Glossary

B	gas phase second virial coefficient, cm^3 mol^{-1}
P^0	pure component vapor pressure, mmHg
R	gas constant
T	absolute temperature, K
t	system temperature, °C
V	pure component liquid molar volume, cm^3 mol^{-1}
x	mole fraction in liquid phase
y	mole fraction in gas phase
γ	activity coefficient
π	system pressure, mmHg

Subscripts

1	more volatile component
2	less volatile component

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Excess Enthalpy, Volume, and Gibbs Free Energy and Viscosity of Ethyl Acetate–Methyl Cellosolve Mixtures

Badri S. Chandak, Govind D. Nageshwar,* and Purshotam S. Mene
Laxminarayan Institute of Technology, Nagpur University, Nagpur, India

Molar excess enthalpies of mixing at 48.2 °C are measured for the ethyl acetate–methyl cellosolve system using an isothermal phase change calorimeter. The molar excess volume is computed from density measurements at 40 °C. The molar excess Gibbs free energies are calculated from isothermal vapor–liquid equilibrium data obtained in a circulation still at 70, 80, and 90 °C. Complete isobaric vapor–liquid equilibrium data at 1 atm pressure are also reported. The activity coefficient data are corrected for vapor phase nonideality, tested for thermodynamic consistency, and correlated by the Wilson equation. The kinematic viscosity of binary mixtures is measured at 40, 50, 60, and 70 °C using an Ostwald viscometer. The viscosity data are correlated by McAllister and excess function models.

The present investigation forms part of a continuing study of excess thermodynamic properties of binary liquid mixtures consisting of methyl cellosolve as one of the components.

Experimental Procedures

Materials. Analytical grade ethyl acetate and methyl cellosolve from British Drug House Co of India were dried over silica and further purified by distillation in a 35-mm glass column packed with 3-mm ceramic insulation beads to a height of 1 m. The column was run at total reflux for 30 min, and the low boiling impurities were drawn off at a very slow rate of rejects. The overhead products were analyzed at various intervals by density and refractive index measurements and the "heart cut" of desired purity was collected for further investigations. The pure component boiling points reported here were obtained in the equilibrium still used in this study. Table I lists the physical properties of the chemicals used.

Apparatus. Calorimetric Measurements. An isothermal phase change calorimeter similar to one described by Dainton (*4*) and further modified by Lakhanpal (*13*) and calibrated in our laboratories by Kalasy (*10*) was used for excess enthalpy measurements at 48.2 °C. The change in the volume of a dilatometric fluid (cetyl alcohol) on phase transformation is a measure of heat change in the mixing process. The conventional bulb breaking method was adopted for mixing the components. The calorimeter was thermostated at 48.2 ± 0.002 °C and the accuracy of any individual measurement of heat change was better than ±0.02 cal mol^{-1}.

Vapor-Liquid Equilibria. The Othmer type of recirculation still (*25*) modified by Mainkar and Mene (*16*) was used to measure the equilibrium data. The still has three separate heating units for the equilibrium chamber to allow for a close approach to adiabatic conditions. For isobaric data the pressure in the still was controlled to 760 ± 1 mmHg using dried air. Two surge tanks between the still and the manostat served to damp out the pressure fluctuations. The data were measured with air present in the condensate returning to the equilibrium still. For isothermal VLE data the still was filled with 230 mL of mixture and heated to boiling. The pressure in the manostat was adjusted for the desired isothermal conditions. After the recirculation had stabilized the still was operated for 2–3 h. The pressure was controlled by a needle valve and indicated by a mercury manometer

Table I. Physical Properties of the Chemicals Used

Properties		Ethyl acetate	Methyl cellosolve
Mol wt		88.10	76.09
Normal bp, °C			
Exptl		77.10	124.5
Lit.		77.15 (*14*)	124.5 (*14*)
Density at 40 °C			
Exptl		0.8753	0.9462
Lit.		0.8759 (*30*)	—
Refractive Index at 30 °C			
Exptl		1.3685	1.3991
Lit.		1.3701[a] (*30*)	1.4004[b] (*14*)
Absolute Viscosity at 40 °C (*Cp*)			
Exptl		0.3481	1.1917
Lit.		0.3600	—
Heat of vaporization at normal bp, cal g-mol^{-1}		87.63 (*15*)	135.0 (*15*)
Antoine constants (70–125 °C)	*A*	7.09808 (*31*)	8.30770 (*5*)
	B	1238.71	2157.00
	C	217.00	273.16
Vapor Pressure at 70 °C (mmHg)			
Exptl		596.00	105.00
Lit.		596.30 (*30*)	105.19 (*5*)

[a] At 25 °C. [b] At 26 °C.

