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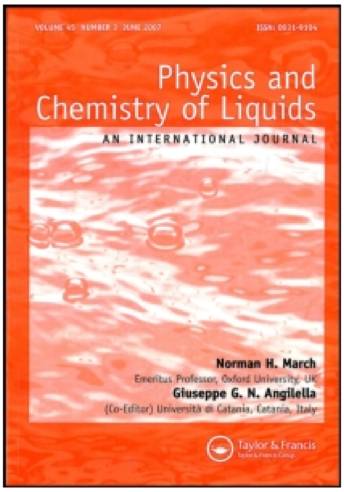

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VAPOR–LIQUID EQUILIBRIA OF 2-BUTANOL + AROMATIC HYDROCARBONS AT 308.15 K

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Vapor–liquid equilibria (VLE) data of 2-butanol + benzene or toluene or *o*- or *m*- or *p*-xylene measured by static method at 308.15 ± 0.01 K over the entire composition range are reported. The excess molar Gibbs free energies of mixing (G^E) for these binary systems have been calculated from total vapor pressure data using Barker's method. The G^E for these binary systems are also analyzed in terms of the Mecke–Kempter type of association model with a Flory contribution term using two interaction parameters and it has been found that this model describes well the G^E values of binary systems benzene or toluene.

Keywords: Vapor–liquid equilibria; Gibbs free energy; 2-Butanol; Aromatic hydrocarbon

INTRODUCTION

Oxygenated compounds like alkanol (C_1–C_4) and ether have been used as octane enhancing additives in gasoline to help in reducing automobile tail-pipe emission [1–5]. These are used as substitutes of poisonous leaded compounds and high octane aromatics in motor fuel. These additives introduce additional oxygen into gasoline which increases combustion efficiency, thereby reducing emission of toxic benzene, smog forming pollutants like volatile organic compounds and nitrogen oxides, and carbon monoxide from motor vehicles. Thus the thermo-physical properties of systems containing these oxygenated compounds would be of great importance in process engineering design and in formulating motor gasoline. Moreover, 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. Alkanol molecules are known to be associated through hydrogen bonding and their mixtures

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with inert solvent like alkane show a pronounced thermodynamic non-ideal behavior. Any excess thermodynamic property (X^E) for such binary system may be considered to consist of two parts, one resulting from breaking up of the hydrogen bonded network and the other due to van der Waals type interaction between the alkyl chain of alkanol and alkane. If these nonpolar solvents are replaced by a polar solvent like aromatic hydrocarbon, known to be electron donor, an additional term corresponding to interactions of π-electrons of the aromatic ring and hydroxyl group of alkanol may be required to describe the thermodynamic behavior of alkanol + an aromatic hydrocarbon. A lot of work has been reported in the literature [6–16] on the excess properties of alkanol + nonpolar solvent, while that of alkanol + polar solvent is relatively rare [16–24]. These considerations prompted us to measure the VLE data on 2-butanol + an aromatic hydrocarbon. In this article, the excess molar Gibbs free energies of mixing (G^E) of 2-butanol + benzene or toluene or *o*- or *m*- or *p*-xylene at 308.15 ± 0.01 K have been calculated from the measured VLE data using Barker's method [25], and their interpretation in terms of the Mecke–Kempter (MK) type association model [9] proposed by Treszczanowicz and Benson are presented. This is in continuation of our earlier work where the excess molar volume (V^E), enthalpy (H^E), and Gibbs energy (G^E) of propanol + an aromatic hydrocarbon at 298.15 K [26,27] and V^E and H^E of an isomer of butanol + an aromatic hydrocarbon at 308.15 K [28–31] have been described in terms of the MK type association model. To the best of our knowledge no VLE data have previously appeared in the literature for the present systems at 308.15 K.

