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Liquid−Liquid Equilibrium, Solid−Liquid Equilibrium, Densities, and Refractivity of a Water, Chloroform, and Acetylacetone Mixture

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ABSTRACT: Molybdenum is chemically combined with other elements and usually extracted from acid leaching a residual solution of molybdenite. Understanding the behavior of the solvents is important for the optimization of the separation procedures. Therefore, binary and ternary liquid−liquid equilibria (LLE) were measured for a mixture of chloroform, acetylacetone, and water at (293.15 to 333.15) K and atmospheric pressure (101.3 ± 0.7 kPa). The solid−liquid equilibrium (SLE) between chloroform and acetylacetone was also determined by visual techniques. This system has a single eutectic point. The experimental LLE and SLE data were well-correlated by the nonrandom two-liquid (NRTL) and universal quasichemical activity coefficient (UNIQUAC) models. In addition, the excess molar volume (V^E) and refractivity (n_D) for the binary chloroform + acetylacetone mixture were measured at 298.15 K over the entire composition range. The experimental data were satisfactorily fit using the Redlich−Kister polynomial.

INTRODUCTION

Molybdenum (Mo) is a metallic element that is most frequently used as an alloying addition in alloys and stainless steels. Its alloying versatility is unmatched because its addition enhances the strength, hardenability, weldability, toughness, elevated temperature strength, and corrosion resistance of materials.[1] Mo is therefore used as a thermocouple and anticathode of X-ray tubes in industry; furthermore, it is used in medicine and is a necessary element for the human body and plants.

Currently, Mo is only known to occur chemically combined with other elements in a natural state. Therefore, a separation procedure of Mo from acid leaching a residual solution of molybdenite concentrate by solvent extraction has been studied. For the extraction of Mo from leach liquor, the selected Mo solvent should have a low solubility in water because a high water solubility decreases its extractive potential. Industrially, acetylacetone is used as a selective solvent of Mo from acidic media of roasted Mo ore, and chloroform is used as a modifier in an aqueous phase.[2,3] For the recycling of the solvent and modifier as well as for saving energy in the equilibrium stage separation process, a suitable phase equilibrium and mixture properties are indispensable.

The ternary liquid−liquid equilibrium (LLE) and binary solid−liquid equilibrium (SLE) for the mixtures of mentioned solvent and modifier are not easily available in the literature; thus, in this work, the ternary LLE for the mixture of water + chloroform + acetylacetone and binary SLE for the system chloroform + acetylacetone were measured. In addition, sub-binary LLE for the systems water + chloroform and water + acetylacetone were determined and compared with reported data in the literature.

The binary LLE data were taken at temperatures ranging from (283.15 to 343.15) K, and the ternary data were taken at (298.15 and 323.15) K at atmospheric pressure because extraction at atmospheric pressure has a potential application for a liquid−liquid extraction.[4,5] The SLE for the binary system chloroform + acetylacetone was measured by a visual technique.[6−8] The determined LLE and SLE data were correlated by using the nonrandom two-liquid (NRTL)[9] and universal quasichemical (UNIQUAC) models.[10]

In addition, the density (ρ) and refractivity (n_D) of pure liquids and mixtures of chloroform + acetylacetone at 298.15 K and atmospheric pressure are reported. These ρ data were used to calculate the excess molar volume (V^E) for the same system. The determined V^E values were correlated with composition using the Redlich−Kister polynomial.[11]

EXPERIMENTAL SECTION

Materials. Acetylacetone ($C_5H_8O_2$, M = 100.13 g·mol^{-1}, CAS-RN 123-54-6) was obtained from Aldrich (USA). Water (H_2O, M = 18.02 g·mol^{-1}, CAS-RN 7732-18-5) and chloroform ($CHCl_3$, M = 119.38 g·mol^{-1}, CAS-RN 67-66-3) were supplied by J.T. Baker (USA) and Samchun (Korea), respectively. Their purity was confirmed by gas chromatographic analysis and by comparing the density with literature values. No significant impurities were detected, which would have been revealed by a single sharp peak using gas chromatography, and each supplier shows the chemicals to be greater than 99 % pure in mass. The purities of above chemicals were periodically checked before the experiment by gas chromatography. The thermophysical properties, UNIQUAC parameters, and purities are summarized in Table 1 with the literature values.[12−16]

