Ternary Excess Molar Enthalpies for Methanol or Ethanol + 1-Propanol + 2-Propanol at the Temperature 298.15 K

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The excess molar enthalpies for the ternary systems methanol + 1-propanol + 2-propanol and ethanol + 1-propanol + 2-propanol have been measured at 298.15 K and atmospheric pressure using a flow microcalorimeter. The ternary experimental results are correlated with a polynomial equation and compared with those predicted by the UNIQUAC associated-solution model having self-association and cross-association constants and binary parameters alone.

Introduction

It is well-known that in alcohol mixtures, alcohol molecules form associated species by hydrogen bonding. On the basis of the association of alcohol, the excess molar enthalpies of the binary and ternary mixtures containing alcohol−hydrocarbon as well as alcohol−alcohol have been explained accurately (Acree, 1984; Nagata et al., 1986; Nagata and Tamura, 1998). Recently, an extended version of the UNIQUAC associated-solution model (Nagata et al., 1988) was proposed to analyze the excess molar enthalpies of ternary mixtures including three different alcohols. The model assumes the linear association and multisolvation of alcohol molecules in the framework of the UNIQUAC model. The model was successfully applied to represent the ternary excess molar enthalpies for the methanol + ethanol + 1-propanol and methanol + ethanol + 2-propanol systems and those for each of the constituent binary systems.

In the present work the ternary excess molar enthalpies for mixtures composed of three alcohols are further studied to test the applicability of the model. We report the ternary excess molar enthalpies of the methanol + 1-propanol + 2-propanol and ethanol + 1-propanol + 2-propanol systems measured at 298.15 K by using a flow microcalorimeter. The experimental results are compared with those predicted by the UNIQUAC associated-solution model using only the binary parameters obtained from the binary correlation of the excess molar enthalpies. The experimental excess molar enthalpies for the binary systems making up the ternary systems have been published: methanol + 1-propanol (Pflug et al., 1968); methanol + 2-propanol (Taylor and Bertrand, 1974); ethanol + 1-propanol (Pflug et al., 1968); ethanol + 2-propanol (Pflug et al., 1968); and 1-propanol + 2-propanol (Polak et al., 1970).

Experimental Section

Materials. Methanol, ethanol, 1-propanol, and 2-propanol were of guaranteed reagent grade (Wako Pure Chemical Industries Ltd., purity > 99.5 mass %). Prior to use the chemicals were degassed using ultrasonic sound and then dried over molecular sieves. GLC analysis detected no significant peaks of impurities for all the components. The densities of the chemicals used, measured with a densimeter (Anton-Paar, DMA58) at 298.15 K, compared well with literature values (Table 1) (Riddick et al., 1986).

Table 1. Densities *d* of the Chemicals

component	d(298.15 K)/g·cm^{-3} exptl	d(298.15 K)/g·cm^{-3} lit.[a]
methanol	0.786 68	0.786 37
ethanol	0.785 03	0.784 93
1-propanol	0.799 75	0.799 60
2-propanol	0.781 22	0.781 26

[a] Riddick et al. (1986).

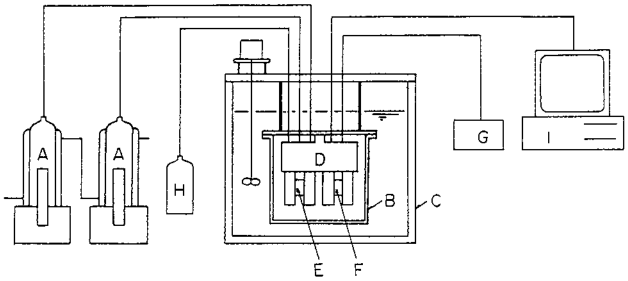

Figure 1. Schematic diagram of flow calorimeter: A, piston displacement pump; B, brass container; C, thermostated water bath; D, aluminum heat sink block; E, flow mixing cell; F, reference cell; G, dc power supply; H, waste reservoir; I, data acquisition system.

