Solubility of 2,6-Diamino-3,5-dinitropyridine and 2,5-Dihydroxyterephthalic Acid in *N*,*N*-Dimethylformamide, Dimethylsulfoxide, Ethanol, and Methanol, *N*,*N*-Dimethylacetamide, and Acetic Acid

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Solubility was reported for 2,6-diamino-3,5-dinitropyridine in *N*,*N*-dimethylformamide at (304.15 to 357.15) K, dimethylsulfoxide at (304.15 to 339.15) K, ethanol at (305.15 to 340.15) K, methanol at (293.15 to 339.15) K, and 2,5-dihydroxyterephthalic acid in *N*,*N*-dimethylformamide at (298.15 to 370.15) K, dimethylsulfoxide at (293.15 to 342.15) K, *N*,*N*-dimethylacetamide at (318.15 to 355.55) K, and acetic acid at (320.15 to 369.15) 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 reasonable mathematical representation, with the average deviations between experimental values and calculated ones being on the order of ± 1.0 % and ± 0.2 % or less. Furthermore, the coupler parameters of the Margules equation (A_{21}, A_{12}) and the enthalpy of fusion ($\Delta_{fus}H$) were obtained by the Levenberg−Marquardt method and Global Optimization method.

Introduction

2,6-Diamino-3,5-dinitropyridine is a yellow crystal. 2,6-Diamino-3,5-dinitropyridine can be used as an insensitive explosive, a multifunctional organic reagent, as well as an important intermediate in the chemistry industry. For instance, 2,6-diamino-3,5-dinitropyridine could prepare 2,6-diamino-3,5-dinitropyridine-1-oxide, an insensitive explosive.[1] Furthermore, it is an important organic intermediate for 2,3,5,6-tetraaminospyridine, which is the monomer for the preparation of polypyridobisimidazole.[2] 2,6-Diamino-3,5-dinitropyridine is mainly prepared by the nitration reaction under the strong acid system.[3] The pure production is always obtained by extraction or crystallization.[4] What's more, solubility data for a solute−solvent system are the starting point to determine or estimate some crystallization parameters and reaction kinetics or thermodynamics study. There, however, has been no report as to its solubility data in some solvents.

2,5-Dihydroxyterephthalic acid is a type of yellow crystal. It is an important component and intermediate in the chemical industry. 2,5-Dihydroxyterephthalic acid could react with scandium chloride and nitrate to form three-dimensional framework compounds.[5] Furthermore, anisotropic homopolymerization of 2,5-dihydroxyterephthalic acid and 2,3,5,6-tetraaminopyridine or 2,5-diamino-1,4-benzenedithiol dihydrochloride and copolymerization of 2,5-dihydroxyterephthalic acid, terephthalic acid, and 2,5-diamino-1,4-benzenedithiol dihydrochloride in polyphosphoric acid could afford high molecular weight rigid-rod polymers and copolymers.[2,6] Recently, it was reported that surface wettability and interfacial adhesion ability of poly(*p*-phenylene benzoxazole), to a large extent, had been improved by introducing 2,5-dihydroxyterephthalic acid into poly(*p*-phenylene benzoxazole) macromolecular chains.[7] The properties of 2,5-dihydroxyterephthalic acid had an important effect on the preparation of the high-property polymers. What's more, solubility data for a solute−solvent system are the starting point to determine or estimate some crystallization parameters and reaction kinetics or thermodynamics study. Although 2,5-dihydroxyterephthalic acid had been produced in scale, there has been no report as to its solubility data in some solvents.

In this work, the solubility of 2,6-diamino-3,5-dinitropyridine in *N*,*N*-dimethylformamide, dimethylsulfoxide, ethanol, methanol, and 2,5-dihydroxyterephthalic acid in *N*,*N*-dimethylformamide, *N*,*N*-dimethylacetamide, dimethylsulfoxide, and acetic acid was measured by the analytical method. The experimental solubility data were correlated by a three-parameter model based on the Margules equation.

