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Excess Molar Enthalpies of 1,2-Propanediol + Alkan-1-ols (C_1−C_6) and Their Correlations at 298.15 K and Ambient Pressure (81.5 kPa)

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Excess molar enthalpies, H_m^E, of 1,2-propanediol + methanol, + ethanol, + propan-1-ol, + butan-1-ol, + pentan-1-ol, and + hexan-1-ol have been measured over the entire range of composition at 298.15 K and ambient pressure (81.5 kPa) using a Parr 1455 solution calorimeter. From the experimental results, the excess partial molar enthalpies, H_i^E, and excess partial molar enthalpies at infinite dilution, $H_i^{E,\infty}$, were calculated. While the excess molar enthalpies are negative for the methanol mixture, those for the ethanol, propan-1-ol, butan-1-ol, pentan-1-ol, and hexan-1-ol mixtures are positive over the entire range of composition of 1,2-propanediol. The H_m^E values increase as the chain length of the alkan-1-ol is increased. The experimental data were correlated by the Redlich−Kister equation and the two thermodynamics models (Wilson and NRTL).

Introduction

Experimental data of excess thermodynamic properties of liquid mixtures provide useful information about molecular interactions and can be used to test thermodynamic models.[1−4] 1,2-Propanediol and alkanol both have a proton donor and proton acceptor group. Therefore, there will be a significant degree of H-bonding, leading to self-association in the pure state, in addition to mutual association in their binaries. The thermophysical behaviors of 1,2-propanediol[5−14] both as a pure compound or in binary mixtures have been studied in recent years. 1,2-Propanediol is used extensively in the food, cosmetic, and pharmaceutical industries. It is an excellent solvent or vehicle for many medications and is nontoxic when used in prescribed amounts.

Excess molar enthalpies, H_m^E, have been measured for 1,2-propanediol + alkan-1-ols (C_1−C_6) over the entire range of composition at 298.15 K and ambient pressure (81.5 kPa). From the experimental results, the excess partial molar enthalpies, H_i^E, and excess partial molar enthalpies at infinite dilution, $H_i^{E,\infty}$, were calculated. The aim of this work is to provide a set of data for the characterization of the size of the alkan-1-ols on the thermodynamic properties of mixtures. These data help to obtain some insight into the intermolecular interactions, particularly hydrogen bonding properties, between the 1,2-propanediol and an alcohol with a different extent of chain length.

Experimental Section

Materials. The sources and the purities declared by the manufacturers are reported in Table 1. These liquids were used without further purification with the exception of hexan-1-ol, which was purified by the standard method of Perrin and Armarego.[15] The purity of solvents was further ascertained by comparing their densities and refractive indices at 298.15 K with the corresponding literature data,[13,16−19] as shown in Table 1.

Densities. The densities of the pure components, ρ, were determined at 298.15 K by means of an Anton Paar DMA 4500 oscillating u-tube density meter, provided with automatic viscosity correction. The temperature in the cell was regulated to ±0.01 K with a solid-state thermostat. Before measurements, the apparatus was calibrated using double-distilled freshwater and dry air. Uncertainty in the density was $\pm 1 \times 10^{-5}$ g·cm^{-3}.

Refractive Indices Measurements. Refractive indices were measured at 298.15 K using a thermostated Abbé refractometer. Water was circulated into the prism of the refractometer by a circulation pump connected to an external thermostated water bath. The uncertainty of the refractive index is on the order of ±0.0002 units, and for the temperature, it was ±0.01 K.

Calorimetric Measurements. The excess molar enthalpies, H_m^E, were measured with a Parr 1455 solution calorimeter. The measurements were carried out in an isolated room at 298 K and ambient pressure (81.5 kPa). In measurement of excess enthalpies, the known weight of 100 mL of component A is taken into the Dewar vessel and about 20 mL of component B is weighted in a sealed glass rotating cell. Both the components are kept in thermal contact with each other in the Dewar mixing chamber at 298.15 K. After attaining thermal equilibrium, component B in the rotating cell is mixed with component A in the Dewar vessel by pushing the glass rod. These experiments were continued until the concentration reached 50−60 vol %. This procedure was repeated by taking component A as component B and component B as component A. The detailed procedures and apparatus used have been described in our earlier publications.[3,20] The miscibility of the components was tested prior to calorimetric measurements, and the two components were found to be completely miscible over the whole concentration range. The temperature in calorimetric measurement can be read to an uncertainty of $\pm 2 \cdot 10^{-3}$ K.