Apparatus and Procedure. The excess molar enthalpies H_m^E were measured with a flow microcalorimeter at (298.15 ± 0.005) K (Nagata et al., 1992). The apparatus is schematically shown in Figure 1. The flow of the first and second components is fed through inlet tubes into the flow mixing cell by the piston displacement pumps A. The pumps regulate the flow rates of the liquids, fluctuations of which between 0.5 and 5.0 mL min^{-1} are less than 0.5%. The total volumetric flow of the mixtures was fixed at 5 mL min^{-1} for all measurements. The ratio of the flow rates of each liquid gives the composition of the liquid mixture. The brass container B was immersed in the thermostated water bath C. The aluminum heat sink block D had two identical compartments for the flow mixing cell E and the reference cell F. The mixtures pass through the flow mixing

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Table 2. Experimental Ternary Excess Molar Enthalpies at 298.15 K for the Methanol + 1-Propanol + 2-Propanol and Ethanol + 1-Propanol + 2-Propanol Systems

Methanol (1) + 1-Propanol (2) + 2-Propanol (3)[a]

$x'_2 = 0.2493$, $H^E_{m,23} = -37.86$ J·mol^{-1}				$x'_2 = 0.4998$, $H^E_{m,23} = -48.96$ J·mol^{-1}				$x'_2 = 0.7488$, $H^E_{m,23} = -35.13$ J·mol^{-1}			
x_1	x_2	ΔH^E_m/J·mol^{-1}	$H^E_{m,123}$/J·mol^{-1}	x_1	x_2	ΔH^E_m/J·mol^{-1}	$H^E_{m,123}$/J·mol^{-1}	x_1	x_2	ΔH^E_m/J·mol^{-1}	$H^E_{m,123}$/J·mol^{-1}
0.0500	0.2369	−7.2	−43.2	0.0500	0.4752	3.9	−42.6	0.0500	0.7114	8.2	−25.2
0.1000	0.2244	−13.2	−47.3	0.1000	0.4502	5.1	−38.9	0.1000	0.6739	15.6	−16.0
0.1500	0.2120	−17.3	−49.5	0.1500	0.4252	6.9	−34.7	0.1500	0.6365	22.9	−7.0
0.2000	0.1995	−21.3	−51.6	0.2000	0.4002	8.9	−30.3	0.2000	0.5991	29.8	1.7
0.2500	0.1870	−22.9	−51.3	0.2500	0.3751	11.5	−25.2	0.2500	0.5616	36.0	9.7
0.3000	0.1745	−24.3	−50.8	0.3000	0.3501	14.2	−20.1	0.3000	0.5242	41.9	17.3
0.3500	0.1621	−23.9	−48.6	0.3500	0.3251	17.3	−14.5	0.3500	0.4876	47.2	24.3
0.4000	0.1496	−22.8	−45.5	0.4000	0.3001	19.9	−9.5	0.4000	0.4493	51.6	30.6
0.4500	0.1371	−21.7	−42.5	0.4500	0.2751	22.5	−4.4	0.4500	0.4119	55.2	35.9
0.5000	0.1247	−20.0	−38.9	0.5000	0.2501	24.2	−0.3	0.5000	0.3744	57.3	39.7
0.5500	0.1122	−17.2	−34.2	0.5500	0.2251	25.7	3.7	0.5500	0.3370	58.5	42.7
0.6000	0.0997	−14.0	−29.1	0.6000	0.2001	27.4	7.8	0.6000	0.2995	58.5	44.5
0.6500	0.0873	−11.7	−25.0	0.6500	0.1751	27.8	10.7	0.6500	0.2621	58.8	46.5
0.7000	0.0748	−9.2	−20.6	0.7000	0.1501	28.0	13.3	0.7000	0.2247	56.7	46.2
0.7500	0.0623	−6.3	−15.8	0.7500	0.1251	27.2	15.0	0.7500	0.1872	53.1	44.3
0.8000	0.0499	−3.5	−11.1	0.8000	0.1000	25.5	15.7	0.8000	0.1498	47.4	40.4
0.8500	0.0374	−1.0	−6.7	0.8500	0.0750	22.1	14.8	0.8500	0.1123	39.6	34.3
0.9000	0.0249	0.6	−3.2	0.9000	0.0500	17.1	12.2	0.9000	0.0749	29.3	25.7
0.9500	0.0125	1.1	−0.8	0.9500	0.0250	10.4	7.9	0.9500	0.0374	16.3	14.5

