	1.698	1.706	−0.47	0.2396	0.4878	0.4854	0.49
0.7463	1.584	1.561	1.45	0.1229	0.3290	0.3285	0.15
0.6541	1.405	1.354	3.63	0.0000	0.2018	0.2022	−0.20
0.5577	1.081	1.120	−3.61				
			$T = 298.15$ K				
1.0000	2.402	2.368	1.42	0.4567	1.220	1.228	−0.66
0.9190	2.383	2.430	−1.97	0.3508	0.9185	0.9278	−1.01
0.8345	2.300	2.345	−1.96	0.2396	0.6476	0.6578	−1.58
0.7463	2.179	2.142	1.70	0.1229	0.4403	0.4269	3.04
0.6541	1.910	1.862	2.51	0.0000	0.2398	0.2427	−1.21
0.5577	1.5392	1.5470	−0.51				
			$T = 303.15$ K				
1.0000	3.088	3.035	1.72	0.4567	1.649	1.658	−0.55
0.9190	2.987	3.069	−2.75	0.3508	1.259	1.273	−1.11
0.8345	2.924	2.955	−1.06	0.2396	0.8643	0.9051	−4.72
0.7463	2.771	2.721	1.80	0.1229	0.6106	0.5752	5.80
0.6541	2.405	2.406	−0.04	0.0000	0.3023	0.308	−1.89
0.5577	2.089	2.042	2.25				
			$T = 308.15$ K				
1.0000	3.961	3.867	2.37	0.4567	2.212	2.213	−0.05
0.9190	3.905	4.013	−2.77	0.3508	1.620	1.705	−5.25
0.8345	3.773	3.918	−3.84	0.2396	1.178	1.218	−3.40
0.7463	3.711	3.629	2.21	0.1229	0.8375	0.7752	7.44
0.6541	3.301	3.211	2.73	0.0000	0.3994	0.4103	−2.73
0.5577	2.795	2.724	2.54				
			$T = 313.15$ K				
1.0000	5.212	5.091	2.32	0.4567	2.776	2.862	−3.10
0.9190	5.181	5.348	−3.22	0.3508	2.138	2.225	−4.07
0.8345	5.111	5.213	−2.00	0.2396	1.588	1.615	−1.70
0.7463	4.793	4.785	0.17	0.1229	1.109	1.045	5.77
0.6541	4.329	4.187	3.28	0.0000	0.5416	0.5536	−2.22
0.5577	3.678	3.525	4.16				
			$T = 318.15$ K				
1.0000	7.029	6.908	1.72	0.4567	3.683	3.718	−0.95
0.9190	7.055	7.196	−2.00	0.3508	2.808	2.920	−3.99
0.8345	6.777	6.932	−2.29	0.2396	2.096	2.155	−2.81
0.7463	6.303	6.284	0.30	0.1229	1.509	1.417	6.10
0.6541	5.577	5.445	2.37	0.0000	0.7391	0.7553	−2.19
0.5577	4.718	4.565	3.24				
			$T = 323.15$ K				
1.0000	9.602	9.421	1.89	0.4567	4.792	4.734	1.21
0.9190	9.420	9.667	−2.62	0.3508	3.609	3.746	−3.80
0.8345	8.950	9.158	−2.32	0.2396	2.700	2.806	−3.93
0.7463	8.365	8.172	2.31	0.1229	2.013	1.883	6.46
0.6541	7.046	6.992	0.77	0.0000	1.002	1.024	−2.20
0.5577	5.914	5.817	1.64				

as a possible mathematical representation for describing how the experimental isothermal solubility of a crystalline solute dissolved in a binary solvent mixture varies with binary solvent composition. S_i is the model constant, and N can be equal to 0, 1, 2, and 3, respectively. Depending on the values of N, four equations can be obtained from eq 3. x_B^0 and x_C^0 refer to the initial mole fraction composition of the binary solvent calculated as if solute A was not present. $(x_A)_i$ is the saturated mole fraction solubility of the solute in pure solvent i.

Substitution of $(1 - x_C^0)$ for x_B^0 in eq 3 with $N = 2$ and subsequent rearrangements result in eq 4

$$\ln x_A = \ln(x_A)_B + [\ln(x_A)_C - \ln(x_A)_B + S_0 + S_1 + S_2]x_C^0 + [-S_0 + 3S_1 + 5S_2]x_C^{02} + [-2S_1 - 8S_2]x_C^{03} + [-4S_2]x_C^{04} \quad (4)$$

which can be written as eq 5

$$\ln x_A = B_0 + B_1 x_C^0 + B_2 x_C^{02} + B_3 x_C^{03} + B_4 x_C^{04} \quad (5)$$

Table 2. Parameters of Equation 5 for Xylitol with the Temperature Range from 293 K to 343 K

T/K	B_4	B_3	B_2	B_1	B_0
293.15	−2.1941	3.5479	−3.5659	4.3355	−8.5061
298.15	−2.4539	4.7874	−5.2323	5.1776	−8.3246
303.15	−2.4954	5.7814	−6.8381	5.8401	−8.0855
308.15	−3.3108	7.2713	−7.7421	6.0250	−7.7987
313.15	−4.5763	9.7079	−9.0579	6.1450	−7.4990
318.15	−5.2905	11.2530	−9.9312	6.1820	−7.1884
323.15	−5.7248	12.1400	−10.2360	6.0396	−6.8836

The experimental solubility data x_A^{exptl} directly correlated with eq 5 and the calculated solubilities x_A^{calcd} are listed in Table 1 together with the relative errors (RE). The values of the five dimensionless parameters B_0, B_1, B_2, B_3, and B_4 are listed in Table 2. The RE is defined as the following

$$\mathrm{RE} = \frac{x_1^{exptl} - x_1^{calcd}}{x_1^{exptl}} \cdot 100 \quad (6)$$

where x_1^{calcd} is the solubility calculated from eq 5 and x_1^{exptl} is the experimental value of solubility. From Table 1, the following conclusions can be drawn: (1) For all selected mixed solvent systems, solubility is a function of temperature and solvent composition, and solubility sharply increases with increasing temperature and ethanol content of the original mixed solvent. (2) Xylitol can crystallize from the mixed solvent of ethanol and acetone by cooling. (3) The calculated solubility of eq 5 shows good agreement with the experimental values. The maximum deviation in RE is 7.44, and the mean absolute deviation is 2.35.

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