Experimental Section

Materials. 2,6-Diamino-3,5-dinitropyridine prepared in the laboratory was recrystallized prior to use.[3] Its purity was the mass fraction greater than 0.9975 % as determined by high-performance liquid chromatography measurement. Its melting point temperature is 595.15 K by thermal gravimetric and thermal decomposition analysis. 2,5-Dihydroxyterephthalic acid prepared in the laboratory was recrystallized prior to use.[2] Its mass fraction purity was above 0.999 % through the high-performance liquid chromatography measurement. Its melting point temperature is 628.35 K by thermal gravimetric and thermal decomposition analysis. *N*,*N*-Dimethylformamide, *N*,*N*-dimethylacetamide, dimethylsulfoxide, ethanol, methanol, and acetic acid (purchased from Tianjin Chemistry Reagent Co., China) are in chromatographically pure grade. The purities of those solvents are the mass fraction higher than 0.999 %.

Experimental Method. The solubility of a solid in a solvent can be measured by the analytical method[8,9] or the synthetic method.[10,11] We adopted the analytical method to determine the solubility of 2,6-diamino-3,5-dinitropyridine and 2,5-dihydroxyterephthalic acid in solvents. The disadvantage of the analytical method is tedious and time-consuming,[12] and its advantage is simple and reliable due to the possibility of measuring a large number of samples simultaneously. The concentrations of 2,6-

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Table 1. Experimentally Determined Solubility Data of 2,6-Diamino-3,5-dinitropyridine (2) in *N*,*N*-Dimethylformamide (1), Dimethylsulfoxide (1), Ethanol (1), and Methanol (1)

T/K	x_2	$(x_2 - x_{2cal})/x_2$	T/K	x_2	$(x_2 - x_{2cal})/x_2$
		N,*N*-Dimethylformamide			
304.15	0.00639	−0.0267	336.15	0.01318	0.0015
306.15	0.00694	−0.0234	338.15	0.01366	0.0022
309.15	0.0076	−0.0191	341.15	0.01409	0.0036
312.15	0.00815	−0.0154	343.15	0.01531	0.0033
315.15	0.00877	−0.0122	346.15	0.01642	0.0038
318.15	0.00944	−0.0095	348.15	0.01824	0.0031
321.15	0.00993	−0.0069	351.15	0.02007	0.0030
323.15	0.01044	−0.0056	354.15	0.02307	0.0023
326.15	0.01095	−0.0035	355.15	0.0261	0.0012
331.15	0.01199	−0.0007	357.15	0.02881	0.0008
333.15	0.01259	0.0001			
		Dimethylsulfoxide			
304.15	0.01505	−0.0014	323.15	0.05325	−0.0002
306.15	0.01518	−0.0001	327.15	0.06382	0.0000
309.15	0.01532	0.0019	329.15	0.06905	0.0001
311.15	0.02177	0.0002	331.15	0.0747	0.0000
314.15	0.02843	−0.0001	333.15	0.08032	0.0001
317.15	0.03607	−0.0002	336.15	0.08835	0.0001
320.15	0.04331	−0.0001	339.15	0.10645	−0.0001
		Ethanol			
305.15	0.000298	0.4020	323.15	0.00561	−0.0060
307.15	0.000783	0.0314	326.15	0.00807	−0.0042
309.15	0.0012	−0.0041	329.15	0.010142	−0.0028
311.15	0.00174	−0.0156	332.15	0.01219	−0.0012
314.15	0.00224	−0.0106	335.15	0.01527	0.0000
316.15	0.00269	−0.0094	338.15	0.01752	0.0011
319.15	0.00314	−0.0032	340.15	0.01996	0.0019
		Methanol			
293.15	0.00184	−0.0199	318.15	0.00915	0.0044
298.15	0.0026	−0.0072	319.15	0.00997	0.004
301.15	0.003345	−0.0056	321.15	0.01853	−0.0004
304.15	0.00341	0.0034	324.15	0.03343	−0.0011
306.15	0.00458	−0.0005	327.15	0.04583	−0.0007
308.15	0.00535	0.0001	330.15	0.05853	−0.0004
310.15	0.006138	0.0008	333.15	0.07279	−0.0001
313.15	0.00696	0.0031	336.15	0.08835	0.0001
315.15	0.008	0.0032	339.15	0.10645	0.0003

diamino-3,5-dinitropyridine and 2,5-dihydroxyterephthalic acid in the above solvents are analyzed through the high-performance liquid chromatography measurement. The solubility data were 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.[13,14]

Analytical Method Procedure. The apparatus and the procedure are similar to the case described in the literature.[15,16] 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 an analytical balance. The balance had a range of measurement up to 160 g, with an uncertainty of ± 0.0001 g. The estimated error in the mole fraction is less than 0.0001.