Ethanol (1) + 1-Propanol (2) + 2-Propanol (3)[b]

$x'_2 = 0.2477$, $H^E_{m,23} = -37.70$ J·mol^{-1}				$x'_2 = 0.5002$, $H^E_{m,23} = -48.95$ J·mol^{-1}				$x'_2 = 0.7488$, $H^E_{m,23} = -35.13$ J·mol^{-1}			
x_1	x_2	ΔH^E_m/J·mol^{-1}	$\Delta H^E_{m,123}$/J·mol^{-1}	x_1	x_2	ΔH^E_m/J·mol^{-1}	$\Delta H^E_{m,123}$/J·mol^{-1}	x_1	x_2	ΔH^E_m/J·mol^{-1}	$\Delta H^E_{m,123}$/J·mol^{-1}
0.0500	0.2353	−4.7	−40.5	0.0500	0.4752	0.7	−45.8	0.0500	0.7114	3.4	−29.9
0.1000	0.2229	−8.5	−42.4	0.1000	0.4502	1.1	−42.9	0.1000	0.6739	5.7	−26.0
0.1500	0.2105	−11.4	−43.4	0.1500	0.4251	1.5	−40.1	0.1500	0.6365	7.6	−22.3
0.2000	0.1982	−13.8	−43.5	0.2000	0.4001	1.6	−37.6	0.2000	0.5991	9.4	−18.7
0.2500	0.1858	−15.9	−44.2	0.2500	0.3751	1.4	−35.3	0.2500	0.5616	11.1	−15.3
0.3000	0.1734	−17.5	−44.0	0.3000	0.3501	1.5	−32.7	0.3000	0.5242	12.5	−12.1
0.3500	0.1610	−18.4	−42.9	0.3500	0.3251	1.8	−30.0	0.3500	0.4876	13.7	−9.2
0.4000	0.1486	−19.1	−42.0	0.4000	0.3001	2.1	−27.3	0.4000	0.4493	14.4	−6.6
0.4500	0.1362	−19.4	−40.1	0.4500	0.2751	2.5	−24.4	0.4500	0.4119	15.1	−4.2
0.5000	0.1238	−19.4	−38.2	0.5000	0.2501	2.9	−21.6	0.5000	0.3744	15.3	−2.3
0.5500	0.1115	−19.1	−36.0	0.5500	0.2251	3.5	−18.5	0.5500	0.3370	15.2	−0.6
0.6000	0.0991	−18.0	−33.1	0.6000	0.2001	3.6	−16.0	0.6000	0.2995	14.9	0.8
0.6500	0.0867	−16.5	−29.7	0.6500	0.1751	3.9	−13.2	0.6500	0.2621	14.3	2.0
0.7000	0.0743	−14.7	−26.0	0.7000	0.1501	4.0	−10.7	0.7000	0.2247	13.4	2.9
0.7500	0.0619	−12.8	−22.3	0.7500	0.1250	4.1	−8.2	0.7500	0.1872	12.2	3.4
0.8000	0.0495	−10.2	−17.8	0.8000	0.1000	4.0	−5.8	0.8000	0.1498	10.6	3.6
0.8500	0.0372	−7.1	−12.8	0.8500	0.0750	3.7	−3.7	0.8500	0.1123	8.6	3.3
0.9000	0.0248	−4.1	−7.9	0.9000	0.0500	3.5	−1.4	0.9000	0.0749	6.2	2.7
0.9500	0.0124	−1.4	−3.2	0.9500	0.0250	2.3	0.2	0.9500	0.0374	3.4	1.6