Table II. Molar Excess Enthalpy and Volume for Ethyl Acetate (1)–Methyl Cellosolve (2)

x_1	H^E, cal mol^{-1}	x_1	V^E, mL mol^{-1}	x_1	V^E, mL mol^{-1}
0.1054	30.67	0.0495	−0.0150	0.5913	0.0471
0.2168	65.98	0.1105	−0.0349	0.6603	0.0499
0.3122	95.14	0.2273	−0.0330	0.6928	0.0489
0.4191	117.21	0.3169	−0.0154	0.7558	0.0437
0.5172	127.20	0.4072	0.0225	0.7961	0.0390
0.6015	125.14	0.4502	0.0346	0.8474	0.0312
0.7049	114.73	0.4982	0.0405	0.9112	0.0202
0.8060	85.92	0.5586	0.0447	0.9436	0.0128
0.8969	60.18				

	Constants in Eq 2			
	C_1	C_2	C_3	RMSD
H^E	496.736	173.847	−14.499	3.88
V^E	0.150	0.330	−0.333	0.0038

to ±1 mmHg. An enclosed type mercury-in-glass thermometer with an accuracy of ±0.1 °C was used for temperature measurements. Equilibrium samples were analyzed by the specific gravity method using pycnometers of 15 mL capacity. All weight measurements were made on a 'Owa labor' analytical balance with a sensitivity of ±0.1 mg.

Viscosity Measurements. Viscosity of binary mixtures of varying compositions was determined with the Ostwald viscometer calibrated against the known viscosity of water at different temperatures. The viscometer was kept in a thermostatically controlled bath for about an hour and the efflux times were measured by visual observations of the meniscus at least three times for each composition agreeing to ±0.5 s.

The limits of error in the experimental data are estimated as follows: enthalpy, ±0.02 cal mol^{-1}; composition, ±0.0002 mole fraction; pressure, ±1 mmHg; temperature, ±0.1 °C.

Results and Discussion

Experimental values of molar excess enthalpy of mixing at 48.2 °C for various mole fractions of ethyl acetate are listed in Table II and presented graphically in Figure 1. The system is endothermic in nature with a maximum heat of mixing of 125.14 cal mol^{-1} at 60 mole % ethyl acetate. The excess molar volume for the system is calculated from the density measurements at 40 °C through the equation.

$$V^E = V - x_1V_1 - x_2V_2 \qquad (1)$$

The pure component liquid molar volumes V_1 and V_2 are obtained from their densities at 40 °C. The data are reported in Table II and presented in Figure 1. The method of least squares was used to fit the results of excess enthalpy and volume with an equation of the form,

$$X^E = x_1x_2 \sum_{q=1}^{n} C_q(1 - 2x_2)^{q-1} \qquad (2)$$

and the constants along with the root-mean-square deviations of the estimate are reported in Table II.

The results of isothermal VLE studies are summarized in Table III. The last column contains values of the excess molar Gibbs free energy.

$$G^E = RT(x_1 \ln \gamma_1 + x_2 \ln \gamma_2) \qquad (3)$$

Table IV reports the equilibrium data at 1 atm pressure. The liquid phase activity coefficients are evaluated from the vapor–liquid equilibrium results using the equation (*7*),

$$\gamma_i = \frac{\pi y_i}{P_i^0 x_i} \exp\left[\frac{(P_i^0 - \pi)(V_i - \beta_i)}{RT}\right] \qquad (4)$$

The vapor pressures of the pure components were calculated over the temperature ranges of the binary as well at their normal boiling points by Antoine (*1*), Miller (semireduced) (*18*), Miller–Erpenbeck (*18*), RPME, and RPMH version of Riedel–Plank–Miller

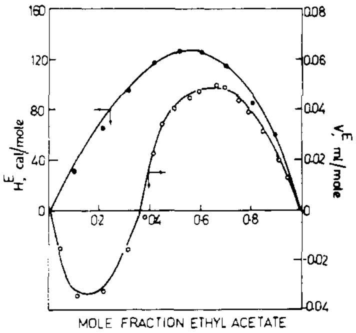

Figure 1. Excess enthalpy (48.2 °C), and volume (40 °C) for ethyl acetate (1)–methyl cellosolve (2).