An excess amount of solute is added to the solvents in a sealed dual-wall flask, which is similar to the case described in the literature.[16] The contents of the vessel were heated very slowly at rates less than 1.5 K·h^{-1} with continuous stirring. To make it attain equilibrium, the solution is constantly stirred for 2.5 h at the specified temperature, and then the stirring is stopped to let the solution settle for 1.5 h. A clear liquid (about 2.5 mL) is taken into the sampling vial by a heated pipet. The mass of the sample was determined using an analytical balance with an uncertainty of ± 0.0001 g. To make the solute dissolute completely at normal temperature, more solvent was added into the sampling vial. By repeating the above procedure at different specified temperatures, a series of samples were obtained. All samples were analyzed by the high-performance liquid chromatography measurement. On the basis of the calibration curve and diluted multiple, the real concentrations of samples at different temperatures can be obtained.

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 as[17]

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

where γ_2 is the liquid-phase activity coefficient of solute; x_2 is the mole fraction of solute; $\Delta_{fus}H$ 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[17]

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

where A_{21} and A_{12} are the coupler parameters of the Margules equation and A_{21} is related to the infinite-dilution activity coefficient (γ_2^∞) of solute by the expression[16]

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

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

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

The Levenberg−Marquardt method and global optimization method were used to regress the experimental solubility data with the above equation to estimate the parameters A_{21}, A_{12}, and $\Delta_{fus}H$.

Results and Discussion

The solubility of 2,6-diamino-3,5-dinitropyridine in *N*,*N*-dimethylformamide, dimethylsulfoxide, ethanol, and methanol was measured by the analytical method in the temperature range of (304.15 to 359.15) K, and the data were shown in Table 1. The solubilities of 2,5-dihydroxyterephthalic acid in *N*,*N*-dimethylformamide, *N*,*N*-dimethylacetamide, dimethylsulfoxide, and acetic acid were measured by the analytical method in the

Table 2. Margules Equation Fitting Parameters of 2,6-Diamino-3,5-dinitropyridine in *N*,*N*-Dimethylformamide, Dimethylsulfoxide, Ethanol, and Methanol

solvent	$\Delta_{fus}H$ kJ·mol^{-1}	A_{21}	A_{12}	γ_2^∞	$10^5\sigma$	R^2	DC
N,*N*-dimethylformamide	68.1516	−7.00003	7.56834	0.00091	8.23567	0.999928	0.999818
dimethylsulfoxide	67.0524	−8.63364	−36.8130	0.00018	1.15022	0.999999	0.999999
ethanol	67.5976	−5.82805	2.17000	0.00294	3.97280	0.999962	0.999962
methanol	66.5200	−7.24039	−3.15103	0.00072	2.40436	0.999999	0.999999

Table 3. Solubility of 2,5-Dihydroxyterephthalic Acid (2) in *N*,*N*-Dimethylformamide (1), Dimethylsulfoxide (1), *N*,*N*-Dimethylacetamide (1), and Acetic Acid (1)

