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Excess Gibbs free energy of butyl acetate with cyclohexane and aromatic hydrocarbons at 308.15 K

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Abstract−Molar excess Gibbs free energies of mixing (C^E) for butyl acetate+cyclohexane or benzene or toluene or *o*- or *m*- or *p*-xylene were calculated by using Barker's method from the measured vapor pressure data by static method at 308.15±0.01 K over the entire composition range. The G^E values for the binary mixtures containing cyclohexane or benzene are positive; while these are negative for toluene, *o*-, *m*- and *p*-xylene system over the whole composition range. The G^E values of an equimolar mixture for these systems vary in the order: cyclohexane>benzene>*o*-xylene>*m*-xylene>*p*-xylene>toluene. The G^E values for these systems were also calculated by Sanchez and Lacome theory using the previously published excess enthalpy and excess volume data. It is found that while values of G^E from Sanchez and Lacombe theory are in reasonably good agreement with those calculated by Barker method for m-xylene and p-xylene mixtures, agreement is very poor for other systems although they predict the sign of G^E data except in the case of mixtures containing benzene.

Key words: Gibbs Free Energy, Vapor Liquid Equilibrium, Static Method, Butyl Acetate, Aromatic Hydrocarbon, Barker's Method, Sanchez and Lacombe Theory

INTRODUCTION

The design of separation processes in industry requires a great knowledge of the behavior of different systems in the phase equilibria [1-6]. Accurate vapor-liquid equilibrium (VLE) data are essential for chemical engineers to design a distillation tower, which is the most important part of petroleum refining and also important to chemists to understand the nature of molecular interactions. Butyl acetate is widely used as a solvent in synthetic resins, adhesives, perfume and paint industry as well as a flavoring agent in the pharmaceutical and food industry. A number of experimental as well as theoretical studies on thermodynamic properties for alkyl ester with alkanes [7-11], amine [12], aromatic hydrocarbons [13-17], alcohols [18,19], chloroalkane [20], and acetonitrile [16] have been reported in the literature. Among them, systemic data for alkyl ester+aromatic polar solvent are relatively rare [13-17]. Alkyl esters are characterized by dipole-dipole interactions in the pure state [5,6]. The degree and strength of dipole-dipole interactions decreases with the increasing size of alkyl group in the esters. The additions of inert solvents like alkane generally break the orientation order of pure alkyl esters to give the positive value of excess thermodynamic functions like excess molar volume (V^E), excess molar enthalpy (H^E), and excess molar Gibb's free energy (G^E) [8,9]. However, these values become considerably less and even negative in the mixtures of alkyl esters+aromatic hydrocarbons [14,15]. Specific interactions of dipole-induced dipole type are postulated to account for such behavior [14,15].

These considerations prompted us to carry out a systemic study on the thermodynamic properties of alkyl esters with aromatic hydrocarbons. In our earlier paper [21], the measured V^E and H^E data for n-butyl acetate+cyclohexane or benzene or toluene or *o*- or *m*- or *p*-xylene were interpreted in terms of a graph theoretical approach [22]. In this paper, we are reporting the excess molar Gibbs free energies of mixing (G^E) for these systems at 308.15±0.01 K calculated from the measured vapor pressure data using Barker's method [23]. We are unaware of any vapor pressure data with which to compare our results.

EXPERIMENTAL SECTION

Butyl acetate (BA) (Merck) was dried over anhydrous magnesium sulphate and then fractionally distilled [24]. The middle fraction of distilled BA was then dried over type 0.3 nm molecular sieves (Merck) in an amber-colored bottle for several days before use. Cyclohexane, benzene, toluene and xylenes (Merck) were purified with standard procedure [25]. The purities of the purified samples were checked by measuring their densities, refractive indices and vapor pressure at 308.15 K, and these compared well with the literature values [24,26-32] as shown in Table 1.