Table III. Isothermal Vapor-Liquid Equilibrium Data for Ethyl Acetate (1)-Methyl Cellosolve (2)

P, mmHg	x_1	y_1	γ_1	γ_2	G^E
		At 70 °C			
105.0	0.0000	0.0000			
122.0	0.0147	0.1215	1.7451	1.0326	21.13
136.0	0.0270	0.2150	1.8724	1.0404	37.76
170.0	0.0720	0.4100	1.6782	1.0267	42.06
230.0	0.1480	0.5925	1.5814	1.0350	66.19
248.0	0.1602	0.6450	1.5226	1.0373	72.12
303.0	0.2610	0.7280	1.4440	1.0429	86.51
321.0	0.2895	0.7505	1.3841	1.0260	76.55
388.0	0.4145	0.8265	1.3139	1.0676	103.28
412.0	0.4740	0.8501	1.2527	1.0881	103.07
449.0	0.5701	0.8750	1.1653	1.2062	114.39
486.0	0.6650	0.9040	1.1142	1.2828	105.87
515.0	0.7855	0.9330	1.0295	1.4781	72.74
560.0	0.8960	0.9645	1.0099	1.7499	46.56
596.0	1.0000	1.0000			
		At 80 °C			
158.0	0.0000	0.0000			
167.0	0.0075	0.0660	1.7288	0.9952	0.35
170.0	0.0110	0.0845	1.6351	0.9921	−1.68
201.5	0.0440	0.2900	1.6595	0.9422	−24.90
227.0	0.0640	0.3595	1.5908	0.9752	4.35
287.0	0.1215	0.5300	1.5560	0.9597	12.34
330.0	0.1650	0.6062	1.5028	0.9672	27.64
385.0	0.2120	0.6703	1.5036	0.9996	60.40
430.0	0.2610	0.7177	1.4564	1.0159	77.03
504.0	0.3603	0.7861	1.3482	1.0366	91.63
560.0	0.4461	0.8281	1.2700	1.0646	99.13
606.0	0.5145	0.8562	1.2285	1.0958	105.45
688.0	0.6832	0.9053	1.1048	1.2480	97.07
833.0	1.0000	1.0000			
		At 90 °C			
233.0	0.0000	0.0000			
245.0	0.0100	0.0785	1.7856	0.9761	12.98
247.0	0.0120	0.0970	1.8534	0.9662	−19.11
263.0	0.0265	0.1733	1.5952	0.9549	−23.44
356.0	0.0781	0.3803	1.5994	1.0170	37.73
444.0	0.1375	0.5372	1.5926	1.0068	50.43
540.0	0.2021	0.6400	1.5616	1.0233	78.28
636.0	0.2920	0.7300	1.4442	1.0125	83.76
720.0	0.3821	0.7860	1.3389	1.0353	95.95
801.0	0.4951	0.8303	1.3089	1.0530	86.57

(*18*, *19*) correlations and compared with the literature values (*9*, *26*). It was observed that the Antoine equation represented the data well for both the components. The gas phase second virial coefficients for ethyl acetate and methyl cellosolve were estimated by O'Connell correlation (*23*) using the parameters reported by Prausnitz et al. (*27*). Yen and Woods (*33*) and Goldhammer (*6*) correlations estimated the liquid molar volumes of ethyl acetate and methyl cellosolve, respectively.

The activity coefficient-composition data were correlated by Vanlaar (*3*), Redlich-Kister (*28*), Wilson (*24*), and NRTL (*29*) equations and the constants were evaluated by the nonlinear least-squares method of Nagahama et al. (*20*) minimizing the ln γ_1/γ_2 function. The Wilson equation represented the data well. The Wilson parameters along with the root mean square deviations in activity coefficient and vapor composition are reported.

		RMSD			
Λ_{12}	Λ_{21}	γ_1	γ_2	y_1	
1.0314	0.4192	0.0524	0.0304	0.0121	at 70 °C
0.6889	0.6928	0.0662	0.0617	0.0215	at 1 atm

The kinematic viscosity data are reported in Table V. The viscosity data are correlated by McAllister (*17*) and excess function models (*11*). The constants were evaluated by the least-squares method and are reported in Table V. The kinematic viscosity of the pure components were fitted in the equation

$$\ln \nu = A' + B'/T \tag{5}$$

where A' and B' are constants and T is absolute temperature, K. These constants over the temperature range of 25–70 °C are

Compound	A'	$10^{-2} B'$
Ethyl acetate	−4.2368	10.400
Methyl cellosolve	−5.3870	17.647

