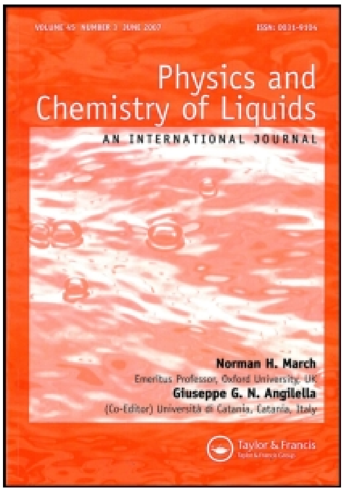

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Measurement and correlation of liquid-liquid equilibria of methanol 2-butanone *n*-alkanes (C_{10}-C_{12}) ternary mixtures

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Measurement and correlation of liquid–liquid equilibria of methanol + 2-butanone + *n*-alkanes (C_{10}–C_{12}) ternary mixtures

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The experimental liquid–liquid equilibrium (LLE) data of the mixtures methanol + 2-butanone + *n*-alkane (C_{10}–C_{12}) were studied at atmospheric pressure in the temperature range 268.15–298.15 K. The equilibrium compositions were measured by gas chromatography from samples splitted isothermally into a glass-stirred device and phase diagrams are reported at each temperature. The non-random two-liquid and universal quasichemical models were used to correlate the experimental tie-lines. A good fit has been obtained as a function of temperature for all the mixtures. The experimental results have been compared with the values predicted by the UNIFAC group contribution method and its modifications. Such predictions provide, however, a rather unsatisfactory description for the experimental results.

Keywords: Liquid–liquid equilibria; 2-Butanone; Methanol; *n*-Alkanes;

1. Introduction

Liquid–liquid equilibria (LLE) are of interest in extraction operations and recovery of valuable products. Nowadays, liquid–liquid extraction has become one of the main separation technologies for separation of complex liquid mixtures. Owing to that, there has been a significant growth in the number of publications devoted to the study on LLE of ternary and quaternary mixtures, including experimental data and correlation parameters. In this way, phase equilibria studies supply essential information to select adequate solvents and the design of extractors. Although simulation programmes provide a variety of possibilities for process synthesis, design and optimization, the quality of the results depends on the quality of the models used. In some cases, the results obtained predicting with group contribution methods might be sufficient, in other cases exact experimental data and a good description of them with the

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thermodynamic models is necessary. Hence, the experimental data and a study to determine the predictive capability of group contribution methods are required.

In this article, LLE of the methanol + 2-butanone + *n*-alkane (C_{10}–C_{12}) systems have been measured. To the authors' knowledge, experimental data for LLE in these mixtures are not available in the literature. The experimental data obtained were correlated with the UNIQUAC [1] and NRTL [2] equations and compared to those obtained by means of the UNIFAC group contribution method [3,4] and its modifications [5,6].