Apparatus and Procedure. To measure the LLE for the binary and ternary systems, a stirred glass cell was used. The equilibrium cell was heated and insulated by using a silicone oil-filled thermostat (Lauda MD 20, Germany) capable of modulating temperatures ranging from (283.15 to 343.15) K. The uncertainty in the temperature of the thermostat was ± 0.02 K, and the temperature

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Table 1. Physical Properties and UNIQUAC Parameters for Used Chemicals

chemical	ρ/g·cm^{-3} at 298.15 K		n_D at 298.15 K		T_{fus}/K		$\Delta_{fus}H^a$	UNIQUAC^a	
	present study	lit. value	present study	lit. value	present study	lit. value ^{a}	J·mol^{-1}	r	q
water	0.99711	0.9970^a	-	-	-	273.15	-	0.9200	1.4000
chloroform	1.47800	1.4780^b	1.4430	1.4432^d	209.27	209.55	8793.90	2.8700	2.4100
acetylacetone	0.97107	0.9719^c	1.4509	1.4511^e	249.57	249.65	14487.10	4.0192	3.5160

[a] Ref 12. [b] Ref 13. [c] Ref 14. [d] Ref 15. [e] Ref 16.

Table 2. LLE Data for the Binary Systems at Different Temperatures

T/K	water (1) + chloroform (2)		water (1) + acetylacetone (2)	
	aqueous phase	organic phase	aqueous phase	organic phase
	x_1	x_1	x_1	x_1
283.15	0.999	0.008	0.978	0.100
293.15	0.999	0.009	0.972	0.120
298.15	0.999	0.011	0.968	0.125
303.15	0.998	0.013	0.966	0.140
313.15	0.997	0.016	0.959	0.167
323.15	0.996	0.018	0.955	0.196
333.15	-	-	0.948	0.243
343.15	-	-	0.938	0.291

in the equilibrium vessel was monitored using a digital thermometer (ASL F250, UK). The samples in the equilibrium cell were rigorously stirred using a magnetic stirrer. The phases were mixed for about 8 h and then allowed to settle for more than 16 h to achieve the complete separation of the two phases at constant temperature. At the equilibrium state, sampling was carefully carried out on the top for the upper phase and from the bottom for the lower phase. No phases were mixed during the sampling procedure. The sample was analyzed using a gas chromatograph (HP 6890N, USA) with an HP-5 (30 m × 0.32 mm × 0.25 μm) capillary column and a thermal conductivity detector. We estimated the uncertainty of the calculated mole fraction to be less than $\pm 1 \cdot 10^{-3}$.[5] The procedure is described in detail elsewhere.[5,8]

The determination of the melting point as the SLE for a given composition was performed using a triple-jacketed glass still, in which the melting process can be observed visually. The apparatus consists of a glass still, cryostat, and a temperature measuring system. Details of the procedures have been described previously.[6] In the equilibrium cell the chemicals were exposed to nitrogen gas to avoid humidification. The equilibrium temperature was measured with a digital thermometer (ASL F250, UK) when the last crystal disappeared in the equilibrium cell. The accuracy of the equilibrium temperature was determined as ± 0.02 K, and the estimated uncertainty of the mole fraction was less than $\pm 1 \cdot 10^{-4}$.[6]

The densities were measured using a digital vibrating glass tube densimeter (Anton Paar, model DMA 5000, Austria). The densimeter was calibrated with water and dried air before the measurement. The uncertainty of the density measurement was stated by manufacturer less than $\pm 5 \cdot 10^{-6}$ g·cm^{-3} within the range of (0 to 3) g·cm^{-3}, and the temperature accuracy was ± 0.01 K. The mixtures were prepared in 5 mL glass vials by a microbalance (A&D, HA202, Japan) with a precision of $\pm 1 \cdot 10^{-5}$ g. The estimated uncertainty in the mole fraction of the mixture sample was less than $\pm 1 \cdot 10^{-4}$.[6] The refractivity (n_D) was measured by a digital precision refractometer (KEM, model RA-520N, Japan). According to the manufacturer, the uncertainty of this refractometer is $\pm 5 \cdot 10^{-5}$ within the range of 1.32 to 1.40 and $\pm 1 \cdot 10^{-4}$ within the range of 1.40 to 1.58. A further detailed description of the measurement procedure can be found in our previous paper.[6]

