

Activity Coefficients of Aroma Compounds in Model Solutions Simulating Apple Juice

Amalia A. Carelli, Guillermo H. Crapiste, and Jorge E. Lozano*

PLAPIQUI (UNS-CONICET), 12 de Octubre 1842, 8000 Bahía Blanca, Argentina

Infinite dilution activity coefficients (γ^∞) of several alcohols, aldehydes, and esters, considered to be the major components of apple aroma, were measured in model food solutions containing glucose, fructose, sucrose, and malic acid. Measurements were made by following the dilution exponential method, at temperatures ranging from 25 to 68 °C. Experimental data were correlated with the one-parameter Wilson model. The results reveal that γ^∞ values for a homologous series increase with the length of the carbon chain. γ^∞ values of lower alcohols and esters increase with temperature, while γ^∞ values for heavier aromas either decrease or remain practically constant with temperature. Apple juice solutes increase the activity coefficients of most of the aroma compounds studied.

INTRODUCTION

Manufacturing of apple juice concentrate involves aroma recovery, an operation by which volatile aromatic compounds contained in natural juice are stripped, together with a certain amount of water vapor, by thermal evaporation. This water solution of aroma compounds is further rectified in a packed column up to a concentration of 150–200-fold, condensed, and cooled to avoid vaporization of more volatile compounds.

To optimize and design the aroma recovery operation, information on thermodynamic properties is required. Apple aroma is a very dilute solution of esters, alcohols, and aldehydes (Carelli and Lozano, 1989), never exceeding levels of a few parts per million in the juice, and from a practical standpoint can be considered at infinite dilution.

Several authors (Lebert and Richon, 1984; Massaldi and King, 1973; Sorrentino et al., 1986) have reported measurements of activity coefficients of chemical compounds at infinite dilution in different aqueous electrolyte and nonelectrolyte solutions. However, information on the most important apple aroma components at different temperatures is scarce. It is well-known (Massaldi and King, 1973; Sorrentino et al., 1986; Chandrasekaran and King, 1972) that activity coefficients can be strongly affected by the type and concentration of soluble solids present in the solvent. The study of this effect in apple juice is difficult because natural juices are very complex mixtures in which the interactions among the many components is difficult to elucidate. To avoid this problem, the use of simplified model food systems has been proposed.

This study aims (1) to present data on the activity coefficients of apple aroma compounds at infinite dilution in solutions that simulate apple juice at different temperatures and (2) to correlate these experimental data with temperature by using some of the one-parameter models based on the Wilson, NRTL, or UNIQUAC equations.

EXPERIMENTAL PROCEDURES

Materials. Water was twice distilled. Aroma compounds were from Aldrich Chemical Co. and were used without further purification since gas chromatographic analysis failed to show any significant impurity. Sucrose, fructose, glucose, and malic acid (L-hydroxybutanedioic acid, 95%) were of grade I from Sigma

Chemical Co. Model system composition was as follows: sucrose, 26.8 g/L; fructose 57.9 g/L; glucose 20.7 g/L; and malic acid 0.42% (Babsky et al., 1986).

Experiment and Procedure. Activity coefficients of aroma components at infinite dilution were determined by following the exponential dilution method, also called the dynamic method (Leroi et al., 1977). This method is based on the stripping of a solute from a solvent by a constant flow of inert gas, N₂ in this study. The variation of solute concentration in the carrier gas is measured by gas-liquid chromatography. Experimental setup and procedure have been described in detail by Leroi et al. (1977). The equilibrium cell used in this work is shown in Figure 1. Cell temperature was measured with a digital thermometer (Fluke Model 2190A) and controlled with a thermostatic water bath (Cole Parmer, Model 1268-20). Fluctuations of no more than 0.1 °C were observed.

Cell pressure was also measured with a U-tube glass manometer with water as manometric fluid. Atmospheric pressure, also required for calculations, was measured with a Forten-type barometer. The accuracy in determination of pressure was ± 0.1 mmHg.

Model solution was prepared by careful mass dilution and stirring, the solutes being weighed to 10⁻⁴ g. Aroma components were injected directly into the cell, containing 40 mL of model solution. The amounts of esters, aldehydes, and alcohols, injected with a chromatographic syringe, were 10, 20, and 40 μ L, respectively.