EXPERIMENTAL SECTION

Benzene, toluene, and xylenes (Merck) were shaken repeatedly with 15 per cent (v/v) concentrated sulphuric acid in a separating funnel in order to eliminate thiophene from them until the acid layer was colorless [32]. After each shaking of a few minutes, the mixtures were allowed to settle and the acid was drawn off. The remaining acid was neutralized by sodium bicarbonate solution and the substance of interest was subsequently washed with distilled water and dried over fused calcium chloride for 24 h. It was then fractionally distilled and the middle fraction of distillate was stored over sodium wire in an amber colored bottle. 2-Butanol (Merck) was dried by refluxing over fused calcium oxide for 5 h and then fractionally distilled [32]. The middle fraction of distilled 2-butanol was then dried over type 3 Å molecular sieves (Merck) in amber colored bottle for several days before use. The purities of the purified samples were checked by measuring their densities, refractive indices, and vapor pressures at 308.15 K. The densities were measured with a precision of $\pm 0.05\,\mathrm{kg\,m^{-3}}$ by a specially designed densimeter, consisting of a bulb of approximate volume $3.5 \times 10^{-5}\,\mathrm{m^3}$ attached to a calibrated capillary through a B-10 standard joint in the manner described by Weissenberger [33]. Air buoyancy correction was also applied to achieve a greater accuracy. Refractive indices were measured with a thermostatically controlled Abbe refractometer (OSAW, India) using sodium D-line with an accuracy of $\pm 1 \times 10^{-4}$. The total vapor pressures of the pure components and their various binary mixtures were measured as a function of liquid phase mole fraction of 2-butanol (x_1) at 308.15 K by the static method [34] in the manner described by Nigam and Mahl [35]. The height of the mercury column in the manometer was read with a cathetometer

TABLE I Densities ρ, refractive indices n_D, and vapor pressures (p) and second virial coefficients (β) of the pure components at 308.15 K

Substance	ρ (kg m^{-3})		n_D		p (kPa)		β (cm^3 mol^{-1})
	This work	*Literature*	*This work*	*Literature*	*This work*	*Literature*	
2-Butanol	0.79394	0.79390[a]	1.3908	1.39100[a]	4.460	4.462[e]	−1281.22
Benzene	0.86296	0.86295[b]	1.4920	1.49170[c]	12.706	12.70[d]	−1272.19
Toluene	0.89290	0.85285[b]	1.4884	1.48870[c]	3.801	3.8036[d]	−1778.40
o-Xylene	0.86742	0.86738[b]	1.5030	1.50295[d]	0.882	0.8800[d]	−2378.60
m-Xylene	0.85155	0.85157[b]	1.4945	1.49464[d]	1.104	1.100[d]	−2347.80
p-Xylene	0.85790	0.84787[b]	1.4883	1.48810[c]	1.117	1.200[d]	−2358.43

[a] Ref. [36]. [b] Ref. [37]. [c] Ref. [38]. [d] Ref. [39]. [e] Ref. [40]. [d] $T = 298.15$ K.

(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 manometer after each measurement and also mercury was changed after every four or five readings. Our experimental values for the density, refractive index, and vapor pressure of the pure compounds compared well with the literature values as shown in Table I. The composition of the liquid phase was determined by measuring the refractive index using an Abbe refractometer in the manner described by Strubl *et al.* [41]. The uncertainties in the liquid phase composition were about 0.01 mol%.

RESULT AND DISCUSSION

The measured vapor pressure (p) for the present 2-butanol (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 II and shown graphically in Fig. 1. The values of Gibbs free energies of mixing G^E as a function of composition are calculated from VLE data using Barker's method [25] of minimizing the pressure residual and are reported in Table II and shown in Figs. 2 and 3. The G^E values are then fitted to the following Redlich and Kister equation

$$G^E = x_1x_2[A + B(x_1 - x_2) + C(x_1 - x_2)^2], \tag{1}$$

where A, B, and C are the adjustable parameters evaluated by least-squares procedure, recorded in Table III. The virial coefficients required for these calculations are calculated from Berthelot's equation [42]

$$\beta = 9RT_c/128P_c - 27RT_c^3/64P_cT^2$$

and critical constant data used therein are taken from the literature [43]. The virial coefficients of binary mixtures are taken as the arithmetic mean of second virial coefficients of pure components, reported in Table I.