The total vapor pressures of the pure components and their various binary mixtures were measured as a function of liquid phase mole fraction of butyl acetate (x_A) at 298.15 K by the static method [33] in the manner described by Nigam and Mahl [34]. The height of the mercury column in the manometer was read with a cathetometer (OSAW, India) with a resolution of ±1×10^{-5} m. The uncertainties in the vapor pressure measurements due to manometer were ±2.66 Pa. Condensation effect on mercury was avoided by vacuum suction of condensed vapors from a manometer after each measurement, and also mercury was changed after every four or five readings. Our experimental values for the vapor pressure of the pure

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Table 1. Measured values of densities (ρ), refractive indices (n_D) and vapor pressures (p) of the pure components at 308.15 K

Compound	ρ/kg m^{-3}		n_D		p/kPa	
	Exptl.	Lit. [Ref.]	Exptl.	Lit. [Ref.]	Exptl.	Lit. [Ref.]
Butyl acetate	865.46	865.5 [28]	1.3881	1.3882 [28]	2.706	2.704 [29]
Cyclohexane	764.49	764.46 [30]	1.4204	1.4201 [31]	20.085	20.082 [32]
Benzene	861.93	861.9 [33]	1.4908	1.4910 [33]	19.983	19.98 [34]
Toluene	850.88	850.9 [33]	1.4885	1.4881 [33]	6.404	6.40 [34]
o-Xylene	867.42	867.38 [30]	1.5032	1.50295[a] [26]	1.687	1.69 [34]
m-Xylene	851.58	851.57 [30]	1.4945	1.49464[a] [26]	1.882	1.88 [34]
p-Xylene	847.52	847.5 [33]	1.4873	1.4875 [33]	2.065	2.06 [34]

[a] At 298.15 K.

Table 2. Measured vapor pressure (p), activity coefficients (γ_1 and γ_2), residual vapor pressure ($\delta p = p_{calcd} - p_{exptl}$), where p_{calcd} is obtained from Barker's method, and molar Gibbs energies of mixing (G^E) as a function of mole fraction x_1 at 308.15 K for butyl acetate (1)+ an aromatic hydrocarbon (2)