Mole fractions of mixtures were determined by mass using a digital balance (AB 204-N Mettler) with an uncertainty of $\pm 1 \cdot 10^{-4}$ g. The uncertainty in the mole fraction is estimated to be lower than $\pm 2 \cdot 10^{-4}$. The performance and reliability of the solution calorimeter was checked by the test mixture cyclohexane + benzene. The excess enthalpies H_m^E agreed with the reported values.[21] The estimated relative uncertainty in the excess enthalpy measurements is $\pm 1 \cdot 10^{-2}$ of the determined value.

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Table 1. Sources, Purity Grades, Densities, ρ, and Refractive Indices, n_D, of the Pure Components at 298.15 K

			ρ/g·cm^{-3}		n_D	
component	source	purity/100 w	exp	lit	exp	lit[19]
1,2-propanediol	Merck	99.5 %	1.03261	1.0327[17]	1.4323	1.4314
methanol	Merck	99.5 %	0.78650	0.78637[16]	1.3270	1.3265
ethanol	Merck	99.8 %	0.78515	0.78504[19]	1.3595	1.3594
propan-1-ol	Merck	99.8 %	0.79951	0.79960[16]	1.3833	1.3837
butan-1-ol	Merck	99.8 %	0.80575	0.80575[18]	1.3974	1.3973
pentan-1-ol	Fluka	99 %	0.81088	0.81077[18]	1.4089	1.4079
hexan-1-ol	Merck	98 %	0.81520	0.81522[18]	1.4166	1.4161

Results and Discussion

Experimental excess molar enthalpies at 298.15 K of the binary mixtures of 1,2-propanediol + alkan-1-ols (C_1-C_6) are reported in Table 2 and are graphically presented in Figure 1 in the form of H_m^E vs the mole fraction (x_1) of 1,2-propanediol. The results can be compared with those published by Karcht et al.[11] and Garcia and Paz Andrade.[25] For the mixtures of methanol and ethanol, our values are somewhat smaller, and for the other mixtures, our results are larger than theirs. The measurements conditions were the same except for pressure, which our measurement pressure was ambient pressure (815 kPa). Also, the types of instruments were different. Their measurements were carried out using a Tian-Calvet and LKB 2277 microcalorimeter, but we used a Parr 1455 macrocalorimeter.

The excess molar enthalpies are positive over the entire range of composition in all mixtures except in the system 1,2-propanediol + methanol, for which H_m^E is negative. This indicates that 1,2-propanediol interacts more strongly with methanol than with other alkanols. The H_m^E values increase as the chain length of the alkan-1-ol is increased. The heat of mixing of these systems may be influenced by factors such as