Our calculated G^E values for all the binary systems are positive over the whole range of mole fraction of 2-butanol (x_1) (Figs. 2 and 3). The G^E *vs.* x_1 plots are

TABLE II Measured vapor pressure (p), activity coefficients (γ_1 and γ_2), residual vapor pressure ($\delta p = p_{\text{exptl}} - p_{\text{calcd}}$), 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 2-butanol (1) + an aromatic hydrocarbon (2)

x_1	p (kPa)	γ_1	γ_2	δp (kPa)	G^E (J mol^{-1})
2-Butanol + benzene					
0.0476	19.98	5.1669	1.0060	−0.27	215
0.1215	20.06	3.7191	1.0367	−0.18	490
0.1926	20.00	2.8605	1.0884	0.00	694
0.2662	19.88	2.2851	1.1634	0.06	848
0.3501	19.65	1.8569	1.2756	0.15	961
0.4664	19.09	1.4909	1.4832	0.14	1016
0.5590	18.24	1.3080	1.7017	−0.05	985
0.6475	17.15	1.1874	1.9709	−0.19	898
0.7148	16.15	1.1205	2.2308	−0.16	795
0.7615	15.37	1.0840	2.4492	0.00	705
0.7970	14.54	1.0610	2.6419	0.03	626
0.8630	12.43	1.0282	3.0804	0.02	456
0.9125	10.02	1.0117	3.5006	−0.24	308
0.9661	6.98	1.0018	4.0794	−0.13	127
2-Butanol + toluene					
0.0595	6.97	4.8966	1.0022	−0.38	247
0.1307	7.62	4.2652	1.0174	−0.56	524
0.2190	8.42	3.3744	1.0700	−0.27	818
0.2875	8.82	2.7486	1.1474	0.07	996
0.3595	9.29	2.2099	1.2739	0.47	1128
0.4399	9.51	1.7518	1.4836	0.69	1202
0.5250	9.47	1.4276	1.8011	0.60	1195
0.5890	9.22	1.2579	2.1111	0.31	1133
0.6540	8.78	1.1392	2.4831	−0.09	1025
0.7095	8.31	1.0718	2.8286	−0.38	900
0.7775	7.62	1.0223	3.2388	−0.58	714
0.8335	6.91	1.0027	3.5048	−0.59	541
0.8870	6.21	0.9966	3.6335	−0.40	366
0.9375	5.47	0.9975	3.5904	−0.17	199
2-Butanol + o-xylene					
0.0580	2.36	4.8521	1.0045	−0.50	246
0.1335	3.29	4.0727	1.0232	−0.65	531
0.1995	4.02	3.5428	1.0521	−0.58	751
0.2520	4.53	3.1889	1.0850	−0.45	905
0.3070	4.98	2.8649	1.1311	−0.30	1047
0.3723	5.48	2.5276	1.2065	−0.04	1186
0.4345	5.80	2.2446	1.3076	0.15	1289
0.5010	6.02	1.9773	1.4622	0.32	1361
0.5897	6.10	1.6720	1.7899	0.42	1389
0.6612	6.01	1.4669	2.2090	0.36	1345
0.7445	5.77	1.2734	3.1201	0.15	1206
0.8225	5.45	1.1371	4.7121	−0.18	976
0.8950	5.09	1.0503	7.6596	−0.50	660
0.9475	4.77	1.0132	11.7048	−0.59	363
2-Butanol + m-xylene					
0.0450	2.34	4.1172	1.0023	−0.29	169
0.1015	2.96	3.6708	1.0114	−0.43	364
0.1625	3.58	3.2819	1.0288	−0.44	556
0.2330	4.24	2.9111	1.5970	−0.35	752
0.3085	4.81	2.5740	1.1093	−0.21	931
0.3512	5.09	2.4023	1.1477	−0.11	1018
0.3955	5.37	2.2354	1.1981	0.02	1095

(*Continued*)