x_1	p/kPa	γ_1	γ_2	δp/kPa	G^E/Jmol^{-1}	x_1	p/kPa	γ_1	γ_2	δp/kPa	G^E/Jmol^{-1}
Butyl Acetate (1)+Cyclohexane (2)						Butyl Acetate (1)+*o*-Xylene (2)					
0.1199	19.73	2.94	1.0545	0.01	449	0.0801	1.81	0.8908	0.9979	0	−27
0.2401	19.33	1.764	1.1756	0.03	667	0.1599	1.96	0.9233	0.9932	0	−44
0.3497	18.54	1.375	1.3018	0.04	724	0.2602	2.15	0.9493	0.9861	−0.01	−58
0.4503	17.35	1.224	1.4051	−0.02	711	0.3311	2.3	0.9605	0.9812	0	−64
0.5805	15.07	1.137	1.5162	−0.01	639	0.4098	2.45	0.9687	0.9764	0	−68
0.6196	13.42	1.12	1.5505	−0.82	606	0.5405	2.72	0.9768	0.9691	0	−70
0.5997	12.55	1.09	1.6355	−0.01	626	0.6798	3.01	0.9842	0.9575	0	−67
0.7604	11.07	1.068	1.7312	−0.17	462	0.7203	3.11	0.9866	0.9522	−0.01	−64
0.8205	9.73	1.045	1.8794	−0.15	385	0.8297	3.32	0.9934	0.9593	−0.02	−50
0.8698	8.17	1.018	2.3546	−0.65	300	0.9309	3.6	0.9985	0.8934	0.03	−26
0.9301	6.04	1.015	2.980	−0.15	179						
Butyl Acetate (1)+Benzene (2)						Butyl Acetate (1)+*m*-Xylene (2)					
0.1302	18.93	1.8155	1.0299	0	264	0.1197	1.97	0.7602	0.9854	0.01	−111
0.2608	17.47	1.4004	1.0947	−0.07	396	0.2295	2.12	0.8803	0.9583	−0.01	−157
0.3396	16.67	1.1868	1.1907	−0.12	432	0.3096	2.29	0.9327	0.939	0	−168
0.4297	15.33	1.1199	1.2582	0	443	0.3898	2.48	0.9567	0.9262	−0.01	−166
0.5495	13.6	1.1077	1.2763	0.12	424	0.4894	2.64	0.9615	0.9225	0	−153
0.5798	13.21	1.0823	1.3244	0.13	414	0.5602	2.77	0.9621	0.9219	0	−146
0.6501	12	1.0691	1.3579	−0.2	384	0.6604	2.98	0.9642	0.9185	0.02	−138
0.6904	10.95	1.0534	1.4093	−0.11	361	0.7509	3.16	0.9713	0.9018	0.02	−120
0.7399	10.11	1.0223	1.587	0.16	327	0.8596	3.39	0.9862	0.8443	0	−90
0.8501	8	1.0043	1.8536	−0.02	226	0.9492	3.49	0.9976	0.7529	0.01	−45
0.92	5.72	1.0001	1.9001	−0.02	134						
Butyl Acetate (1)+Toluene (2)						Butyl Acetate (1)+*p*-Xylene (2)					
0.1298	5.81	0.7462	0.9844	−0.03	−130	0.0808	2.13	0.7628	0.9947	0.01	−66
0.2395	5.41	0.8434	0.968	0	−186	0.1894	2.21	0.8519	0.9784	−0.03	−125
0.3392	5.11	0.9017	0.9326	0.02	−209	0.2696	2.39	0.8897	0.966	0.03	−147
0.4403	4.83	0.9377	0.9099	0.02	−207	0.3602	2.48	0.9135	0.9546	−0.01	−159
0.5006	4.64	0.9518	0.898	−0.01	−200	0.4408	2.59	0.9249	0.9468	−0.03	−166
0.5607	4.5	0.9621	0.8872	−0.01	−188	0.4897	2.69	0.9299	0.9424	0	−171
0.6994	4.2	0.9788	0.8617	−0.01	−155	0.5799	2.88	0.9387	0.9322	0.04	−168
0.7692	4.05	0.9855	0.8454	−0.02	−125	0.6907	3.02	0.9532	0.9069	−0.02	−163
0.8499	3.97	0.9925	0.8199	0.04	−95	0.7803	3.2	0.9691	0.8655	−0.13	−145
0.9104	3.84	0.9969	0.7937	0	−60	0.8911	3.48	0.9895	0.7762	0.02	−97

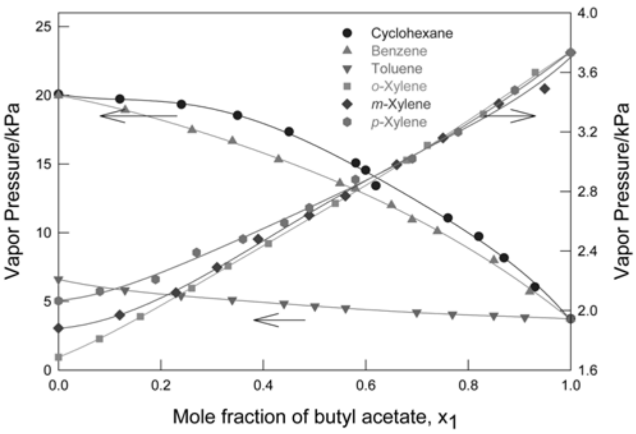

Fig. 1. Experimental vapor pressures as a function of mole fraction of butyl acetate (x_1); the smooth lines represent the calculated vapor pressures for the corresponding systems from Barker's method.

compounds compared well with the literature values as shown in Table 1. The composition of the liquid phase was determined by measuring the refractive index with an Abbe refractometer in the manner described by Strubl et al. [35]. The uncertainties in the liquid phase composition were about 0.01 mol per cent.