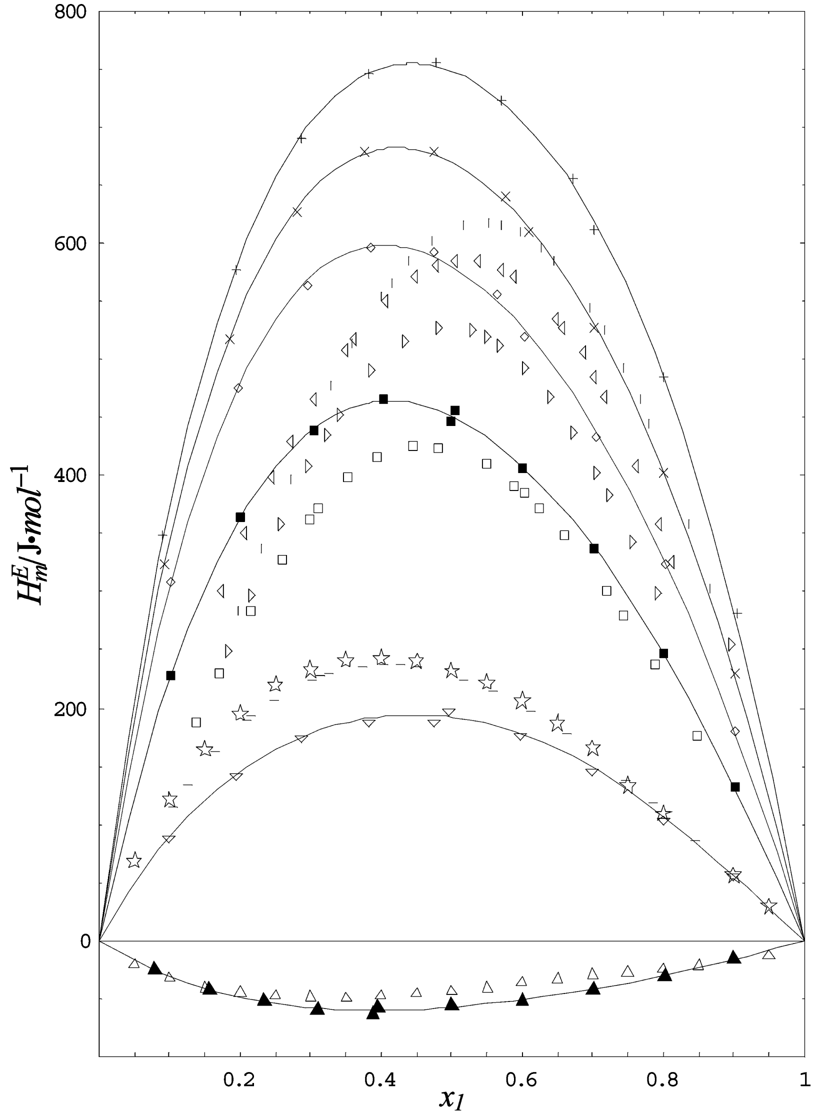

Figure 1. Experimental excess molar enthalpies of 1,2-propanediol (1) + alkan-1-ols (2) at 298.15 K. This work: ▲, methanol; ▽, ethanol; ■, propan-1-ol; ◇, butan-1-ol; ×, pentan-1-ol; +, hexan-1-ol. Karcht et al.:[12] △, methanol; ☆, ethanol. Garcia and Paz Andrade:[25] —, ethanol; □, propan-1-ol; right-facing unfilled triangle, butan-1-ol; left-facing unfilled triangle, pentan-1-ol; |, hexan-1-ol. Solid lines were calculated from the coefficient of eq 1 given in Table 3.

Table 2. Excess Molar Enthalpies, H^{E}_{m}, for the Binary Systems of 1,2-Propanediol + Alkan-1-ols at 298.15 K

x_1	$H^E_m/J\cdot mol^{-1}$ exp	$(\delta H^E_m)^a$	x_1	$H^E_m/J\cdot mol^{-1}$ exp	$(\delta H^E_m)^a$
		1,2-propandiol (1) + methanol (2)			
0.0787	−25.3	0.0	0.4998	−56.7	1.2
0.1565	−42.0	0.5	0.6004	−51.8	−0.3
0.2336	−53.2	0.1	0.7015	−42.5	−0.3
0.3099	−59.8	−0.8	0.8026	−31.1	−0.5
0.3885	−63.2	−2.4	0.9000	−16.4	0.7
0.3952	−58.6	2.2			
		1,2-propandiol (1) + ethanol (2)			
0.0997	89.90	0.3	0.4952	198.5	6.1
0.1948	143.6	0.0	0.5972	178.7	0.9
0.2875	175.7	0.5	0.6991	147.6	−1.3
0.3810	189.7	−1.4	0.8005	105.9	−1.0
0.4756	188.6	−4.9	0.9004	56.90	1.5
		1,2-propandiol (1) + propan-1-ol (2)			
0.1008	227.7	1.3	0.5049	455.9	5.6
0.2010	364.1	−1.2	0.6007	407.3	−0.8
0.3036	439.6	−0.9	0.7015	338.0	−0.2
0.4025	466.3	1.9	0.8011	246.3	0.3
0.5001	446.0	−5.7	0.9009	132.3	0.0
		1,2-propandiol (1) + butan-1-ol (2)			
0.1007	308.5	3.5	0.5644	556.7	6.4
0.1971	476.0	−1.5	0.6047	519.5	−5.0
0.2945	564.7	−3.1	0.7062	433.6	−3.9
0.3853	596.5	−1.0	0.8047	323.3	0.6
0.4758	592.2	3.7	0.9026	179.6	2.3
		1,2-propandiol (1) + pentan-1-ol (2)			
0.0914	324.0	3.7	0.5770	640.5	5.6
0.1843	518.5	−0.7	0.6082	609.2	−5.4
0.2821	627.9	−4.7	0.7027	527.5	−3.1
0.3755	678.2	0.8	0.8020	402.8	0.1
0.4761	680.0	3.7	0.9014	229.9	2.4
		1,2-propandiol (1) + hexan-1-ol (2)			
0.0905	348.7	4.1	0.5711	723.5	1.0
0.1929	577.3	−1.5	0.6718	656.4	5.8
0.2872	691.1	−3.6	0.7021	611.7	−8.3
0.3828	746.5	−0.3	0.7996	484.8	−3.1
0.4778	756.4	3.8	0.9040	281.6	5.3