[a] Mixtures were obtained by adding pure methanol with {$x'_2$1-propanol + (1 − x'_2)2-propnaol}. [b] Mixtures were obtained by adding pure ethanol with {$x'_2$1-propanol + (1 − x'_2)2-propnaol}.

cell and then flow out of the cell into the waste reservoir H. Mixing occurs at the flow mixing cell. The differences of heat flux between these two cells were converted to voltages by two pairs of thermomodules and then recorded by the data acquisition system I fitted with a digital voltmeter. After equilibrium voltages are established, excess molar enthalpies are calculated from the voltages using calibration constants. The calibration constants of the thermomodule were obtained by supplying a known amount of direct current to a calibrated heater in the flow mixing cell using the dc power supply G. Agreement between our results and the literature values for the benzene + cyclohexane system (Elliott and Wormald, 1976; Nagata et al., 1992) was good. The experimental errors were within ±0.5% of H^E_m in excess molar enthalpies and ±0.00005 in mole fraction. Three experimental runs were performed for ternary mixtures formed by adding methanol or ethanol to binary mixtures of 1-propanol and 2-propanol. A ternary mixture may be considered as a pseudobinary mixture composed of methanol or ethanol and one binary mixture. The excess molar enthalpy $H^E_{m,23}$ of the ternary mixture of composition x_1, x_2, and x_3 can be expressed as follows:

$$H^E_{m,123} = \Delta H^E_m + (1 - x_1)H^E_{m,23} \tag{1}$$

where ΔH^E_m is the molar enthalpy measured for the pseudobinary mixture, $H^E_{m,23}$ is the molar enthalpy for the initial binary 1-propanol (2) + 2-propanol (3) mixture, and x_1 is the mole fraction of methanol or ethanol. The values of $H^E_{m,23}$ at three specified compositions (the approximate compositions of these mixtures were 25, 50, and 75 mol % of component 2) were interpolated by means of a spline fit.

Results

The ternary excess molar enthalpies for the methanol (1) + 1-propanol (2) + 2-propanol (3) and ethanol (1) +

Table 3. Coefficients $a_{n,ij}$ of Eq 3, Absolute Arithmetic Mean Deviations, and Standard Deviations

system (i−j)	$a_{1,ij}$	$a_{2,ij}$	$a_{3,ij}$	$a_{4,ij}$	$\delta(H^E_m)$/J·mol^{-1}	$\sigma(H^E_m)$/J·mol^{-1}
methanol−1-propanol	329.35	101.03	15.96	4.56	0.1	0.2
methanol−2-propanol	−302.63	125.72	−8.07	1.05	0.0	0.0
ethanol−1-propanol	76.72	11.34	8.69	−1.58	0.1	0.1
ethanol−2-propanol	−181.80	23.33	−11.64	6.88	0.2	0.3
1-propanol−2-propanol	−195.64	14.56	3.16	−2.68	0.1	0.2

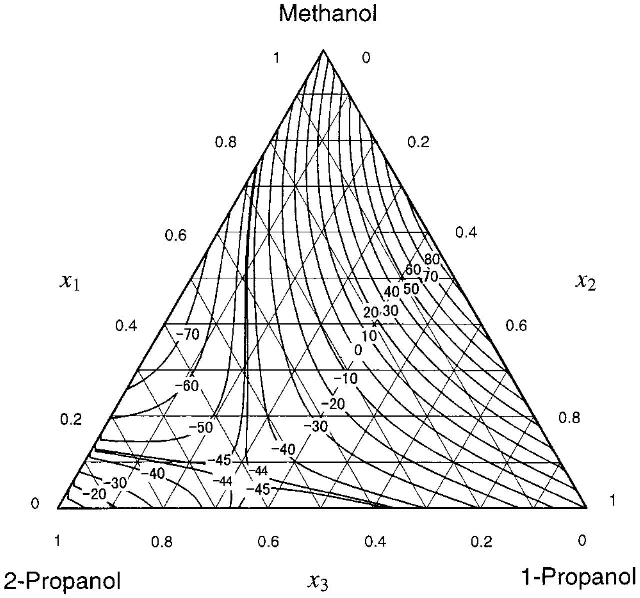

Figure 2. Contours of excess molar enthalpies for the ternary methanol (1) + 1-propanol (2) + 2-propanol (3) system at 298.15 K: —, calculated from eqs 2−4.