TABLE II Continued

x_1	p (kPa)	γ_1	Γ_2	δp (kPa)	G^E (J mol^{-1})
0.4825	5.74	1.9362	1.3415	0.22	1206
0.5520	5.84	1.7225	1.5213	0.28	1251
0.6320	5.83	1.5054	1.8514	0.26	1243
0.7130	5.67	1.3202	2.4282	0.12	1160
0.7770	5.49	1.2005	3.2085	−0.06	1030
0.8440	5.21	1.1027	4.6241	−0.34	823
0.9135	4.96	1.0334	7.4252	−0.50	521
2-Butanol + p-xylene					
0.0480	2.67	2.5576	1.0007	0.15	118
0.1020	3.36	2.5196	1.0018	0.38	246
0.1645	3.96	2.5129	1.0022	0.37	393
0.2170	4.44	2.5164	1.0018	0.36	517
0.2835	4.98	2.5077	1.0031	0.29	673
0.3514	5.48	2.4616	1.0120	0.22	831
0.4125	5.85	2.3764	1.0347	0.17	966
0.4930	6.27	2.1982	1.1048	0.23	1124
0.5801	6.58	1.9406	1.2790	0.39	1250
0.6710	6.70	1.6437	1.6942	0.58	1299
0.7428	6.63	1.4221	2.4093	0.59	1250
0.8050	6.27	1.2568	3.6876	0.23	1123
0.8795	5.76	1.1054	7.3721	−0.46	843

symmetrical for the system containing benzene or toluene, but these skewed toward the high mole fraction of 2-butanol, having a maximum G^E at $x_1 = 0.6$–0.7. For an equimolar mixture, G^E values vary in the order: *o*-xylene > *m*-xylene > toluene > *p*-xylene > benzene.

Mecke–Kempter Association Model

This model [9] was proposed by Treszczanowicz and Benson to describe excess thermodynamic properties of binary alkanol + alkane mixtures. It assumes that molar excess Gibbs energy of mixing ($G^E_{\text{MK model}}$) is composed of three type of contributions—the combinatorial contribution as described by Flory–Huggin theory [44], the chemical (MK type association of alkanol) contribution [9], and a physical contribution described by Flory theory [45,46]:

$$G^E_{\text{MK model}} = G^E_{\text{comb}} + G^E_{\text{MK}} + G^E_{\text{F}} \tag{2}$$

The combinatorial contribution is given by relation

$$G^E_{\text{comb}} = RT[x_1 \ln(\phi_1/x_1) + x_2 \ln(\phi_2/x_2)], \tag{3}$$

the chemical (MK) contribution is given by

$$G^E_{\text{MK}} = \left(x_1 RT/K^\phi \phi_1\right)\left[\phi_1(1+K^\phi)\ln(1+K^\phi) - \left(1+K^\phi\phi_1\right)\ln\left(1+K^\phi\phi_1\right)\right], \tag{4}$$