[a] $(\delta H^E_m = H^E_{m,R-K} - H^E_{m,exp})$.

(i) the absorption of heat due to the dissociation of self-associated 1,2-propanediol or alkanols and (ii) the liberation of heat as a result of possible a hydrogen bonding interaction between 1,2-propanediol and alkanols. The actual value of H^E_m would depend on the relative strength of the two opposing effects.

To represent excess enthalpy as a function of temperature and composition at constant pressure, some kind of equation and thermodynamic model is required. Typical excess enthalpy models contain parameters that are fitted to experimental data on binary mixtures and then used to predict similar data in a multicomponent system. The excess molar enthalpies were correlated by the Redlich−Kister equation and two kinds of local composition models (Wilson and NRTL).

Redlich−Kister Equation. The excess molar enthalpies were correlated by the Redlich−Kister[22] equation

$$H^E_m/J\cdot mol^{-1} = x_1x_2\sum_{r=1} A_r(x_1 - x_2)^{r-1} \quad (1)$$

where x_1 is the mole fraction of 1,2-propanediol and A_r represents the adjustable parameters. The parameters in eq 1 were estimated by the least-squares fit method, and in each case, the optimum number of coefficients is ascertained from examination of the variation in the standard deviation, $\sigma(H^E_m)$, as given by

$$\sigma(H^E_m)/J\cdot mol^{-1} = [\sum_{k=1}^{n}\{H^E_{m,cal} - H^E_{m,exp}\}^2_k/(n-p)]^{1/2} \quad (2)$$

where n is the number of experimental points and p is the number of adjustable parameters A_r. The adjustable parameters A_r and standard deviation σ are given in Table 3.

Wilson Model. The model proposed by Wilson[23] is based on the concept of local composition. The explicit form of excess molar enthalpies of binary mixture developed through Wilson's model is

$$H^E_m/J\cdot mol^{-1} = x_1x_2\left(\frac{\lambda_{12}\Lambda_{12}}{x_1 + \Lambda_{12}x_2} + \frac{\lambda_{21}\Lambda_{21}}{\Lambda_{21}x_1 + x_2}\right) \quad (3)$$

where

$$\Lambda_{12} = \frac{V_2}{V_1}\exp\left(-\frac{\lambda_{12}}{RT}\right), \quad \Lambda_{21} = \frac{V_1}{V_2}\exp\left(-\frac{\lambda_{21}}{RT}\right)$$

λ_{12} and λ_{21} are model parameters and V_i is molar volume. The adjustable parameters λ_{21} and λ_{12} in the Wilson equation were obtained by a nonlinear least-squares fit and along with standard deviation, $\sigma(H^E_m)$ values are given in Table 4.