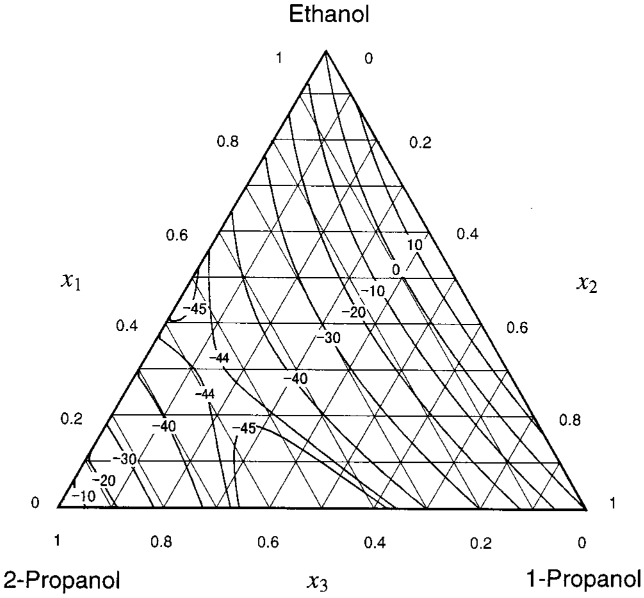

Figure 3. Contours of excess molar enthalpies for the ternary ethanol (1) + 1-propanol (2) + 2-propanol (3) system at 298.15 K: —, calculated from eqs 2−4.

1-propanol (2) + 2-propanol (3) systems at 298.15 K are given in Table 2 and fitted to an equation of the form

$$H^E_{m,123} = H^E_{m,12} + H^E_{m,13} + H^E_{m,23} + x_1x_2x_3\Delta_{123} \qquad (2)$$

where $H^E_{m,ij}$ is calculated from a polynomial equation of the form

$$H^E_{m,ij}/\mathrm{J\cdot mol^{-1}} = x_ix_j\sum_{n=1}^{p} a_{n,ji}(x_i - x_j)^{n-1} \qquad (3)$$

where $a_{n,ji}$ are the coefficients to be obtained by an unweighted least-squares method. Table 3 presents the coefficients of eq 3 for the five binary systems and the standard deviation, calculated by using $\sigma(H^E_m) = \{\sum_{i=1}^{m}(H^E_{exp} - H^E_{cal})^2/(m - p)\}^{0.5}$, where m is the number of experimental data points and p is the number of parameters. The additional term Δ_{123} in eq 2 is expressed by

$$\Delta_{123}/RT = \sum_{j=1}^{p} b_j(1 - 2x_1)^{j-1}/\{1 - k(1 - 2x_1)\} \qquad (4)$$