2. Experimental section

The chemicals used in this work were of chromatographic quality. Methanol and 2-butanone were supplied by Merck and *n*-alkanes were supplied by Sigma. The purities are better than 0.998 for methanol and 2-butanone and better than 0.994 mass fraction for the *n*-alkanes. The maximum water contents (determined with a Coulometric Moisture Model CA-06) were 1.5×10^{-2}, 3.7×10^{-2} and $<10^{-3}$ for methanol, 2-butanone and *n*-alkanes, respectively. The liquids were degassed in an ultrasonic bath for 4 h, stored in inert argon (N55, less than 3×10^{-6} by volume in water) and dried over molecular sieves type 0.3 nm for methanol and 0.4 nm for 2-butanone and *n*-alkanes, 1.6 mm (Aldrich cat. n° 20,860-4 or 20,858-2, respectively). The chemicals were checked using gas chromatography. The device for LLE determination consisted of a jacketed glass cell with a magnetic stirrer. The cell was connected to a PolyScience bath model 9510, whose temperature stability was $\pm10^{-2}$ K. The cell temperature was measured with a Yokogawa 7563 digital thermometer with a precision of $\pm10^{-2}$ K, calibrated with an Anton Paar MKT-100 digital thermometer (precision $\pm10^{-3}$ K, temperature scale ITS-90) over the entire temperature range. The samples were prepared by mass using a Mettler AX 205 DeltaRange balance with a precision of ±0.00008 g, which allows an accuracy of $\pm4\times10^{-4}$ mole fraction. Each mixture was stirred for 60 min and then left to settle for at least 12 h. After equilibrium was attained, the phases were allowed to separate and small samples of approximately 2 cm^3 were taken from the upper and lower phases with a syringe. The compositions were determined by gas chromatography using a Hewlett-Packard HP-6890 Series GC System chromatograph equipped with a flame ionization detector and a HP-1 19091Z-215 (methyl siloxane, $50\times3.2\times10^{-4}\times1.05\times10^{-6}$ m film thickness) capillary column. The carrier gas used was helium and the flow rate was 0.8 $cm^3\,min^{-1}$. Both injector and detector temperatures were maintained at $T=493.15$ K. The initial column temperature was $T=363.15$ K and the final temperature was $T=438.15$ K. An internal standard calibration method was used. The reference samples were analysed using the same chromatographic conditions as the equilibrium samples. The estimated accuracy in mole fraction for the equilibrium data was 3×10^{-3} in the *n*-alkane rich region (phase I) and 2×10^{-3} in the methanol rich region (phase II).

3. Results and discussion