The sample valve (a six-port external volume Valco valve, heated at 260 °C with heated valve enclosure) injected sample (1.0 cm³ of esters and aldehydes and 2.0 cm³ of alcohols) into the gas chromatograph (Varian 3700). The chromatograph was equipped with a data processing station (Vista 402), an FID detector, and a 1 m \times 3.2 mm stainless steel GLC column packed with Porapak-Q. Carrier gas (N₂, minimum purity 99.5%) flow was 28 cm³/min, and temperature conditions were as follows: FID, 280 °C; column, 180 °C.

The working equation for γ^∞ calculation was derived from the equilibrium conditions and mass balance in the cell (Leroi et al., 1977) and was valid for volatile solvent, without taking into account the solute distribution correction (Duhem and Vidal, 1978)

$$\gamma^\infty = [(X(t)/Y(t)) + 1](P_{sv}/P_s) \quad (1)$$

$$X(t) = \ln(S_i/S_{i,0})$$

$$Y(t) = \ln[1 - (P_s DP_{sv} t / (P_i - P_{sv}) NRT)]$$

* To whom correspondence should be addressed.

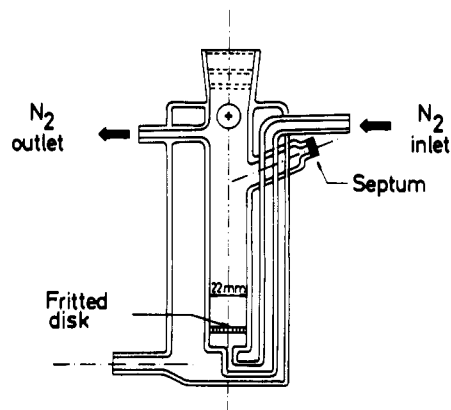


Figure 1. Sketch of the equilibrium cell.

Table I. Antoine Constants^a of Aroma Compounds

compound	A	B	C	T range, K	ref
ethyl acetate	14.1366	2790.50	-57.15	260-385	b
ethyl butyrate	13.9837	3127.60	-60.15	288-432	b
butyl acetate	14.1686	3151.09	-69.15	295-435	b
ethyl isobutyrate	14.8788	3515.11	-40.76	240-380	d
ethyl valerate	15.3818	4074.13	-40.59	280-435	c
pentyl acetate	15.3830	4103.45	-40.94	315-425	c
trans-2-hexenal	15.3857	4007.22	-47.56	280-415	c
hexanal	15.4971	3952.08	-38.12	280-415	c
benzaldehyde	14.3351	3748.62	-66.12	300-460	b
ethanol	16.8969	3803.98	-41.68	270-369	b
propanol	15.5285	3166.38	-80.15	285-400	b
butanol	15.2010	3137.02	-94.43	288-404	b
2-methyl-1-butanol	14.2558	2752.19	-116.30	307-402	b
hexanol	16.0848	4055.45	-76.49	308-430	b
model solution	16.7154	4076.80	-36.46	298-339	e

^a Antoine equation: $\ln P = A - B/(C + T)$; P = vapor pressure, kPa; T = temperature, K. ^b Reid et al. (1977). ^c Dreisbach (1952). ^d Stull (1947). ^e Taylor and Rowlinson (1955).

where N is the total moles of solvent in the dilution cell; D is the flow of carrier gas through the cell at T , cm³/min; R is the gas constant; T is the cell temperature, K; S_i is the area of solute peak, P_t is the total pressure in the cell; P_{sv} is the vapor pressure of solvent at T , kPa; P_s is the vapor pressure of solute at T , kPa; and t is time, min.

Vapor pressures of apple aroma compounds (P_s) were calculated with the Antoine equation. The Antoine constants were obtained from Reid et al. (1977) or calculated from nonlinear regression of P_s vs T data reported by Dreisbach (1952) and Stull (1947) and are listed in Table I. Vapor pressure of solvent was estimated from nonlinear regression of P_{sv} vs T data of glucose solutions (Taylor and Rowlinson, 1955), a reasonable assumption taking into account the concentration and composition of the model solution (Crapiste and Lozano, 1988). The γ^∞ values were obtained from linear regression of $X(t)$ vs $Y(t)$ data.