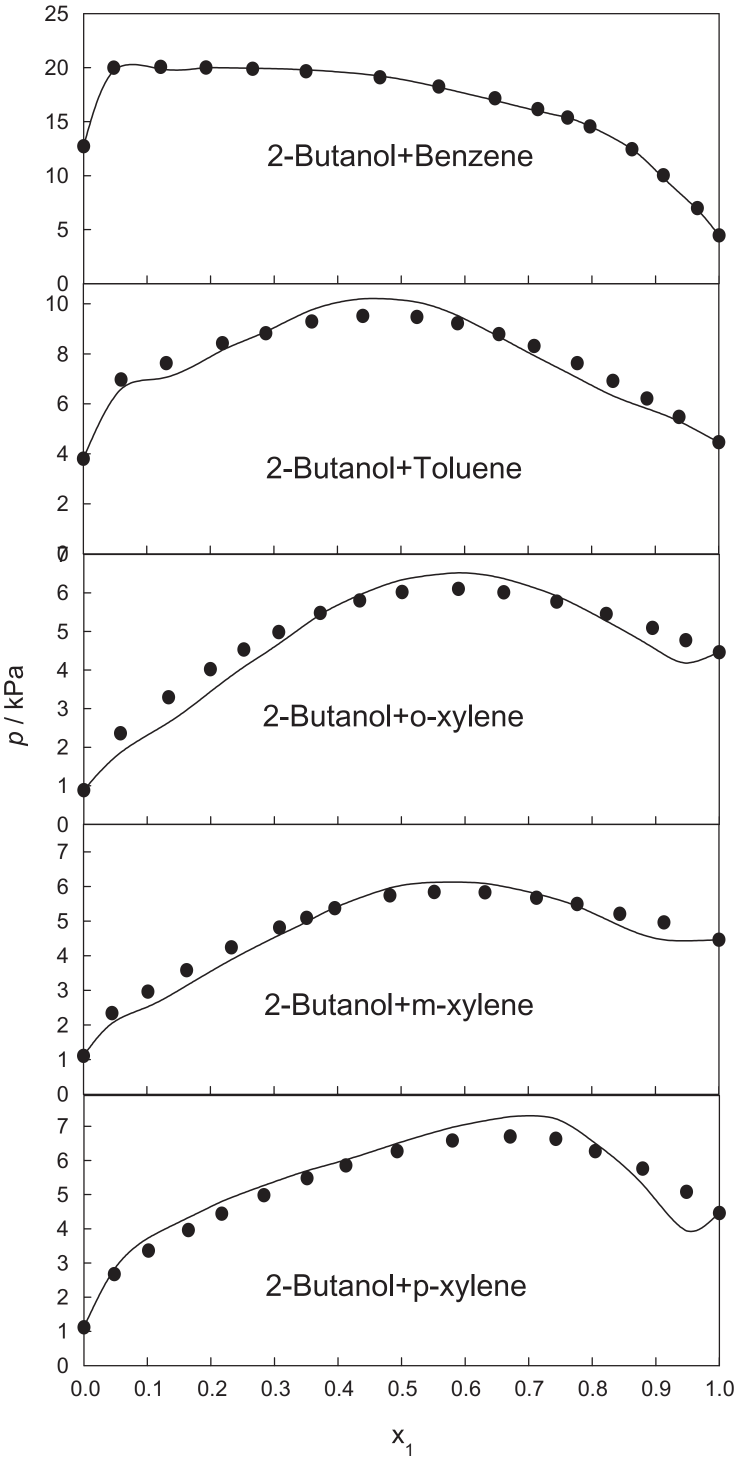

FIGURE 1 Experimental (•) and calculated (-) vapor pressures.

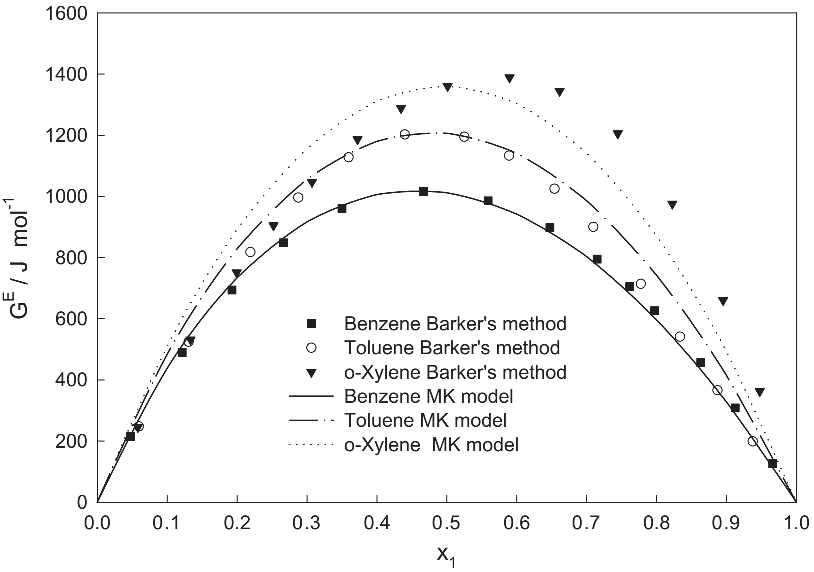

FIGURE 2 Molar excess Gibbs energy of 2-butanol (1) + aromatic hydrocarbon (2).