The values of the coefficients b_j and k, the absolute arithmetic mean deviation $\delta(H^E_m) = \sum_{i=1}^{m}|H^E_{exp} - H^E_{cal}|/m$, and the standard deviation obtained in fitting eqs 3 and 4 to the experimental ternary H^E_m are $b_1 = 0.01511$, $b_2 = 0.02182$, $b_3 = 0.10190$, $k = -1.04028$, $\delta(H^E_m) = 0.4$ J·mol^{-1}, and $\sigma(H^E_m) = 0.5$ J·mol^{-1} for the methanol (1) + 1-propanol (2) + 2-propanol (3) system and $b_1 = 0.02579$, $b_2 = 0.01343$, $b_3 = 0.03609$, $k = -1.07589$, $\delta(H^E_m) = 1.0$ J·mol^{-1}, and $\sigma(H^E_m) = 1.1$ J·mol^{-1} for the ethanol (1) + 1-propanol (2) + 2-propanol (3) system. Figures 2 and 3 show contours of excess molar enthalpies for the methanol + 1-propanol + 2-propanol and ethanol + 1-propanol + 2-propanol systems. The local minimum value of the excess molar enthalpies of the ternary methanol (1) + 1-propanol (2) + 2-propanol (3) mixture, calculated from eq 2, was −44.3 J·mol^{-1} at the ridge point $x_1 = 0.070$ and $x_2 = 0.310$, and that of the ethanol (1) + 1-propanol (2) + 2-propanol (3) mixture was −44.2 J·mol^{-1} at $x_1 = 0.293$ and $x_2 = 0.144$.

Table 4. Molecular Size and Surface Parameters

component	r_I	q_I
methanol	1.15	1.12
ethanol	1.69	1.55
1-propanol	2.23	1.98
2-propanol	2.23	1.98

Table 5. Self-Association Constants for Pure Alcohols and Cross-Association Constants between Unlike Alcohols at 323.15 K

component	K_I	component (I−J)	K_{IJ}
methanol	173.9	methanol−1-propanol	72.0
ethanol	110.4	methanol−2-propanol	70.0
1-propanol	87.0	ethanol−1-propanol	49.0
2-propanol	49.1	ethanol−2-propanol	47.6
		1-propanol−2-propanol	36.1

Data Analysis

The experimental excess enthalpies for the binary and ternary systems were analyzed by means of an extended version of the UNIQUAC associated-solution model proposed by Nagata et al. (1998). The model assumes alcohols A, B, and C form linear chains A_i, B_j, and C_k and complexes $(A_iB_j)_k$, $(B_iA_j)_k$, $(A_iC_j)_k$, $(C_iA_j)_k$, $(B_iC_j)_k$, and $(C_iB_j)_k$ according to successive chemical equilibria in a ternary mixture. Further, formation of ternary multicomplexes is followed by A_i, B_j, and C_k. The general formulas of the chemical

Table 6. Binary Parameters and Absolute Arithmetic Mean Deviations

system ($I-J$)	no. of data points	C_{JI}/K	C_{IJ}/K	D_{JI}	D_{IJ}	$\delta(H_m^E)$/J·mol^{-1}
methanol−1-propanol	21	1545.9	393.78	5.9782	0.7101	0.5
methanol−2-propanol	9	−567.81	−2437.1	−2.5365	-8.7288	2.2
ethanol−1-propanol	15	2159.6	1612.8	7.9383	5.8346	0.2
ethanol−2-propanol	19	866.42	794.82	2.9057	2.5056	2.3
1-propanol−2-propanol	19	1696.9	243.21	6.5314	0.2815	0.7