A more simplified expression of eq 1 has been previously used for γ^∞ calculation of volatiles in food model systems (Lebert and Richon, 1984; Sorrentino et al., 1986) but failed to represent the solvent volatility.

Four determinations of activity coefficients γ^∞ were performed and averaged for each temperature and solute analyzed. Ethyl acetate in water was used in this work to check the accuracy of the experimental procedure. Agreement of activity coefficients obtained in this study with those reported in the literature for the ethyl acetate-water system (Sorrentino et al., 1986; Chandrasekaran and King, 1972) was within 0.5%.

Table II. Activity Coefficient at Infinite Dilution of Aroma Compounds in Model Solution and Dependence of the Wilson-Hiranuma Parameter C_{12}^∞ on Temperature

compound	T, K	γ^∞	a	$b \times 10^3$	r
ethyl acetate	298.13	86.2 ± 1.9	2.7038	-5.076	-0.998
	303.15	97.3 ± 1.1			
	323.19	101.8 ± 2.6			
	337.65	108.3 ± 6.5			
ethyl butyrate	298.18	1132 ± 66	2.6451	-6.093	-0.995
	308.37	1202 ± 42			
	323.09	1251 ± 27			
	329.65	1233 ± 65			
	338.29	1121 ± 62			
butyl acetate	298.33	1201 ± 46	2.6653	-5.8744	-0.996
	308.26	1220 ± 80			
	323.81	1270 ± 74			
	337.93	1125 ± 83			
ethyl isobutyrate	298.22	1121 ± 90	2.3262	-5.1804	-0.986
	308.29	1220 ± 76			
	323.52	1255 ± 84			
	338.17	1062 ± 32			
ethyl valerate	315.37	5145 ± 373	1.8476	-3.4605	-0.999
	323.68	4743 ± 239			
	331.55	4372 ± 167			
	337.77	4028 ± 227			
pentyl acetate	315.63	5478 ± 384	1.6714	-3.4605	-0.999
	323.53	4875 ± 272			
	330.74	4457 ± 360			
trans-2-hexenal	338.76	3993 ± 557	2.3196	-4.2050	-0.983
	314.64	687 ± 42			
	323.40	743 ± 66			
	331.00	675 ± 56			
hexanal	340.13	658 ± 19	2.1081	-4.2163	-0.960
	298.56	1310 ± 95			
	308.39	1566 ± 110			
	323.18	1379 ± 80			
benzaldehyde	338.56	1132 ± 60	2.913	-5.6913	-0.970
	323.35	559 ± 30			
	333.01	636 ± 66			
ethanol	341.09	593 ± 77	2.3970	-1.9000	-0.976
	312.24	4.79 ± 0.34			
	323.93	5.55 ± 0.48			
propanol	339.18	5.72 ± 0.54	2.460	-2.584	-1.000
	311.70	19.96 ± 1.70			
	323.36	20.19 ± 0.90			
butanol	338.13	20.46 ± 0.64	2.7904	-3.9906	-0.998
	307.95	67.4 ± 4.7			
	323.26	71.6 ± 3.0			
2-methyl-1-butanol	337.71	70.1 ± 2.7	2.9278	-4.7647	-0.999
	308.32	214.8 ± 16.1			
	323.24	199.8 ± 13.7			
hexanol	338.07	187.1 ± 14.9	2.4268	-3.9153	-0.999
	314.41	1047 ± 131			
	323.49	985 ± 47			
	337.56	866 ± 35			

$${}^a C_{12} = a + bT.$$

THEORETICAL BACKGROUND

Comparisons between predicted values from one-parameter Wilson, NRTL, and UNIQUAC equations were also performed.