RESULTS AND DISCUSSION

The measured vapor pressure (p) for the present *n*-butyl acetate (1)+benzene, or toluene, or *o*-, or *m*-, or *p*-xylene (2) systems at 308.15 K over the entire composition range are reported in Table 2 and shown graphically in Fig. 1. The values of excess molar Gibbs free energies of mixing G^E as a function of composition are calculated by fitting VLE data using Barker's method [23] of minimizing the residual pressure (δp) to Redlich-Kister Eq. (1) and are reported in Table 2 along with activity coefficients and δp and are shown in Fig. 2.

$$G^E/RT = x_1x_2\sum_{j=0}^{2}A_j(x_2-x_1)^j \tag{1}$$

where A_j are the adjustable parameters and reported in Table 3 along with standard deviation in vapor pressure.

The activity coefficients γ_A and γ_B are obtained by differentiation of the above Eq. (2)

$$\ln\gamma_1 = x_2^2\left[A_0+\sum_{j=1}^{m}\{A_j(x_2-x_1)^j-2jA_jx_1(x_2-x_1)^{j-1}\}\right] \tag{2}$$

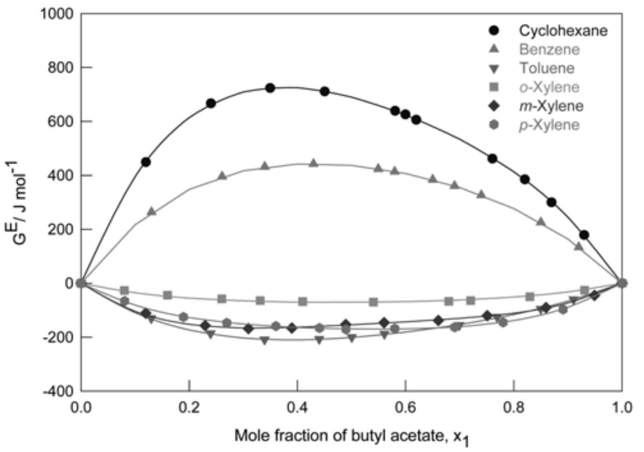

Fig. 2. Molar excess Gibbs free energies as a function of mole fraction of butyl acetate (x_1).

$$\ln\gamma_2 = x_1^2\left[A_0+\sum_{j=1}^{m}\{A_j(x_2-x_1)^j-2jA_jx_2(x_2-x_1)^{j-1}\}\right] \tag{3}$$

where subscripts '1' and '2' stand for butyl acetate and second component, respectively. The calculated vapor pressure is then given by:

$$p_{calcd} = x_1\gamma_1p_1^*R_1 + x_2\gamma_2p_2^*R_2 \tag{4}$$

using for non-ideality of the vapor phase the corrections:

$$R_1 = \exp[\{(V_1^*-\beta_{11})(p-p_1^*)-p\delta_{12}y_2^2\}/RT] \tag{5}$$

$$R_2 = \exp[\{(V_2^*-\beta_{22})(p-p_2^*)-p\delta_{12}y_1^2\}/RT] \tag{6}$$

where V_1^* and V_2^* are the molar volumes, y_1 and y_2 are the vapor phase mole fractions of butyl acetate and second component, respectively and δ_{12} is the cross second virial coefficient of the (1+2) binary mixture and defined as:

$$\delta_{12} = 2\beta_{12}-\beta_{11}-\beta_{22} \tag{7}$$

where β_{11}, β_{22}, β_{12} and are the second virial coefficients of butyl acetate, second component, and (1+2) mixture, respectively. In this case $\delta_{12}=0$, as we assume that vapor is an ideal mixture of imperfect gases:

$$\beta_{12} = (\beta_{11}+\beta_{22})/2 \tag{8}$$

The virial coefficients required for these calculations are calculated from Berthelot's equation [36] and critical constant data used therein are taken from the literature [37].