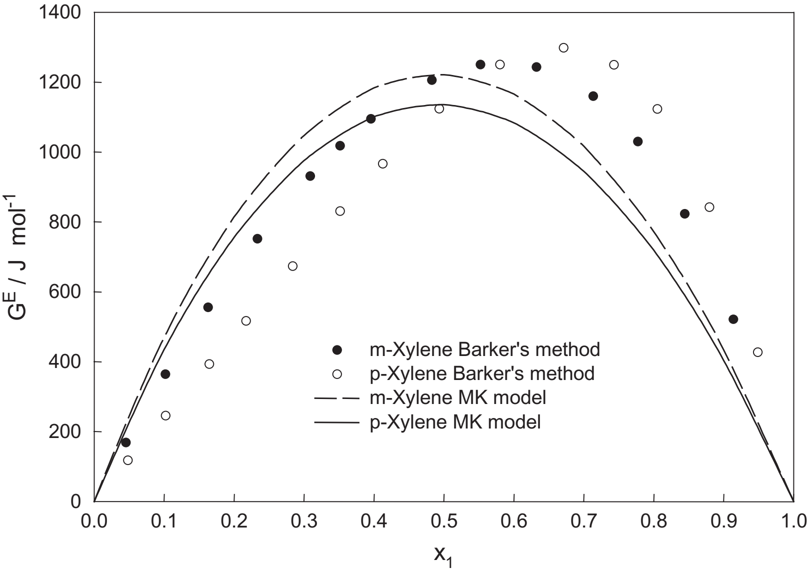

FIGURE 3 Molar excess Gibbs energy of 2-butanol (1) + aromatic hydrocarbon (2).

where

$$K^{\phi} = \exp\left[1 - \left(\Delta h_{\mathrm{H}}^{0} - T\Delta s_{\mathrm{H}}^{0}\right)/RT - \ln\left(V^{*}/17.12\,\mathrm{cm}^{3}\,\mathrm{mol}^{-1}\right)\right] \tag{5}$$

and the physical contribution is given by Flory

$$G_{\mathrm{F}}^{\mathrm{E}} = H_{\mathrm{F}}^{\mathrm{E}} - TS_{\mathrm{F}}^{\mathrm{E}} \tag{6}$$

TABLE III Adjustable parameters A, B, and C of Eq. (1)

System	A (J mol^{-1})	B (J mol^{-1})	C (J mol^{-1})
2-Butanol + benzene	4049	−483	307
2-Butanol + toluene	4824	−595	−1193
2-Butanol + *o*-xylene	5440	−1566	560
2-Butanol + *m*-xylene	4886	1585	586
2-Butanol + *p*-xylene	4546	3438	1378

where

$$H_{\mathrm{F}}^{\mathrm{E}} = x_1\theta_2\left(V_1^*/\tilde{V}\right)\chi_{12} + \sum\left[x_iP_i^*V_i^*\left(\tilde{V}_i^{-1} - \tilde{V}^{-1}\right)\right] \tag{7}$$

and

$$S_{\mathrm{F}}^{\mathrm{E}} = x_1\theta_2V_1^*Q_{12} - 3\sum\left(x_iP_i^*V_i^*/T_i^*\right)\ln\left[\left(\tilde{V}_i^{-1/3} - 1\right)/\left(\tilde{V}^{-1/3} - 1\right)\right]. \tag{8}$$

Calculation of $G_{\mathrm{MK\ model}}^{\mathrm{E}}$ requires the knowledge of association constants like Δv_{H}^0, Δh_{H}^0, and Δs_{H}^0 for these binary systems. These parameters were calculated from $V_{\mathrm{exptl}}^{\mathrm{E}}$ and $H_{\mathrm{exptl}}^{\mathrm{E}}$ values of an equimolar binary mixture in our earlier work [28] as suggested by Treszczanowicz and Benson [9]. Using these values of Δh_{H}^0 and Δs_{H}^0, K^{ϕ} was calculated from Eq. (5). Using this value of K^{ϕ}, $G_{\mathrm{F}}^{\mathrm{E}}$ is computed from Eq. (6). Calculation of the physical contribution $G_{\mathrm{F}}^{\mathrm{E}}$ from Eq. (6) requires two unknown interaction parameters χ_{12} and Q_{12}. In order to calculate χ_{12}, $V_{\mathrm{F}}^{\mathrm{E}}$ at equimole fractions is calculated from the relation

$$V_{\mathrm{F}}^{\mathrm{E}} = V_{\mathrm{exptl}}^{\mathrm{E}} - V_{\mathrm{MK}}^{\mathrm{E}}, \tag{9}$$

where

$$V_{\mathrm{MK}}^{\mathrm{E}} = \Delta v_{\mathrm{H}}^0 x_1 h\left(K^{\phi}, \phi_1\right) \tag{10}$$

and

$$h\left(K^{\phi}, \phi_1\right) = \left[\phi_1\ln\left(1 + K^{\phi}\right) - \ln\left(1 + K^{\phi}\phi_1\right)\right]/K^{\phi}\phi_1. \tag{11}$$