Wilson Equation. The Wilson expression for a binary mixture with the solute at infinite dilution is (Kruming et al., 1980)

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

where

Table III. Performance of Different Models To Represent Activity Coefficients at Infinite Dilution

compound/model	% error in γ^∞	$C_{12} = a + bT$		
		a	$b \times 10^3$	r
ethyl acetate				
Wilson-Hiranuma	1.38	2.7038	-5.076	-0.998
NRTL	1.38	2.2519	-3.037	-0.997
UNIQUAC	1.37	2.1813	-2.593	-0.995
trans-2-hexenal				
Wilson-Hiranuma	3.17	2.3196	-4.205	-0.983
NRTL	3.18	1.9076	-2.010	-0.963
UNIQUAC	3.18	1.7464	-1.843	-0.942
butanol				
Wilson-Hiranuma	1.72	2.7904	-3.9906	-0.998
NRTL	1.81	2.3888	-2.6237	-0.997
UNIQUAC	1.72	2.0146	-1.1738	-0.984

$$A_{ji} = 1 - (V_j/V_i) \exp[-(g_{ji} - g_{ii})/RT] \quad (3)$$

and

$$g_{21} = g_{12} = (g_{11}g_{22})^{1/2}(1 - c_{12}) \quad (4)$$

In these equations $V_{i,j}$ is the liquid molar volume of components i and j (m^3/mol), g_{ii} is a constant proportional to the energy of interaction between molecules of species i , and C_{12} is a parameter to be determined experimentally. Hiranuma and Honma (1975) proposed that for systems where the infinite dilution activity coefficient is on the order of 10 or greater values of

$$g_{ii} = -\Delta U_i/3 \quad (5)$$

and

$$V_j/V_i = 1 \quad (6)$$

can be used, where ΔU_i is the energy of vaporization of the i th component (J/mol), and can be calculated from

$$\Delta U_i = [RT^2 d(\ln P_i^0)/dT] - RT \quad (7)$$

where P_i^0 is the vapor pressure of the i th component.

NRTL Equation. The activity coefficient data have been also correlated by means of the NRTL equation, a one-parameter expression valid at infinite dilution that can be expressed, after the nonrandomness parameter α_{12} is set equal to 0.4 and on the basis of $V_2 > V_1$, as

$$\ln \gamma_2^\infty = (V_2/V_1 0.4RT) \{ (g_{12} - g_{22}) + \exp[-(g_{21} - g_{11})/RT] (g_{12} - g_{11}) \} \quad (8)$$

where the parameters g_{ij} were assumed to be given by eq 4 and the parameters g_{ij} by the expression

$$g_{ii} = -0.08\Delta U_i/q_{ij} \quad (9)$$

where

$$q_{12} = 1 \quad q_{21} = (V_2/V_1)^{1/2} \quad (10)$$

UNIQUAC Model. The UNIQUAC model, derived from statistical mechanical arguments (Abrams and Prausnitz, 1975), was expressed as the combination of a combinatorial term, which takes into account liquid-phase nonidealities due to differences in molecular size and shape, and a residual term, which takes into account nonidealities due to intermolecular interactions. When activity coefficients are calculated at infinite dilutions, UNIQUAC equations can be simplified considerably to give the expressions

$$\ln \gamma_{c2}^\infty = \ln (r_2/r_1) + 5q_2 \ln (q_2 r_1 / r_2 q_1) + 5(r_2 - q_2) - (r_2 - 1) - r_2/r_1 [5(r_1 - q_1) - (r_1 - 1)] \quad (11)$$

$$\ln \gamma_{r2}^\infty = q_2(1 + g_{12}/RT - g_{22}/RT) - \exp[-(g_{12} - g_{11})/RT] \quad (12)$$

$$\ln \gamma_2^\infty = \ln \gamma_{c2}^\infty + \ln \gamma_{r2}^\infty \quad (13)$$

where

$$g_{ii} = -0.5\Delta U_i/q_i$$

On the other hand, g_{ij} is given by eq 4; r_i and q_i are pure components structural parameters, obtained from Gmehling et al. (1982).

RESULTS AND DISCUSSION

Table II shows the experimental values of activity coefficients at infinite dilution obtained in this study. Tabulated data are average values of four replicated runs. Confidence intervals, with a significance level $\alpha = 5\%$, are also reported. Error analysis indicated that, on the average, the deviation in measured activity coefficients at infinite dilution was less than 7%, with the exception of those for benzaldehyde and pentyl acetate at high temperatures.