T/K	x_2	$(x_2 - x_{2cal})/x_2$	T/K	x_2	$(x_2 - x_{2cal})/x_2$
		N,*N*-Dimethylformamide			
298.15	0.01274	−0.02198	356.15	0.02775	0.004685
301.15	0.01345	−0.01859	358.15	0.03105	0.003865
308.15	0.01474	−0.01289	363.15	0.03839	0.002865
331.15	0.01903	−0.00053	367.15	0.04268	0.002577
344.15	0.02227	0.003592	370.15	0.04826	0.001865
353.15	0.02443	0.005731			
		Dimethylsulfoxide			
293.15	0.0273	−0.0033	326.15	0.06214	0.000644
303.15	0.03417	−0.00088	330.15	0.07419	0.000135
307.15	0.03694	0.000000	333.15	0.0827	−0.00012
313.15	0.04004	0.000999	336.15	0.09008	−0.00022
317.15	0.04625	0.000865	342.15	0.10438	−0.00038
321.15	0.05172	0.000967			
		N,*N*-Dimethylacetamide			
318.15	0.0034	−0.03529	343.15	0.00874	0.003432
323.15	0.00376	−0.01862	345.15	0.00988	0.003036
328.15	0.00414	−0.00966	348.15	0.01111	0.0045
333.15	0.0053	−0.00189	351.15	0.01474	0.002714
338.15	0.00646	0.003096	353.15	0.01654	0.002418
340.15	0.00768	0.002604	355.55	0.01861	0.002149
		Acetic Acid			
320.15	0.00443	−0.03612	356.15	0.00938	0.008529
328.15	0.00515	−0.01942	360.15	0.01023	0.010753
336.15	0.00618	−0.00647	366.15	0.01186	0.011804
343.15	0.00716	0.001397	369.15	0.0141	0.009929
352.15	0.00857	0.007001			

temperature range of (298.15 to 370.15) K, and the data were shown in Table 3.

Equation 3 was used to regress the experimental data in Tables 1 and 3, and the parameters (A_{21}, A_{12}, and $\Delta_{fus}H$) in eq 3 were obtained and shown in Tables 2 and 4, respectively. The values of the root-mean-square deviations (rmsd) and the correlation coefficients (R^2), determination coefficient (DC), and γ_2^∞ were also listed in Tables 2 and 4. The rmsd of the mole fraction is defined as

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

where N is the number of experimental points; $x_{2\ cali}$ represents the solubility calculated from eq 3; and x_{2i} represents the experimental solubility value. The comparisons between experimental data and calculated ones are shown in Tables 1 and 3 and Figures 1, 2, and 3. The average deviation between experimental values and calculated ones is 1.0 % and 0.2 %, respectively. It can be seen that the solubility model can be used to describe the variation with temperature of the solubility of solute.

According to the data shown in Table 2, it is obvious that the greater the solubility of 2,6-diamino-3,5-dinitropyridine in the solvent, the smaller the γ_2^∞ is, which is in accord with the thermodynamics. The solubility curves of 2,6-diamino-3,5-dinitropyridine in *N*,*N*-dimethylformamide, dimethylsulfoxide, ethanol, and methanol are shown in Figures 1 and 2, respectively. It is seen that the solubility of 2,6-diamino-3,5-dinitropyridine in these solvents decreased in the sequence dimethylsulfoxide, methanol, *N*,*N*-dimethylformamide, and ethanol. On the other hand, the solubility of 2,6-diamino-3,5-dinitropyridine is much larger in the stronger polar and inertia solvents such as dimethylsulfoxide. 2,6-Diamino-3,5-dinitropyridine could form the intermolecular hydrogen bond with some solvents containing an oxygen or hydroxyl group due to its amino and nitrogen, which could improve its solubility in these solvents. The molecular rearrangement in the solution depends on the possibility of hydrogen bond formation between solute and solvent.[18] The hydrogen bonds promote the formation of the new crystal structure. Furthermore, the influence of the temperature on the solubility of 2,6-diamino-3,5-dinitropyridine is not obvious below 320 K; however, the influence becomes more obvious in dimethylsulfoxide and methanol above 320 K. Therefore, dimethylsulfoxide and methanol should be used to purify 2,6-diamino-3,5-dinitropyridine. As seen from Figure 2, the temperature dependence of the solubility of 2,6-diamino-

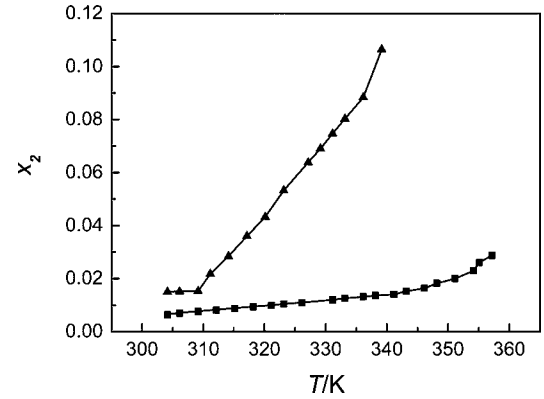

Figure 1. Solubility of 2,6-diamino-3,5-dinitropyridine (2) in *N*,*N*-dimethylformamide and dimethylsulfoxide: ■, experimental data in *N*,*N*-dimethylformamide; ▲, experimental data in dimethylsulfoxide; —, calculated data.