Thermodynamic Consistency. The isothermal vapor-liquid equilibrium data at 70 °C were tested for thermodynamic consistency by Herington's criterion (*8*) as suggested by Null (*22*), through the equation

$$\int_0^1 \ln \gamma_1/\gamma_2 \, dx_1 = -\int \frac{V^E}{RT} dP \tag{6}$$

The right-hand integral of eq 6 was evaluated from excess molar volume at 40 °C and the graphical differential of the P–x curve

Table IV. Isobaric Vapor-Liquid Equilibrium Data for Ethyl Acetate (1)-Methyl Cellosolve (2) at 760 mmHg

t, °C	x_1	y_1	$y_{1(\text{calcd})}$[a]	Z_1	Z_2	γ_1	γ_2
124.5	0.0000	0.0000					
121.8	0.0160	0.1080	0.1050	1.0762	0.9974	2.1341	0.9798
119.0	0.0355	0.1901	0.2128	1.0713	0.9945	1.7967	0.9889
113.2	0.0775	0.3505	0.3829	1.0610	0.9887	1.7252	0.9970
110.0	0.1032	0.4241	0.4560	1.0554	0.9856	1.6877	1.0092
106.5	0.1360	0.5100	0.5282	1.0488	0.9822	1.6731	1.0010
102.5	0.1940	0.6140	0.6348	1.0423	0.9785	1.5588	0.9978
98.5	0.2530	0.6820	0.6979	1.0345	0.9747	1.4702	0.9880
95.0	0.3201	0.7380	0.7506	1.0294	0.9714	1.3788	1.0122
91.0	0.4105	0.7920	0.7944	1.0227	0.9677	1.2864	1.0707
88.0	0.4854	0.8310	0.8134	1.0177	0.9649	1.2415	1.1128
83.9	0.6292	0.8856	0.8550	1.0109	0.9611	1.1487	1.2205
81.0	0.7695	0.9250	0.9024	1.0062	0.9583	1.0694	1.4371
79.5	0.8315	0.9430	0.9142	1.0032	0.9569	1.0557	1.5836
78.0	0.9402	0.9801	0.9683	1.0013	0.9555	1.0160	1.6520
77.1	1.0000	1.0000					

[a] $y_{1(\text{calcd})}$ calculated by Wilson equation.

Table V. Kinematic Viscosity of Ethyl Acetate (1)-Methyl Cellosolve (2)

	Kinematic viscosity cm^2 s^{-1}			Kinematic viscosity cm^2 s^{-1}	
x_1	40 °C	50 °C	x_1	60 °C	70 °C
0.0000	1.2595	1.0716	0.0000	0.9152	0.7901
0.0565	1.1214	0.9635	0.0565	0.8367	0.7291
0.1105	1.0352	0.8952	0.1105	0.7807	0.6752
0.1505	0.9755	0.8402	0.2273	0.6785	0.5902
0.2273	0.8729	0.7625	0.3183	0.6051	0.5355
0.3169	0.7728	0.6755	0.4072	0.5450	0.4825
0.3704	0.7234	0.6372	0.4560	0.5114	0.4601
0.4072	0.6915	0.6107	0.5054	0.4855	0.4382
0.4502	0.6675	0.5855	0.5448	0.4674	0.4222
0.4982	0.6210	0.5562	0.6036	0.4421	0.4026
0.5586	0.5801	0.5223	0.6434	0.4269	0.3856
0.5914	0.5603	0.5051	0.6945	0.4124	0.3703
0.6603	0.5222	0.4745	0.7503	0.3932	0.3521
0.6929	0.5065	0.4588	0.8018	0.3772	0.3389
0.7558	0.4772	0.4336	0.8495	0.3631	0.3275
0.7961	0.4588	0.4195	0.8904	0.3525	0.3207
0.8474	0.4402	0.4022	0.9451	0.4301	0.3085
0.9112	0.4196	0.3831	1.0000	0.3290	0.3004
0.9436	0.4115	0.3750			
1.0000	0.3975	0.3622			

	McAllister			Excess function			
t, °C	ν_{12}	ν_{21}	RMSD	A''	B''	C''	RMSD
40	0.4926	0.6977	0.0085	0.5101	−0.0100	0.1166	0.0089
50	0.4512	0.6225	0.0048	0.4431	−0.0171	0.0797	0.0049
60	0.3944	0.5722	0.0041	0.3930	−0.0214	0.0940	0.0047
70	0.3473	0.5334	0.0055	0.3830	0.0125	0.1721	0.0061

and was found to be negligible. The consistency index was found to be 0.0466 (Figure 2). The isobaric data also satisfied Herington's consistency criterion (*8*) and Black (*2*) and Norrish-Twigg (*21*) tests. The latent heat of vaporization needed in applying the Norrish-Twigg test was calculated by Watson's correlation (*32*) using latent heats at normal boiling points from the literature (*14*).