The C_{12} parameter of the Wilson-Hiranuma, NRTL, and UNIQUAC models was determined from the experimental values of γ^∞ at each temperature by using a Newton-Raphson iterative procedure. It was found that effect of temperature on C_{12} can be represented by a linear relationship. After correlation, model predictions were compared with experimental data. Some results are given in Table III. No significant differences between γ^∞ predictions of the above models were observed. However, although both NRTL and UNIQUAC equations predicted values that were in good agreement with measurements, results indicated that the Wilson equation (after Hiranuma) gave a better correlation of C_{12} as a function of temperature. Similar results were obtained for the other solutes but are not shown to avoid repetition. Thus, the Wilson-Hiranuma model was retained for this study and the corresponding C_{12} parameters are also given in Table II.

Figures 2, 3, and 4 also compare experimental data with those predicted by the Wilson-Hiranuma equation for alcohols, esters, and aldehydes, respectively. The results indicated that, for a homologous series, the activity coefficients at infinite dilution increase with the length of the carbon chain, particularly in the case of esters and alcohols. The behavior is in agreement with the trends obtained in previous studies by Kieckbush and King (1979), Lebert and Richon (1984), and Sorrentino et al. (1986).

It is also noteworthy that activity coefficients of lower alcohols and esters increased with temperature. Contrarily, heavier aromas reduced its values when the temperature was increased. On the other hand, it appears that γ^∞ values for ethyl butyrate, butyl acetate, ethyl isobutyrate, and butanol remained practically constant with temperature in the range analyzed.

The observed behavior for alcohols is in agreement with γ^∞ in pure water calculated from Pierotti et al. (1959), which predicted increasing values of γ^∞ with temperature for ethanol, propanol, and butanol and a decrease in γ^∞ for hexanol. Kieckbush and King (1979) studied the effect of temperature on partition coefficients of n -acetates in water and polysaccharide solutions. The authors reported that partition coefficients for the series methyl acetate-

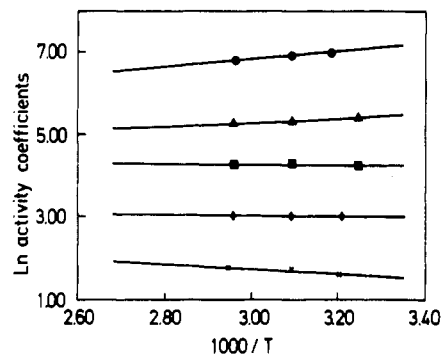


Figure 2. Variation of the activity coefficient at infinite dilution for various alcohols with respect to temperature. (x) Ethanol; (♦) propanol; (■) butanol; (▲) 2-methyl-1-butanol; (●) hexanol. Full lines represent Wilson-Hiranuma predictions.

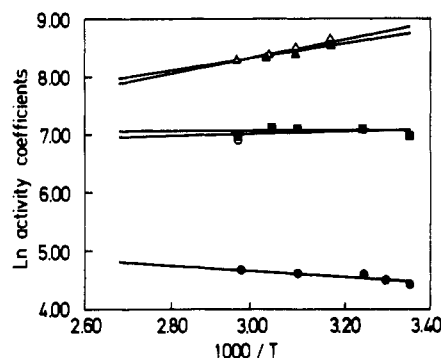


Figure 3. Infinite dilution activity coefficient of several esters as a function of temperature. (●) Ethyl acetate; (■) ethyl butyrate; (○) ethyl isobutyrate; (▲) ethyl valerate; (△) pentyl acetate. Full lines represent Wilson-Hiranuma predictions.