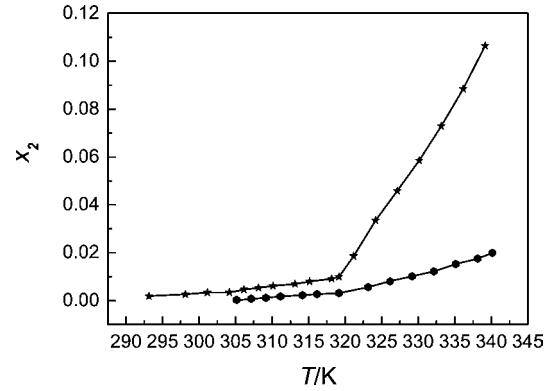

Figure 2. Solubility of 2,6-diamino-3,5-dinitropyridine (2) in ethanol and methanol: ★, experimental data in methanol; ●, experimental data in ethanol; —, calculated data.

Table 4. Margules Equation Fitting Parameters of 2,5-Dihydroxyterephthalic Acid in *N*,*N*-Dimethylformamide, Dimethylsulfoxide, *N*,*N*-Dimethylacetamide, and Acetic Acid

solvents	$\Delta_{fus}H$ kJ·mol^{-1}	A_{12}	A_{21}	σ	γ_2^∞	R^2	DC
N,*N*-dimethylformamide	140.923	9.601	−21.309	0.000156	$5.57928 \cdot 10^{-10}$	0.999914	0.999817
dimethylsulfoxide	143.353	−9.392	−27.872	0.000041	$7.88133 \cdot 10^{-13}$	0.999997	0.999997
N,*N*-dimethylacetamide	139.038	6.900	−17.833	0.000049	$1.80314 \cdot 10^{-08}$	0.999953	0.999900
acetic acid	136.303	2.890	−16.604	0.00012	$6.16201 \cdot 10^{-08}$	0.999796	0.998507

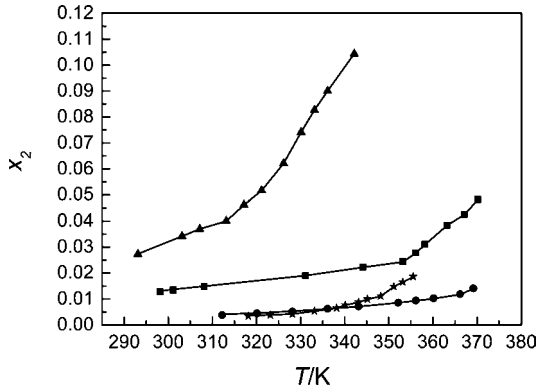

Figure 3. Solubility of 2,5-dihydroxyterephthalic acid (2) in *N*,*N*-dimethylformamide, dimethylsulfoxide, *N*,*N*-dimethylacetamide, and acetic acid: ■, experimental data in *N*,*N*-dimethylformamide; ▲, experimental data in dimethylsulfoxide; ★, experimental data in *N*,*N*-dimethylacetamide; ●, experimental data in acetic acid; —, calculated data.