NRTL Equation. Renon and Prasunitz[24] modified Wilson's model for local mole fraction by introducing the constant α_{ij} to account for the nonrandomness of liquids solutions. The nonrandomness parameter, α_{ij}, is an empirical constant. According to the modification, the explicit form of excess molar enthalpies is

$$H^E_m/J\cdot mol^{-1} = -x_1x_2\left[\frac{Z_1N_1\alpha_{12}\Delta g_{21} - \Delta g_{21}G_{21}N_1 - Z_1x_2G_{21}\alpha_{12}\Delta g_{21}}{N_1^{\,2}} + \frac{Z_2N_2\alpha_{12}\Delta g_{12} - \Delta g_{12}G_{12}N_2 - Z_2x_1G_{12}\alpha_{12}\Delta g_{12}}{N_2^{\,2}}\right] \quad (4)$$

where $G_{21} = \exp(-\alpha_{12}\Delta g_{21}/RT)$, $N_1 = x_1 + x_2G_{21}N_2 = x_2 + x_1G_{12}$, $G_{12} = \exp(-\alpha_{12}\Delta g_{12}/RT)$, $Z_1 = (\Delta g_{21}/RT)G_{21}$, $Z_2 = (\Delta g_{12}/RT)G_{12}$; Δg_{12} and Δg_{21} are the two adjustable parameters remaining, because the nonrandomness parameters, α_{ij}, were taken to be constant for each binary mixtures. The adjustable

Table 3. Redlich−Kister Equation Fitting Coefficients, A_r, in Equation 1 and Standard Deviation, σ (H^E_m), in Equation 2 for 1,2-Propanediol (1) + Alkan-1-ols (2) at 298.15 K

system	A_1	A_2	A_3	A_4	$\sigma(H^E_m)$ $J\cdot mol^{-1}$
1,2-propanediol (1) + methanol (2)	−231.7	96.5	−52.6	−3.1	1.4
1,2-propanediol (1) + ethanol (2)	768.0	−155.8	63.0	−127.4	3.4
1,2-propanediol (1) + propan-1-ol (2)	1806.8	−576.8	287.9	−93.0	3.5
1,2-propanediol (1) + butan-1-ol (2)	2322.9	−727.7	580.0	−183.8	4.6
1,2-propanediol (1) + pentan-1-ol (2)	2679.8	−626.0	793.1	−249.2	4.6
1,2-propanediol (1) + hexan-1-ol (2)	2994.2	−455.9	1036.7	−225.4	5.6