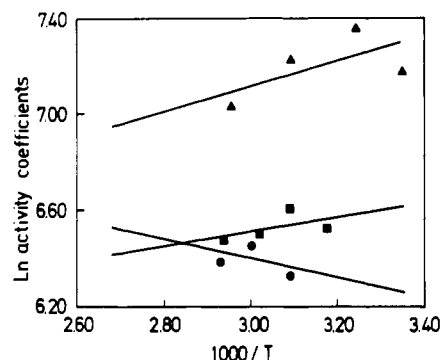


Figure 4. Variation of activity coefficient at infinite dilution of the more important aldehydes of apple aroma, with temperature. (▲) Hexanal; (■) *trans*-2-hexenal; (●) benzaldehyde. Full lines represent Wilson-Hiranuma predictions.

pentyl acetate increase with temperature in the range 25–40 °C. The results of the present paper indicate a similar trend for ethyl and butyl acetate but an opposite behavior for pentyl acetate at higher temperatures. On the other hand, Table II suggests that activity coefficients for ethyl butyrate, ethyl isobutyrate, and butyl acetate also increase slightly up to about 60 °C and then fall off. According to that, it appears that a change of behavior could be expected at higher temperatures. Since the slope of $\ln \gamma^\infty$ vs $1/T$ is associated with the excess molar enthalpy, this would imply a change from exothermic to endothermic mixtures. It can be seen that some discrepancies were also found for aldehydes. However, the small differences might be the result of experimental errors, so that a complete study on the effect of temperature on γ^∞ of esters and aldehydes in water should be done to clarify this point.

Experimental values of activity coefficients at 20–25 °C

Table IV. Activity Coefficient at Infinite Dilution of Aroma Compounds in Pure Water and Model Solution

compound	activity coefficients, γ^∞	
	water	model solution
ethyl acetate	70 ^a –77 ^b	86
ethyl butyrate	1153 ^a	1178
butyl acetate	1236 ^a	1246
ethyl valerate	4883 ^a	6872
pentyl acetate	5289 ^a	7889
<i>trans</i> -2-hexenal	1162 ^a	687
hexanal	1012 ^a –1000 ^c	1310
ethanol	3.37 ^a –3.55 ^d	4.79
propanol	11.4 ^a –11.2 ^d	19.96
butanol	42.9 ^a –43 ^d	67.4
hexanol	676 ^a –645 ^e	1047

^a Calculated from Pierotti et al. (1959). ^b Sorrentino et al. (1986). ^c Chandrasekaran and King (1972). ^d Sorrentino et al. (1986). ^e Lebert and Richon (1984).

of some aroma compounds diluted in pure water and those calculated from the equation of Pierotti et al. (1959) are presented in Table IV. They are compared with γ^∞ results obtained in this work evaluated by using the Wilson equation. It can be seen that γ^∞ values in model solutions of most of the volatiles studied are higher than those reported in pure water. As a general conclusion, it appears that the presence of apple juice solutes leads to an increase in activity coefficients of aroma components.

GLOSSARY

- A, B, C* = Antoine constants
A_{ij} = parameter in Wilson equation
C₁₂ = parameter in Wilson, NRTL, and UNIQUAC models
D = flow of carrier gas through cell at *T*, cm³/min
g_{ij} = parameter in Wilson, NRTL, and UNIQUAC models
n = moles of solutes
N = total moles of solvent in dilution cell
P_t = total pressure in the cell, kPa
P = vapor pressure, kPa
q_{ij} = parameter in NRTL equation
R = gas constant, 8.3143 J/mol·K
S_i = area of solute peak
t = time, min
T = cell temperature, K
 ΔU_i = energy of vaporization of *i*th component, J/mol
V_i = liquid molar volume of components *i*, m³
X(t), Y(t) = functions defined in eq 1

Greek letters

- γ^∞ = activity coefficient at infinite dilution

Subscripts

- i, j* = components
0 = initial time
s = solute
sv = solvent
c = combinatorial
r = residual

LITERATURE CITED

- Abrams, D. S.; Prausnitz, J. M. Statistical Thermodynamics of Liquid Mixtures: A New Expression for the Excess Gibbs Energy of Partly or Completely Miscible Systems. *AIChE J.* 1975, 21, 116–128.