$$\beta = 9RT_C/128P_C - 27RT_C^3/64PT_C^2 \tag{9}$$

Table 3. Adjustable parameters A_j (j=0-2) of Eq. (1) and standard deviation s(p)

System	A_0 /J mol^{-1}	A_1/J mol^{-1}	A_2/J mol^{-1}	s(p)/kPa
Butyl Acetate (1)+Cyclohexane (2)	1.0776	0.4085	0.4852	0.361
Butyl Acetate (1)+Benzene (2)	0.6815	0.1451	0.2259	0.120
Butyl Acetate (1)+Toluene (2)	−0.314	−0.1162	−0.1043	0.022
Butyl Acetate (1)+*o*-Xylene (2)	−0.1093	−0.0082	−0.0545	0.014
Butyl Acetate (1)+*m*-Xylene (2)	−0.2398	−0.0836	−0.2245	0.012
Butyl Acetate (1)+*p*-Xylene (2)	−0.2639	0.0226	−0.1633	0.056

The virial coefficients of binary mixtures are taken as the arithmetic mean of second virial coefficients of pure components. The standard deviations are defined by:

$$s(p)=\left[\sum_{i=1}^{N}(\delta p)_i^2/(N-m)\right]^{1/2} \qquad (10)$$

where δp_is are the residual pressures according to Barker's method, N is the number of experimental points, and m is the number of parameters in the corresponding analytical equation.

The G^E values are found to be positive for a system containing cyclohexane or benzene and negative for other systems over the entire composition range, and for an equimolar composition G^E values follow the sequence: cyclohexane>benzene>*o*-xylene>*m*-xylene>*p*-xylene>toluene.

1. Sanchez and Lacombe Theory

According to Sanchez and Lacombe theory [38,39], the molar excess Gibbs free energy of mixing, G^E, can be represented as

$$G^E_{Sanchez}=H^E_{Sanchez}-RT\left[\sum_{i=1}^{2}x_i\ln\phi_i+r_{mix}(V^*_{mix}-1)\ln(1-\tilde{\rho}_{mix})+\ln\tilde{\rho}_{mix}\right]$$
$$-\sum_{i=1}^{2}x_i[r_i^o(V^*_{mix}-1)\ln(1-\rho_i)+\ln\tilde{\rho}_{mix}] \qquad (11)$$

where

$$H^E_{Sanchez}=2\phi_1\phi_2 r_{mix}\tilde{\rho}_{mix}\chi_{12}+RTr_{mix}\left[\sum_{i=1}^{2}((\tilde{\rho}_i\phi_i^o-\tilde{\rho}_{mix}\phi_i)/\tilde{T}_i)\right] \qquad (12)$$

$$\chi_{12}=\varepsilon^*_{11}+\varepsilon^*_{22}-2\varepsilon^*_{12} \qquad (13)$$

$$\phi_i=(m_i/\rho^*_i)/\sum_{i=1}^{2}(m_i/\rho^*_i) \qquad (14)$$

$$m_i=x_iM_i/\sum_{i=1}^{2}(x_iM_i) \qquad (15)$$

Table 4. Values of various parameters of pure component used in Sanchez and Lacombe theory along with the interaction energy ε^*_{12} for the binary system

Compound	ε^*_{ii} Kcal mol^{-1}	V^*_i cm^3 mol^{-3}	r_i	T^*_i K	P^*_i Atm	ρ^*_i gm cm^{-3}	E^*_{12} Kcal mol^{-1}
Butyl acetate	0.99	10.50	11.03	498	3890	1.003	
cyclohexane	0.987	10.79	8.65	497	3780	0.902	0.9106
Benzene	1.039	9.80	8.02	523	4380	0.994	0.9702
Toluene	1.080	11.22	8.50	543	3970	0.966	1.0023
o-Xylene	1.115	12.24	9.141	561	3760	0.949	1.0200
m-Xylene	1.114	12.11	9.21	560	3800	0.952	1.0141
p-Xylene	1.135	12.03	9.14	571	3890	0.965	1.0190