In table 1, the experimental equilibria data of the mixtures methanol + 2-butanone + *n*-alkane (C_{10}–C_{12}) are gathered. The LLE compositions for the binary

Table 1. Experimental LLE data of the studied ternary mixtures.

x_1^{I}	x_2^{I}	x_1^{II}	x_2^{II}	x_1^{I}	x_2^{I}	x_1^{II}	x_2^{II}
Methanol (1) + 2-butanone (2) + *n*-decane (3)							
268.15 K							
0.046	0.000	0.985	0.000	0.118	0.133	0.729	0.220
0.052	0.014	0.955	0.028	0.133	0.154	0.693	0.247
0.058	0.027	0.928	0.052	0.149	0.177	0.654	0.273
0.066	0.043	0.896	0.081	0.170	0.202	0.617	0.297
0.075	0.060	0.864	0.109	0.202	0.234	0.567	0.323
0.081	0.072	0.838	0.130	0.229	0.259	0.532	0.337
0.094	0.095	0.798	0.165	0.291	0.300	0.465	0.349
0.101	0.107	0.773	0.185				
278.15 K							
0.060	0.000	0.984	0.000	0.125	0.098	0.804	0.151
0.268	0.228	0.597	0.278	0.134	0.109	0.783	0.167
0.066	0.013	0.957	0.022	0.151	0.129	0.754	0.190
0.075	0.027	0.931	0.045	0.169	0.149	0.722	0.211
0.083	0.038	0.911	0.063	0.199	0.178	0.678	0.240
0.086	0.051	0.887	0.085	0.218	0.194	0.648	0.256
0.095	0.065	0.862	0.105	0.318	0.253	0.547	0.292
0.113	0.082	0.833	0.128				
288.15 K							
0.075	0.000	0.980	0.000	0.145	0.086	0.830	0.122
0.311	0.201	0.638	0.233	0.165	0.106	0.798	0.145
0.087	0.016	0.951	0.024	0.189	0.128	0.762	0.171
0.095	0.027	0.932	0.041	0.213	0.146	0.733	0.189
0.103	0.040	0.908	0.062	0.246	0.168	0.695	0.210
0.110	0.054	0.884	0.080	0.364	0.221	0.590	0.245
0.136	0.076	0.846	0.110				
298.15 K							
0.091	0.000	0.974	0.000	0.194	0.096	0.815	0.122
0.109	0.015	0.951	0.021	0.224	0.115	0.785	0.143
0.124	0.031	0.925	0.042	0.264	0.138	0.742	0.166
0.137	0.042	0.906	0.057	0.304	0.161	0.700	0.185
0.156	0.063	0.871	0.083	0.356	0.181	0.643	0.202
0.170	0.077	0.847	0.101				
Methanol (1) + 2-butanone (2) + *n*-undecane (3)							
268.15 K							
0.042	0.000	0.990	0.000	0.101	0.139	0.725	0.238
0.046	0.017	0.955	0.033	0.117	0.165	0.683	0.271
0.053	0.034	0.921	0.065	0.135	0.196	0.635	0.309
0.063	0.054	0.883	0.100	0.153	0.226	0.591	0.339
0.071	0.074	0.845	0.134	0.170	0.258	0.547	0.366
0.084	0.099	0.799	0.174	0.213	0.304	0.487	0.393
0.094	0.122	0.758	0.210	0.249	0.331	0.447	0.402
0.094	0.122	0.758	0.210	0.290	0.361	0.399	0.399
278.15 K							
0.054	0.000	0.989	0.000	0.118	0.123	0.771	0.191
0.059	0.015	0.960	0.026	0.138	0.149	0.735	0.221
0.067	0.034	0.925	0.058	0.153	0.169	0.702	0.245
0.077	0.049	0.899	0.081	0.176	0.206	0.649	0.283
0.085	0.067	0.867	0.111	0.216	0.244	0.590	0.318
0.092	0.082	0.841	0.131	0.248	0.265	0.560	0.329
0.108	0.105	0.804	0.165	0.289	0.296	0.513	0.346
288.15 K							
0.063	0.000	0.987	0.000	0.170	0.141	0.762	0.190
0.075	0.017	0.956	0.027	0.191	0.165	0.725	0.216