Any G^E data available at more than one temperature were tested for mutual consistency with H^E data using the Gibbs-Helmholtz equation

$$H^E = \frac{\delta(G^E/T)}{\delta(1/T)} \tag{7}$$

The equation related the slope of the G^E/T curve with respect to the reciprocal temperature to excess enthalpy. The test is performed at $x_1 = 0.5$ using G^E values at 70, 80, and 90 °C from isothermal vapor-liquid equilibrium data. The curve is extrapolated to 48.2 °C where its slope is 135 cal mol^{-1}. The agreement is good indicating the mutual consistency of G^E and H^E data (Figure 3).

The present binary system showed the positive deviations from Raoult's law. The esters are proton acceptors, and methyl cellosolve has a proton donor group. Spectroscopic evidence indicated that intermolecular H bonding in methyl cellosolve persists in CCl_4 solution (*12*). This is a typical example of a mixture of associated liquid and slightly polar liquid (ethyl acetate), showing firstly the excess heat of mixing being positive with a maximum in a mixture weak in the polar component, secondly the excess free energy is large, positive, and nearly symmetrical in composition. The activity coefficient of ethyl acetate in solution decreases with increase in temperature as seen from the isothermal data. The negative deviation of viscosity from rectilinear dependence on mole fraction indicates the presence of some dominant dispersive forces in the mix-

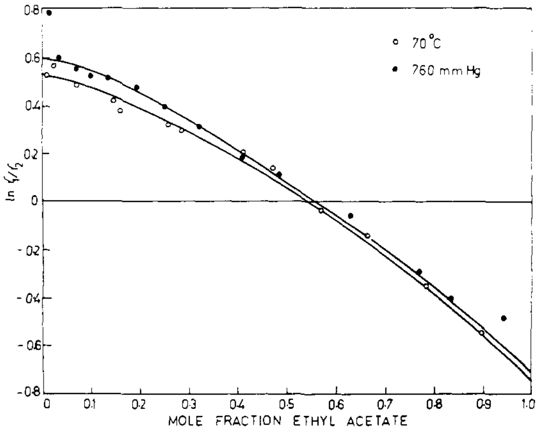

Figure 2. Thermodynamic consistency by Herington's criteria.

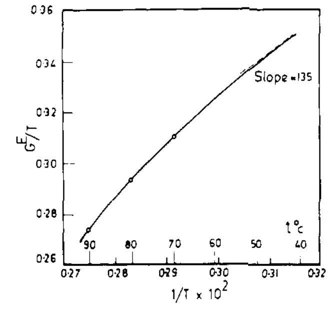

Figure 3. Mutual consistency of G^E and H^E data for ethyl acetate (1)-methyl cellosolve (2).

tures. The rise in temperature will disrupt the acetate–alcohol association as well as the self-association of methyl cellosolve. This is observed from the decrease in the activation energies ν_{12} and ν_{21} with increase in temperature from Table V.

Acknowledgment

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Glossary

A,B,C	constants in Antoine vapor pressure equation
A'',B'',C''	excess function model constants
A',B'	constants in eq 5
C_1,C_2,C_3	constants in representing the excess properties by eq 2
G^E	molar excess Gibbs free energy, cal mol^{-1}
H^E	molar excess enthalpy, cal mol^{-1}
n	number of coefficients in eq 2
P^0	vapor pressure of pure components, mmHg
P	system pressure, mmHg
R	gas constant
T	absolute temperature, K
t	system temperature, °C
V	liquid molar volume of pure component, mL mol^{-1}
V^E	molar excess volume, mL mol^{-1}
x_i	mole fraction of component *i* in liquid phase
y_i	mole fraction of component *i* in vapor phase
X^E	molar excess property
Z_1,Z_2	vapor phase nonideality correction
RMSD	root mean square deviation = $((\gamma_{i(\text{exptl})} - \gamma_{i(\text{calcd})})^2/N)^{1/2}$
N	number of data points

Greek letters

β	gas phase second virial coefficient of pure component, mL mol^{-1}
γ	activity coefficient of the component in liquid phase
π	total pressure, mmHg
ν	kinematic viscosity, cm^2 s^{-1}
$\Lambda_{12},\Lambda_{21}$	Wilson parameters
ν_{12},ν_{21}	parameters in McAllister model

Subscripts

1	more volatile component
2	less volatile component
calcd	calculated
i	component *i*

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