Table 5. Comparison of G^E values from Barker's method (at rounded mole fractions) with their corresponding values calculated from Sanchez and Lacombe (S & L) theory for various butyl acetate (BA) (1)+(2) mixtures

x_1	G^E/J mol^{-1}					
	Barker	S & L	Barker	S & L	Barker	S & L
	BA (1)+Cyclohexane (2)		BA (1)+Benzene (2)		BA (1)+Toluene (2)	
0.1	395	208	217	−191	−109	−414
0.2	614	534	348	−203	−173	−547
0.3	709	775	417	−220	−203	−615
0.4	725	902	442	−261	−210	−654
0.5	690	913	437	−321	−201	−677
0.6	624	815	407	−389	−181	−685
0.7	534	622	355	−445	−153	−668
0.8	413	359	277	−463	−116	−607
0.9	245	78	164	−388	−66	−457
	BA (1)+*o*-Xylene (2)		BA (1)+*m*-Xylene (2)		BA (1)+*p*-Xylene (2)	
0.1	−34.8	−316	−104	−351	−81	−341
0.2	−54.9	−356	−152	−417	−127	−398
0.3	−65.3	−340	−166	−419	−151	−391
0.4	−69.6	−314	−163	−402	−164	−367
0.5	−70.0	−297	−154	−386	−169	−347
0.6	−67.5	−295	−143	−378	−169	−337
0.7	−61.7	−306	−130	−375	−161	−337
0.8	−50.8	−315	−111	−366	−138	−335
0.9	−31.7	−281	−73	−309	−89	−291

$$\mathrm{r}_{mix}=\sum_{i=1}^{2}(\mathrm{x}_i\mathrm{r}_i) \tag{16}$$

$$\mathrm{r}_i=\mathrm{r}_i^o\mathrm{V}_i^*/\mathrm{V}_{mix}^* \tag{17}$$

$$\mathrm{V}_{mix}^*=\sum_{i=1}^{2}\phi_i^o\mathrm{V}_i^* \tag{18}$$

$$\phi_i^o=(\mathrm{m}_i/\rho_i^*\mathrm{V}_i^*)/\sum_{i=1}^{2}(\mathrm{m}_i/\rho_i^*\mathrm{V}_i^*) \tag{19}$$

$$\tilde{\mathrm{V}}_{mix}=1/\tilde{\rho}_{mix} \tag{20}$$

In these equations, x_i, M_i, ϕ_i, ϕ_i^o, V_i^*, ρ_i, $\tilde{\mathrm{T}}_i$, r_i, r_i^o and ε_{11}^* have the same significance as described by Sanchez and Lacombe [38,39]. Besides a knowledge of the reduced density of the mixture, ρ_{mix}, evaluation of G^E according to this theory requires a knowledge of r_i^o, ρ_i, ε_{11}^* and V_i^* parameters for these compounds. These were taken from the literature [36] and reported in Table 4. Further, though the reduced density of the mixtures can in principle be evaluated from the following equation of state of the mixture proposed by Lacombe and Sanchez [39]

$$\tilde{\rho}_{mix}^2+\tilde{\mathrm{P}}+\tilde{\mathrm{T}}[\ln(1-\tilde{\rho}_{mix})+\tilde{\rho}_{mix}(1-1/\mathrm{r}_{mix})]=0 \tag{21}$$

where for the binary mixture

$$\tilde{\mathrm{T}}=\mathrm{T}/\mathrm{T}^* \qquad \mathrm{T}^*=\varepsilon^*/\mathrm{R} \tag{22}$$

$$\tilde{\mathrm{P}}=\mathrm{P}/\mathrm{P}^* \qquad \mathrm{P}^*=\varepsilon^*/\mathrm{V}^* \tag{23}$$