This value of $V_{\mathrm{m,F}}^{\mathrm{E}}$ is then used to calculate the Flory interaction parameter χ_{12} using the following set of equations [45,46]

$$V_{\mathrm{m,F}}^{\mathrm{E}} = V^*\left[\tilde{V} - \sum\left(\tilde{V}_i/\phi_i\right)\right] \tag{12}$$

$$\tilde{T} = \left[\left(\tilde{V}^{1/3} - 1\right)/\tilde{V}^{4/3}\right] \tag{13}$$

$$\chi_{12} = \left[\sum\left(\phi_iP_i^*\right) - \sum\left(\phi_iP_i^*\tilde{T}_i/\tilde{T}\right)\right]/\phi_1\theta_2, \tag{14}$$

TABLE IV The values of association parameters $\Delta v_{\mathrm{H}}^{0}$, $\Delta h_{\mathrm{H}}^{0}$, $\Delta s_{\mathrm{H}}^{0}$, and K^{ϕ} of the Mecke–Kempter model and the Flory interaction parameters χ_{12} and Q_{12} for various 2-butanol (1) + an aromatic hydrocarbons (2) systems at 308.15 K

System	K^{ϕ}	Q_{12} (J cm^{-3})	χ_{12} (J cm^{-3})	$\Delta h_{\mathrm{H}}^{0}$ (J mol^{-1})	$\Delta s_{\mathrm{H}}^{0}$ (J K^{-1} mol^{-1})	$\Delta v_{\mathrm{H}}^{0}$ (cm^3 mol^{-1})
2-Butanol + benzene	6.0405	−0.0752	4.2995	−17 774.56	−32.06	−4
2-Butanol + toluene	6.1477	−0.1017	0.7065	−16 890.48	−29.02	−4
2-Butanol + *o*-xylene	5.4436	−0.1227	0.1103	−18 107.70	−34.18	−4
2-Butanol + *m*-xylene	6.1990	−0.0905	1.9496	−16 381.49	−27.28	−4
2-Butanol + *p*-xylene	5.6990	−0.0832	0.5851	−17 414.95	−31.47	−4

where all the terms have their usual significance [9,44–46]. The other interaction parameter Q_{12} is calculated from Eq. (6) by assuming that at an equimole fraction

$$G_{\mathrm{F}}^{\mathrm{E}} = G^{\mathrm{E}} - G_{\mathrm{comb}}^{\mathrm{E}} - G_{\mathrm{MK}}^{\mathrm{E}} \tag{15}$$

and by using Eqs. (6)–(8). Method of calculation of all of these parameters was discussed in detail in our earlier work [26]. The values of χ_{12} and Q_{12}, along with association parameters $\Delta v_{\mathrm{H}}^{0}$, $\Delta h_{\mathrm{H}}^{0}$, and $\Delta s_{\mathrm{H}}^{0}$ for all the present systems are recorded in Table IV.

$G_{\mathrm{MK\ model}}^{\mathrm{E}}$ at other mole fractions for all these binary systems are calculated from Eq. (2) and are plotted against mole fraction of 2-butanol (x_1) in Figs. 2 and 3. It has been observed from plots of $G_{\mathrm{MK\ model}}^{\mathrm{E}}$ and G^{E} against x_1 that the $G_{\mathrm{MK\ model}}^{\mathrm{E}}$ values are in good agreement with G^{E} for 2-butanol + benzene or toluene, whereas the agreement is poor in case of 2-butanol + *o*-xylene or *m*-xylene or *p*-xylene.

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