(*Continued*)

Table 1. Continued.

x_1^{I}	x_2^{I}	x_1^{II}	x_2^{II}	x_1^{I}	x_2^{I}	x_1^{II}	x_2^{II}
0.089	0.037	0.924	0.056	0.209	0.190	0.689	0.240
0.098	0.052	0.899	0.077	0.247	0.217	0.646	0.263
0.107	0.069	0.873	0.100	0.310	0.246	0.589	0.286
0.124	0.093	0.835	0.132	0.395	0.277	0.516	0.295
0.143	0.111	0.805	0.156				
298.15 K							
0.084	0.000	0.984	0.000	0.174	0.114	0.806	0.150
0.092	0.015	0.961	0.021	0.197	0.134	0.773	0.172
0.099	0.030	0.937	0.042	0.214	0.151	0.747	0.190
0.113	0.046	0.911	0.064	0.240	0.176	0.714	0.213
0.126	0.059	0.891	0.081	0.268	0.198	0.677	0.233
0.140	0.077	0.858	0.107	0.292	0.212	0.654	0.245
0.149	0.093	0.838	0.124	0.342	0.231	0.613	0.258
Methanol (1) + 2-butanone (2) + *n*-dodecane (3)							
268.15 K							
0.039	0.000	0.994	0.000	0.090	0.147	0.714	0.260
0.041	0.014	0.962	0.028	0.105	0.167	0.689	0.280
0.047	0.030	0.933	0.058	0.110	0.186	0.659	0.306
0.051	0.044	0.900	0.085	0.118	0.208	0.623	0.335
0.058	0.062	0.874	0.114	0.124	0.223	0.595	0.359
0.065	0.077	0.844	0.141	0.140	0.253	0.552	0.386
0.073	0.091	0.816	0.166	0.151	0.275	0.522	0.409
0.078	0.109	0.785	0.194	0.177	0.313	0.478	0.434
0.084	0.128	0.754	0.223	0.182	0.331	0.453	0.448
278.15 K							
0.053	0.000	0.992	0.000	0.123	0.145	0.749	0.221
0.056	0.017	0.962	0.028	0.133	0.163	0.720	0.245
0.062	0.032	0.935	0.054	0.141	0.188	0.684	0.275
0.074	0.047	0.901	0.083	0.156	0.208	0.650	0.300
0.080	0.066	0.875	0.110	0.172	0.234	0.615	0.327
0.090	0.086	0.841	0.140	0.194	0.262	0.577	0.351
0.101	0.104	0.814	0.165	0.223	0.296	0.531	0.376
0.113	0.125	0.782	0.193	0.288	0.347	0.457	0.395
288.15 K							
0.062	0.000	0.992	0.000	0.151	0.157	0.749	0.213
0.070	0.022	0.955	0.031	0.176	0.182	0.722	0.237
0.080	0.038	0.938	0.050	0.198	0.208	0.677	0.267
0.095	0.065	0.895	0.089	0.222	0.232	0.642	0.290
0.108	0.083	0.858	0.121	0.237	0.252	0.609	0.308
0.126	0.112	0.815	0.159	0.273	0.276	0.577	0.327
0.142	0.141	0.775	0.192	0.296	0.292	0.525	0.335
298.15 K							
0.084	0.000	0.990	0.000	0.178	0.139	0.788	0.175
0.090	0.019	0.966	0.024	0.193	0.155	0.759	0.196
0.099	0.037	0.933	0.051	0.211	0.181	0.726	0.224
0.111	0.058	0.901	0.079	0.220	0.191	0.713	0.235
0.114	0.059	0.900	0.080	0.249	0.214	0.685	0.251
0.127	0.076	0.877	0.101	0.278	0.234	0.652	0.268
0.139	0.090	0.853	0.121	0.291	0.242	0.642	0.274
0.152	0.108	0.830	0.139	0.309	0.253	0.623	0.283
0.160	0.115	0.823	0.147	0.322	0.256	0.614	0.286
0.168	0.129	0.805	0.162	0.369	0.275	0.576	0.297