$$\tilde{\mathrm{V}}_{mix}=\mathrm{V}/\mathrm{V}^*=1/\tilde{\rho}_{mix} \tag{24}$$

$$\varepsilon_{mix}^*=\sum_{i=1}^{2}(\phi_i\varepsilon_{ii}^*)-\mathrm{RT}\sum\sum\phi_1\phi_2\chi_{12} \tag{25}$$

yet very few mixtures actually satisfy [40-42] this equation of state. To obviate this problem, we evaluated the reduced density of the mixture from its experimental V^E data using the relation:

$$\mathrm{V}^E=\left[1/\tilde{\rho}_{mix}-\sum_{i=1}^{2}(\phi_i\mathrm{V}_i^*)\right]\mathrm{r}_{mix}\mathrm{V}_{mix}^* \tag{26}$$

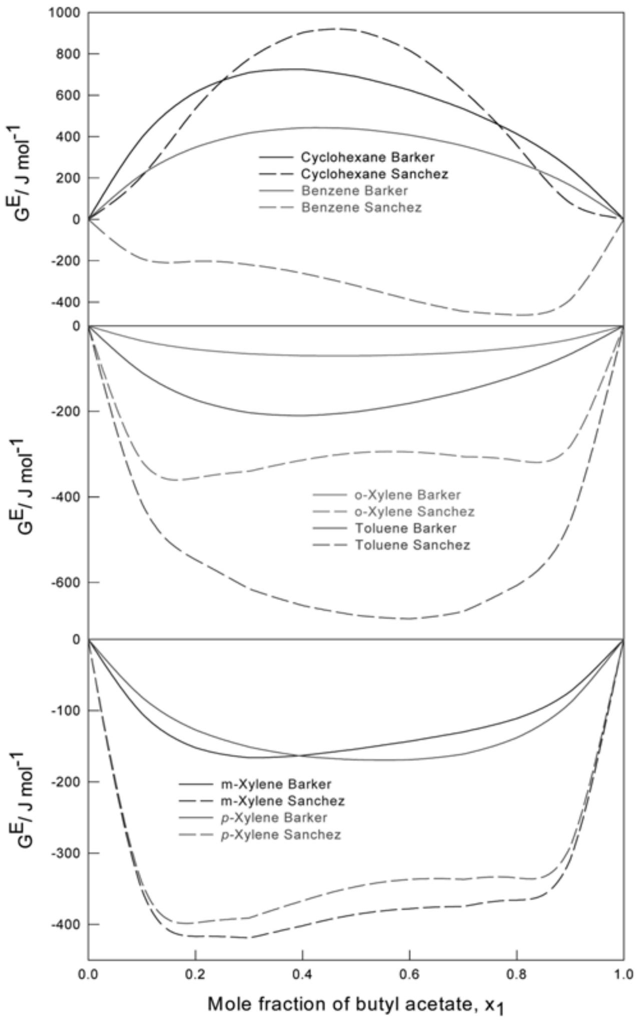

Fig. 3. Comparison of molar excess Gibbs free energies as a function of mole fraction of butyl acetate (x_1); solid line represent Barker's method and dashed line the Sanchez and Lacombe theory.

The interaction parameter χ_{12} for the mixture, required for these calculations, was evaluated by using experimental H^E and V^E data [24] for an equimolar mixture via Eqs. (12) and (26) and was subsequently used to evaluate the corresponding G^E data at rounded mole fraction for these binary systems. Such calculated values of G^E are shown in Table 5 and compared with the G^E values calculated from vapor pressure data by using the Barker method; the comparison is shown in Figs. 3-5. It is found that while values of G^E from the Sanchez and Lacombe theory are in reasonably good agreement with those calculated by the Barker method for *m*-xylene and *p*-xylene mixtures, agreement is very poor for other systems although they predict the sign of G^E data except in cases of mixtures containing benzene.

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