3,5-dinitropyridine in methanol has a clear break. To a large extent, the simple 2,6-diamino-3,5-dinitropyridine molecules with hydrophilic nitrogen groups could cause specific and dramatic interactions with the hydroxyl of methanol and promote its solubility at higher temperature. From another point of view, the lone electron pair of the amino nitrogen on the outer ring of 2,6-diamino-3,5-dinitropyridine had the electron cloud of amino migrate to the pyridine ring because of the p-π conjugate effect, which makes amino alkalescence weakened and the proton activity improved. In addition, the space location obstruct of methanol is weaker, and the boiling point was lower than other solvents. Along with the increasing temperature, the thermal movement between these molecules becomes more strenuous. On the basis of the above reasons, therefore, there is a clear break of solubility in the methanol at higher temperature. Table 2 shows that the enthalpies of fusion $\Delta_{fus}H$ of 2,6-diamino-3,5-dinitropyridine in different solvents are consistent, obtained by the Levenberg−Marquardt method and global optimization method. The average value is 67.3304 kJ·mol^{-1}.

According to the data shown in Table 4, it is obvious that the greater the solubility of DHTA in the solvent, the smaller the γ_2^{∞} is, which is in accord with the thermodynamics. The solubilities curves of 2,5-dihydroxyterephthalic acid in *N*,*N*-dimethylformamide, *N*,*N*-dimethylacetamide, dimethylsulfoxide, and acetic acid are shown in Figure 3. The solubility of 2,5-dihydroxyterephthalic acid in these solvents increases along with the rising temperature. It is found that the solubility of 2,5-dihydroxyterephthalic acid in these solvents decreased in the sequence dimethylsulfoxide, *N*,*N*-dimethylformamide, *N*,*N*-dimethylacetamide, and acetic acid. On the other hand, the solubility of 2,5-dihydroxyterephthalic acid is much greater in the stronger polar and inertia solvents, such as dimethylsulfoxide and *N*,*N*-dimethylformamide. Importantly, the intermolecular hydrogen bond formation between 2,5-dihydroxyterephthalic acid and these solvents improves its solubility due to the hydroxyl group of 2,5-dihydroxyterephthalic acid. In addition, based on the Lewis theory, *N*,*N*-dimethylformamide is an alkaline aprotic solvent, which enhances the solvent effect between 2,5-dihydroxyterephthalic acid and *N*,*N*-dimethylformamide molecules and promotes the solubility of 2,5-dihydroxyterephthalic acid in *N*,*N*-dimethylformamide fatherly. As seen from Figure 3, the influence of the temperature on the solubility of 2,5-dihydroxyterephthalic acid is much greater in dimethylsulfoxide below 355 K, compared to other solvents. On the other hand, the influence of the temperature on the solubility of 2,5-dihydroxyterephthalic acid is not obvious in *N*,*N*-dimethylacetamide and acetic acid. Especially, the solubility of 2,5-dihydroxyterephthalic acid in acetic acid is greater at higher temperature than in acetic acid at the lower temperature; however, the solubility in acetic acid is smaller than in *N*,*N*-dimethylacetamide gradually above 336 K. To a large extent, *N*,*N*-dimethylacetamide's amino promotes the solubility of 2,5-dihydroxyterephthalic acid, compared to acetic acid. On the basis of the above reasons, therefore, dimethylsulfoxide and *N*,*N*-dimethylformamide should be used to purify 2,5-dihydroxyterephthalic acid. Table 4 shows that the enthalpies of fusion ($\Delta_{fus}H$) of 2,5-dihydroxyterephthalic acid in different solvents are consistent, obtained by the Levenberg−Marquardt method and global optimization method. The average value is 139.904 kJ·mol^{-1}.

Conclusion

Solubility was reported for 2,6-diamino-3,5-dinitropyridine in *N*,*N*-dimethylformamide at (304.15 to 357.15) K, dimethylsulfoxide at (304.15 to 339.15) K, ethanol at (305.15 to 340.15) K, methanol at (293.15 to 339.15) K, and 2,5-dihydroxyterephthalic acid in *N*,*N*-dimethylformamide at (298.15 to 370.15) K, dimethylsulfoxide at (293.15 to 342.15) K, *N*,*N*-dimethylacetamide at (318.15 to 355.55) K, and acetic acid at (320.15 to 369.15) K. The Margules equation was used to fit the experimental data, and the average deviations between experimental values and calculated ones are on the order of ± 1.0 % and ± 0.2 % or less. Furthermore, the parameters (A_{21}, A_{12}, and $\Delta_{fus}H$) were obtained by the Levenberg−Marquardt method and global optimization method.

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