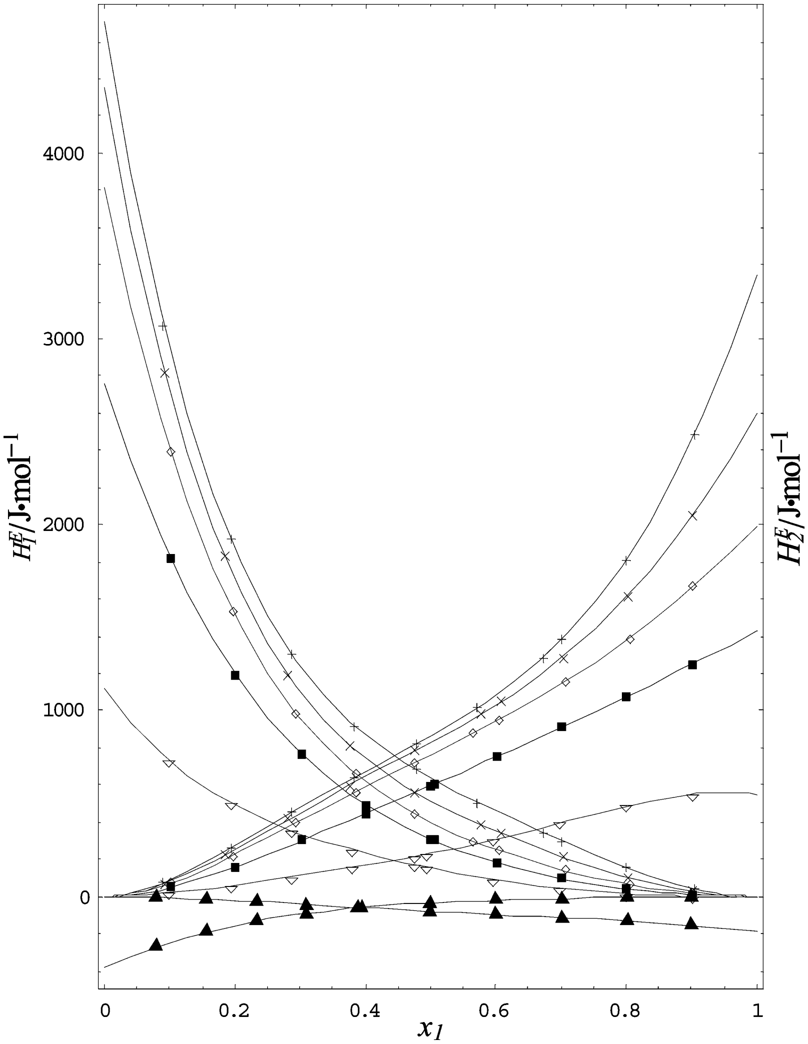

Figure 2. Excess partial molar enthalpies and their values at infinite dilution of 1,2-propanediol (1) + alkan-1-ols (2) at 298.15 K: ▲, methanol; ▽, ethanol; ■, propan-1-ol; ◇, butan-1-ol; ×, pentan-1-ol; +, hexan-1-ol. Solid lines were calculated from eq 5.

Table 4. Parameters λ_{21}, λ_{12}, Δg_{21}, Δg_{12}, and $\alpha_{12} = \alpha_{21}$ Used in Equations 3 and 4 for Calculating H_m^E Derived at 298.15 K

	(J·mol^{-1})					σ/(J·mol^{-1})	
system	λ_{21}	λ_{12}	Δg_{21}	Δg_{12}	α_{ij}	Wilson	NRTL
1,2-propanediol (1) + methanol (2)	−323.4	303.5	356.1	−466.0	0.96	1.6	4.0
1,2-propanediol (1) + ethanol (2)	124.6	887.9	857.0	207.7	0.96	3.4	3.5
1,2-propanediol (1) + propan-1-ol (2)	518.4	2456.9	2253.3	734.7	0.54	3.2	3.0
1,2-propanediol (1) + butan-1-ol (2)	991.6	3570.6	3106.5	1314.0	0.48	4.5	3.7
1,2-propanediol (1) + pentan-1-ol (2)	1659.3	4044.6	3521.2	1950.0	0.45	5.7	3.6
1,2-propanediol (1) + hexan-1-ol (2)	2754.4	4049.4	3894.5	2704.8	0.42	21.7	4.4

parameters (Δg_{21}, Δg_{12}) and the randomness parameter (α_{ij}) in the NRTL equation were obtained by a nonlinear least-squares fit and along with standard deviation, $\sigma(H_m^E)$, are given in Table 4.

The excess partial molar enthalpies, H_i^E, can be determined from excess molar enthalpies data using

$$H_i^E/\text{J·mol}^{-1} = H_m^E + (1 - x_i)(\partial H_m^E/\partial x_i)_{T,P} \quad (5)$$

where $(\partial H_m^E/\partial x_i)_{T,P}$ is calculated from eq 1 using the parameters in Table 3.

The excess partial molar enthalpy at infinite dilution $H_i^{E,\infty}$ can be determined from the following equation

$$H_i^{E,\infty}/\text{J·mol}^{-1} = (\partial H_m^E/\partial x_i)_{x_i=0,T,P} \quad (6)$$

The excess partial molar enthalpy at infinite dilution, $H_i^{E,\infty}$, appears to be of particular interest. In the limit of infinite dilution, solute−solute interactions disappear. Thus, the values of the excess partial molar enthalpies at infinite dilution provide insight into solute−solvent interactions. We can consider 1,2-propanediol at infinite dilution ($x_1 = 0$) in alcohols and of alcohols at infinite dilution ($x_1 = 1$) in 1,2-propanediol. All excess partial molar enthalpies, H_i^E, and their values at infinite dilution, $H_i^{E,\infty}$, are graphically represented in Figure 2. The H_1^E and H_2^E values of methanol are negative and for other alkanols are positive over the entire range of 1,2-propanediol composition.

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