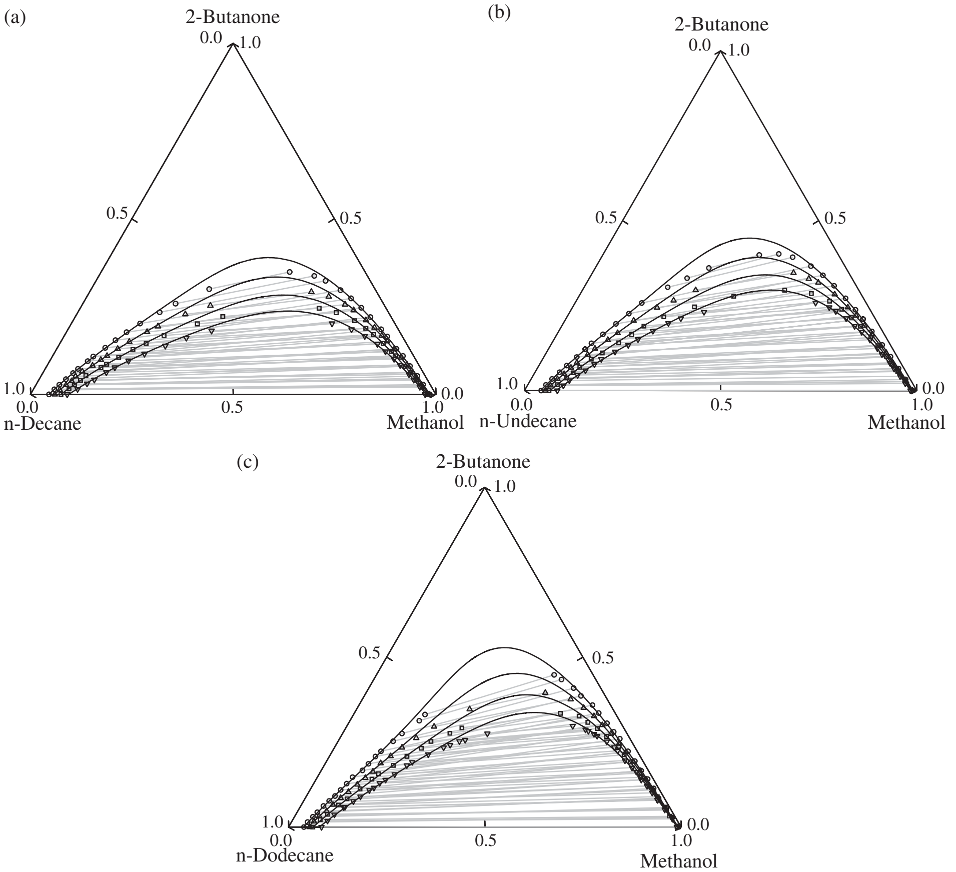

Figure 1. Experimental tie-lines and binodal curves correlated by the NRTL equation for (a) methanol + 2-butanone + *n*-decane, (b) methanol + 2-butanone + *n*-undecane and (c) methanol + 2-butanone + *n*-dodecane. Experimental tie-lines: (o—o), 268.15 K, (△—△), 278.15 K, (□—□), 288.15 K, (∇—∇), 298.15 K; (—) NRTL equation.

systems (methanol + *n*-alkane) have been previously published [7]. Figure 1 shows the experimental data of all mixtures in the whole temperature range. As expected, the size of the two-phase region decreases with an increase in temperature. On the other hand, it can be observed that the immiscibility region also decreases with the alkane chain decreasing.

The UNIQUAC [1] and NRTL [2] equations (Equations (1) and (5), respectively) were applied to correlate the experimental data.

UNIQUAC:

$$\ln\gamma_i = \ln\frac{\Phi_i}{x_i} + \frac{z}{2}q_i\ln\frac{\theta_i}{\Phi_i} + l_i - \frac{\Phi_i}{x_i}\sum_{j=1}^{nc} x_j l_j - q_i\ln\sum_{j=1}^{nc}\theta_j\tau_{ji} + q_i - q_i\sum_{j=1}^{nc}\frac{\theta_j\tau_{ij}}{\sum_{k=1}^{nc}\theta_k\tau_{kj}}x_j l_j \qquad (1)$$

Table 2. Parameters of NRTL and UNIQUAC equations and root mean square deviations σ. The parameters a_{ij} and b_{ij} have units of (cal mol^{-1}) and (cal K^{-1} mol^{-1}), respectively.

Methanol (1) + 2-butanone (2) + *n*-decane (3)

NRTL

ij	12	21	13	31	23	32
a_{ij}	1865.161	−3129.793	3136.721	1681.503	1415.980	44.138
b_{ij}	−9.109	14.725	−7.201	0.339	−5.360	3.513

	$\sigma(x_1^{\mathrm{I}})$	$\sigma(x_2^{\mathrm{I}})$	$\sigma(x_1^{\mathrm{II}})$	$\sigma(x_2^{\mathrm{II}})$
268.15 K	0.017	0.005	0.011	0.003
278.15 K	0.017	0.004	0.008	0.002
288.15 K	0.022	0.003	0.009	0.002
298.15 K	0.012	0.001	0.007	0.001

UNIQUAC

ij	12	21	13	31	23	32
a_{ij}	617.991	−854.662	2167.059	227.606	329.849	−93.808
b_{ij}	−2.443	3.520	−2.455	−0.604	−0.622	0.596

	$\sigma(x_1^{\mathrm{I}})$	$\sigma(x_2^{\mathrm{I}})$	$\sigma(x_1^{\mathrm{II}})$	$\sigma(x_2^{\mathrm{II}})$
268.15 K	0.026	0.008	0.013	0.004
278.15 K	0.029	0.007	0.008	0.003
288.15 K	0.037	0.007	0.008	0.003
298.15 K	0.029	0.004	0.007	0.002

Methanol (1) + 2-butanone (2) + *n*-undecane (3)

NRTL

ij	12	21	13	31	23	32
a_{ij}	1825.197	−2843.129	3046.139	1784.524	956.720	1160.759
b_{ij}	−8.591	13.154	−6.671	0.649	−3.597	−0.629

	$\sigma(x_1^{\mathrm{I}})$	$\sigma(x_2^{\mathrm{I}})$	$\sigma(x_1^{\mathrm{II}})$	$\sigma(x_2^{\mathrm{II}})$
268.15 K	0.025	0.011	0.015	0.007
278.15 K	0.018	0.005	0.006	0.002
288.15 K	0.038	0.007	0.017	0.004
298.15 K	0.011	0.002	0.003	0.001