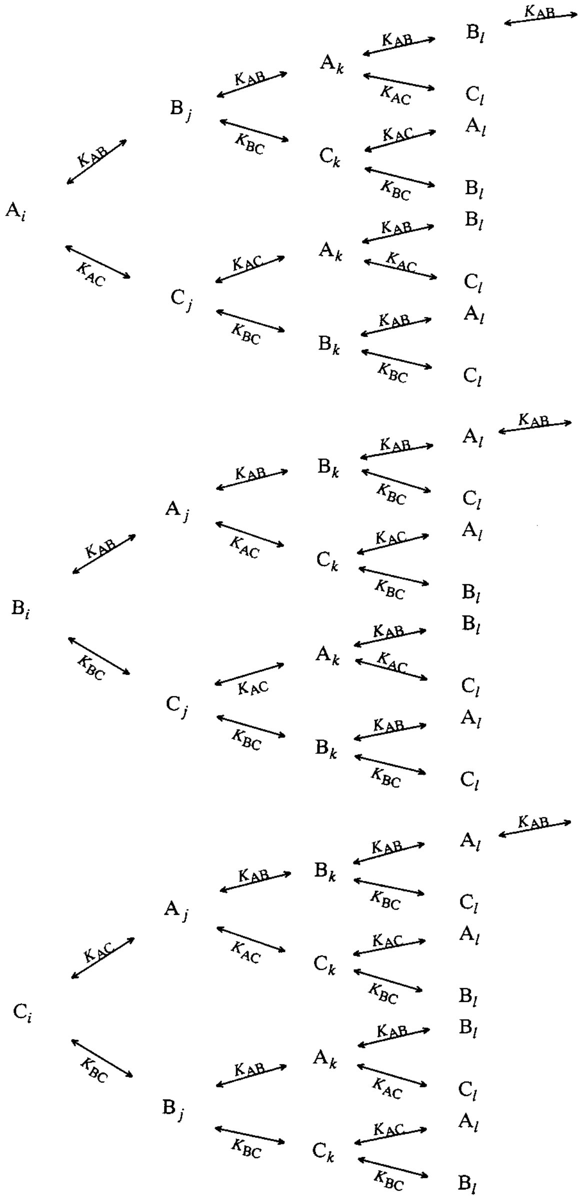

Figure 4. Ternary chemical complex-forming scheme.

complexes are expressed by $A_i(B_jA_k)_l$, $B_i(A_jB_k)_l$, $A_i(C_jA_k)_l$, $C_i(A_jC_k)_l$, $B_i(C_jB_k)_l$, and $C_i(B_jC_k)_l$, where i, j, k, and l range from 1 to infinity. Figure 4 shows a typical example of forming the ternary multicomplexes, starting from A_i, B_i, and C_i to third step, where the solvation equilibrium constant of $I-J$ pair-bonding formation is expressed by K_{IJ}. The equilibrium association constants of alcohols A, B, and C at 323.15 K, expressed by K_A, K_B, and K_C, were taken from Brandani (1983), and the enthalpy of hydrogen-bonding formation was $h_A = h_B = h_C = h_{AB} = h_{AC} = h_{BC} = -23.2$ kJ·mol^{-1} for all alcohols (Stokes and Burfitt, 1973). The temperature dependence of the equilibrium constants is fixed by the van't Hoff equation. The enthalpies of hydrogen-bonding formation were assumed to be independent of temperature. The pure-component molecular size and surface parameters r_I and q_I were estimated by the method of Vera et al. (1977). Tables 4 and 5 summarize the values of the pure-component molecular parameters and the self-association constants of the pure components and the cross-association constants between unlike alcohols. In fitting the model to binary H_m^E data, the energy parameter a_{ij} is assumed to be a linear function of temperature: $a_{ij} = C_{ij} + D_{ij}(T/K - 273.15)$. The coefficients C_{ij} and D_{ij} were obtained by minimizing the sum-of-squares between the experimental binary H_m^E and values calculated by the model using a simplex method (Nelder and Mead, 1965). Table 6 gives the binary coefficients and the absolute arithmetic-mean deviation $\delta(H_m^E)$ between the binary experimental and calculated values.

The model with the association and solvation constants and binary parameters alone predicted the ternary excess molar ethalpies. The absolute arithmetic mean deviations of the excess molar enthalpies were 2.9 J·mol^{-1} for the methanol + 1-propanol + 2-propanol system and 2.6 J·mol^{-1} for the ethanol + 1-propanol + 2-propanol system. Good agreement between the experimental and calculated results was obtained.

Conclusion

Ternary excess molar enthalpies have been measured for the methanol + 1-propanol + 2-propanol and ethanol + 1-propanol + 2-propanol systems at 298.15 K, and the results are compared well with those calculated by using the polynomial equation and the UNIQUAC associated-solution model. Further, the model taking into account the mulitisolvation among different alcohol multimers can successfully reproduce the excess molar enthalpies of the mixtures composed of three alcohols by using only the binary information.

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