- Babsky, N.; Toribio, J. L.; Lozano, J. E. Influence of Storage on the Composition of Clarified Apple Juice Concentrate During Storage. *J. Food Sci.* 1984, 49, 889-892.
- Carelli, A.; Lozano, J. E. Apple Aroma from Argentina: Quality Evaluation by Capillary Gas Chromatography. *HRC & CC* 1989, 12, 488-490.
- Chandrasekaran, S. K.; Judson King, C. Multicomponent Diffusion and Vapor-Liquid Equilibria of Dilute Organic Components in Aqueous Sugar Solutions. *AIChE J.* 1972, 18, 513-526.
- Crapiste, G. H.; Lozano, J. E. Effect of concentration and Pressure on the Boiling Point Rise of Apple Juice and Related Sugar Solutions. *J. Food Sci.* 1988, 53, 865-895.
- Dreisbach, R. R. *Pressure-Volume-Temperature Relationships of Organic Compounds*, 3rd ed.; Handbook Publishers: 1952.
- Duhem, P.; Vidal, J. Extension of the Dilutor Method to Measurement of High Activity Coefficients at Infinite Dilution. *Fluid Phase Equilib.* 1978, 2, 231-235.
- Gmehling, J.; Rasmussen, P.; Fredenslund, A. Vapor-Liquid Equilibria by UNIFAC Group Contribution, Revision and Extension 2. *Ind. Eng. Chem. Process Des. Dev.* 1982, 21, 118-127.
- Hiranuma, M.; Honma, K. Estimation of Unlike-Pair Potential Parameter in Single Parameter Wilson Equation. *Ind. Eng. Chem. Process Des. Dev.* 1975, 14, 221-226.
- Kieckbush, T. G.; Judson King, C. Partition Coefficients for Acetates in Food Systems. *J. Agric. Food Chem.* 1979, 27, 504-507.
- Kruming, A. E.; Rastogi, A. K.; Rusak, M. E.; Tassios, D. Prediction of Binary Vapor-Liquid Equilibrium from One Parameter Equations. *Can. J. Chem. Eng.* 1980, 58, 663-669.
- Lebert, A.; Richon, D. Infinite Dilution Activity Coefficients of n-Alcohols as a Function of Dextrin Concentration in Water-Dextrin Systems. *J. Agric. Food Chem.* 1984, 32, 1156-1161.
- Leroi, J. C.; Masson, J. C.; Renon, H.; Sannier, H. Accurate Measurement of Activity Coefficients at Infinite Dilution by Inert Gas Stripping and Gas Chromatography. *Ind. Eng. Chem. Process Des. Dev.* 1977, 16, 139-144.
- Massaldi, H. A.; Judson King, C. Simple Technique to Determine Solubilities of Sparingly Soluble Organics: Solubility and Activity Coefficients of d-Limonene, n-Butylbenzene and n-Hexyl Acetate in Water and Sucrose Solutions. *J. Chem. Eng. Data* 1973, 18, 393-397.
- Pierotti, G. J.; Deal, C. H.; Den, E. L. Activity Coefficients and Molecular Structure. *Ind. Eng. Chem.* 1959, 51, 95-102.
- Reid, R.; Sherwood, T.; Prausnitz, J. *The Properties of Gases and Liquids*; McGraw-Hill: New York, 1977.
- Sorrentino, F.; Voilley, A.; Richon, D. *Sci. Aliments* 1984, 4, 105-110.
- Sorrentino, F.; Voilley, A.; Richon, D. Activity Coefficients of Aroma Compounds in Model Food Systems. *AIChE J.* 1986, 32, 1988-1993.
- Stull, D. R. Vapor Pressure of Pure Substances. *Ind. Eng. Chem.* 1947, 39, 517-540.
- Taylor, J. B.; Rowlinson, J. S. The Thermodynamic Properties of Aqueous Solutions of Glucose. *J. Trans. Faraday Soc.* 1955, 51, 1183-1192.

Received for review November 26, 1990. Accepted June 3, 1991.

Registry No. Ethyl acetate, 141-78-6; ethyl butyrate, 105-54-4; butyl acetate, 123-86-4; ethyl isobutyrate, 97-62-1; ethyl valerate, 539-82-2; pentyl acetate, 628-63-7; *trans*-2-hexenal, 6728-26-3; hexanal, 6728-26-3; benzaldehyde, 100-52-7; ethanol, 64-17-5; propanol, 71-23-8; butanol, 71-36-3; 2-methyl-1-butanol, 137-32-6; hexanol, 111-27-3.