UNIQUAC

ij	12	21	13	31	23	32
a_{ij}	559.996	−695.940	2090.072	215.510	201.536	88.289
b_{ij}	−2.152	2.863	−1.976	−0.542	−0.068	−0.149

	$\sigma(x_1^{\mathrm{I}})$	$\sigma(x_2^{\mathrm{I}})$	$\sigma(x_1^{\mathrm{II}})$	$\sigma(x_2^{\mathrm{II}})$
268.15 K	0.031	0.012	0.018	0.008
278.15 K	0.025	0.007	0.008	0.003
288.15 K	0.049	0.010	0.018	0.005
298.15 K	0.023	0.004	0.005	0.002

(*Continued*)

Table 2. Continued.

Methanol (1) + 2-butanone (2) + *n*-dodecane (3)

NRTL

ij	12	21	13	31	23	32
a_{ij}	1791.637	−3236.228	3254.803	1072.886	440.236	2568.022
b_{ij}	−8.190	14.231	−7.300	3.811	−1.743	−5.433

	$\sigma(x_1^{\mathrm{I}})$	$\sigma(x_2^{\mathrm{I}})$	$\sigma(x_1^{\mathrm{II}})$	$\sigma(x_2^{\mathrm{II}})$
268.15 K	0.009	0.005	0.003	0.002
278.15 K	0.021	0.008	0.008	0.004
288.15 K	0.017	0.005	0.009	0.003
298.15 K	0.020	0.004	0.005	0.002

UNIQUAC

ij	12	21	13	31	23	32
a_{ij}	738.124	−971.362	2429.476	72.829	211.499	143.527
b_{ij}	−2.733	3.821	−3.069	−0.018	−0.073	−0.348

	$\sigma(x_1^{\mathrm{I}})$	$\sigma(x_2^{\mathrm{I}})$	$\sigma(x_1^{\mathrm{II}})$	$\sigma(x_2^{\mathrm{II}})$
268.15 K	0.010	0.004	0.005	0.002
278.15 K	0.025	0.009	0.010	0.004
288.15 K	0.024	0.007	0.011	0.004
298.15 K	0.029	0.006	0.006	0.003

with

$$l_i = \frac{z}{2}(r_i - q_i) - (r_i - 1) \tag{2}$$

where γ_i is the activity coefficient for compound i, x_i is the molar fraction, z is the coordination number (taken to be 10), nc is the number of compounds, r_i and q_i are the volume and surface area parameters and θ_i and Φ_i are the area and volume fraction, respectively:

$$\theta_i = \frac{q_i x_i}{\sum_{j=1}^{nc} q_j x_j} \tag{3}$$

and

$$\Phi_i = \frac{r_i x_i}{\sum_{j=1}^{nc} r_j x_j} \tag{4}$$

NRTL:

$$\ln \gamma_i = \frac{\sum_{j=1}^{nc} \tau_{ji} G_{ji} x_j}{\sum_{k=1}^{nc} G_{ki} x_k} + \sum_{j=1}^{nc} \frac{x_j G_{ij}}{\sum_{k=1}^{nc} G_{kj} x_k} \left(\tau_{ij} - \frac{\sum_{k=1}^{nc} x_k \tau_{kj} G_{kj}}{\sum_{k=1}^{N} G_{kj} x_k} \right) \tag{5}$$

with

$$G_{ji} = \exp\left[-\tau_{ji} \alpha_{ji}\right] \tag{6}$$

where α_{ji} is the non-random parameter.

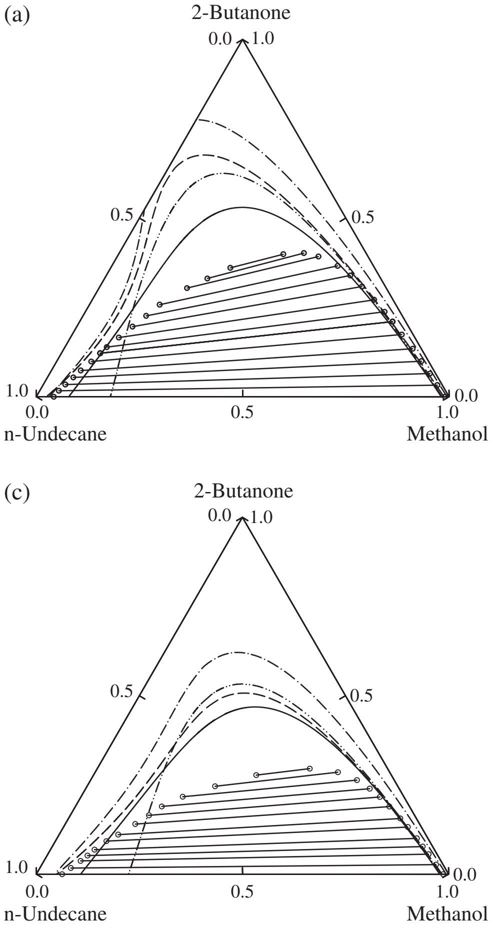

Figure 2. Experimental tie-lines (o——o) and prediction by UNIFAC-Original (——),UNIFAC-LLE (– · · –), UNIFAC-Dortmund (– –) and UNIFAC-Lyngby (– · –) for methanol + 2-butanone + *n*-undecane at (a) 268.15 K, (b) 278.15 K, (c) 288.15 K and (d) 298.15 K.

The temperature dependence of the energetic parameter τ_{ij} for the UNIQUAC and NRTL equations was used, respectively, as follows [8]:

$$\tau_{ij} = \exp -\left\{\left(a_{ij} + b_{ij}T\right)/RT\right\} \tag{7}$$

$$\tau_{ij} = \left(a_{ij} + b_{ij}T\right)/RT \tag{8}$$

where a_{ij} (cal mol^{-1}) and b_{ij} (cal K^{-1} mol^{-1}) are the adjustable parameters, and subscripts $i,j = 1,2,3$ refer to the three components of the ternary mixture under consideration (methanol (1), 2-butanone (2) and *n*-alkane (3)).

Table 2 shows these fitting parameters (a_{ij} and b_{ij}) and their root mean square deviations. The non-random parameter α_{ji} of NRTL equation has been fixed to 0.3 for all cases. A good accuracy has been achieved for all studied mixtures by means of both equations, although NRTL provided best results at the top of the binodal curves. For instance, figure 1 shows the correlation results using the NRTL equation.

Prediction of LLE for the ternary systems studied has been carried out by the UNIFAC group contribution method [3] with parameters obtained from vapour–liquid equlibrium [9] or LLE data [4] (UNIFAC-ELL) and its modifications: UNIFAC-Dortmund [5,10,11] and UNIFAC-Lyngby [6].

For example, figure 2 shows the predicted results of the mixture methanol + 2-butanone + *n*-undecane. An unsatisfactory description was obtained by means of these group contribution methods. All methods overestimate the split region at each temperature and the UNIFAC-Lyngby method provided the worst prediction. This fact could be more clearly observed by the predicted immiscibility and by the non-symmetric tendency towards the 2-butanone + *n*-alkane linear mixture, which estimates the LLE for these binary mixtures at low temperatures. On the contrary, these binary mixtures show a homogeneous experimental trend over the whole range of temperatures [7].

4. Conclusions

The LLE of the mixtures methanol + 2-butanone + *n*-alkane (C_{10}–C_{12}) were determined in the temperature range of 268.15–298.15 K and at atmospheric pressure. Both the UNIQUAC and the NRTL equations were able to describe the correlations in these mixtures, showing low deviation values from the experimental data.

Prediction by means of the UNIFAC group contribution method and its modifications is not suitable. No method is currently available to predict adequately the split region of these mixtures. This fact is observed, especially at low temperatures, where the results are not even qualitative. Such an effect reveals the importance of a wider disposal of thermodynamics liquid split data, and to review the various models for an adequate description of multicomponent heterogeneous systems. Consequently, it is also important to have experimental data or correlation parameters for the design of extraction units.

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