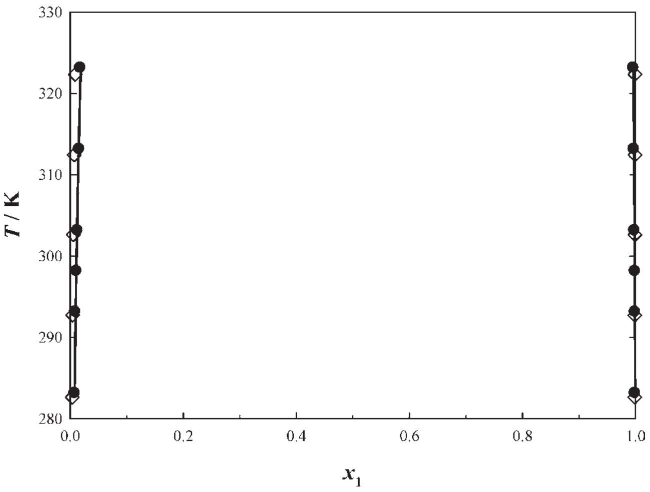

Figure 1. LLE for the binary system of water (1) + chloroform (2) from T = (283.15 to 323.15) K. ●, experimental data; ◇, data from ref 17; —, calculated lines from the UNIQUAC model.

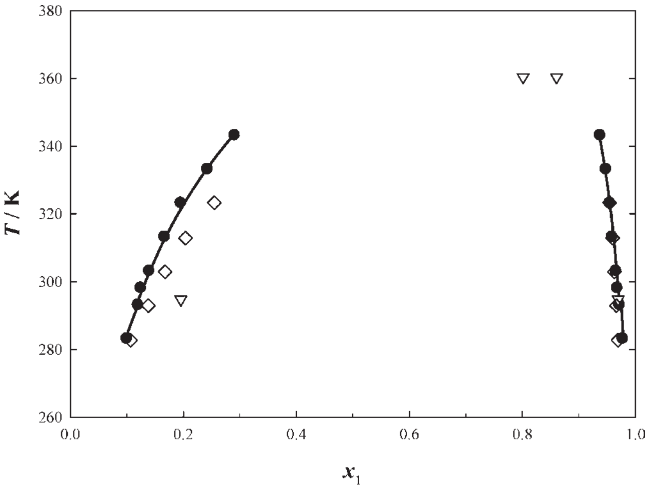

Figure 2. LLE for the binary system of water (1) + acetylacetone (2) from T = (283.15 to 343.15) K. ●, experimental data; ◇, data from ref 17; ▽, data from ref 18; —, calculated lines from the UNIQUAC model.

Table 3. Correlated NRTL and UNIQUAC Model Parameters for the Binary Systems[a]

water (1) + chloroform (2)	NRTL		UNIQUAC	
	ij = 12	*ij* = 21	*ij* = 12	*ij* = 21
a_{ij} (K)	3166.895	1794.990	1420.105	800.179
b_{ij}	−5.652	−3.227	−3.613	−0.434
α	0.20		-	
rmsd (mole %)	0.04		0.04	

water (1) + acetylacetone (2)	NRTL		UNIQUAC	
	ij = 12	*ij* = 21	*ij* = 12	*ij* = 21
a_{ij} (K)	188.657	1379.112	190.450	591.896
b_{ij}	2.403	−3.787	−0.771	−0.029
α	0.20		-	
rmsd (mole %)	0.25		0.17	

[a] Interaction energy parameters: $(g_{ij} - g^{jj})$ or $(u^{ij} - u^{jj})/\mathrm{J \cdot mol^{-1}} = (a_{ij} + b_{ij}T)R$.

Table 4. LLE Data for the Ternary System Water (1) + Chloroform (2) + Acetylacetone (3) at (298.15 and 323.15) K

aqueous phase			organic phase		
x_1	x_2	x_3	x_1	x_2	x_3
		T/K = 298.15 K			
0.968	0.000	0.032	0.125	0.000	0.875
0.977	0.000	0.023	0.123	0.061	0.816
0.988	0.000	0.012	0.113	0.125	0.762
0.990	0.000	0.010	0.105	0.182	0.713
0.993	0.000	0.007	0.088	0.309	0.603
0.993	0.000	0.007	0.075	0.444	0.481
0.995	0.001	0.004	0.047	0.619	0.343
0.998	0.001	0.001	0.025	0.745	0.230
0.998	0.001	0.001	0.016	0.868	0.116
0.999	0.001	0.000	0.011	0.989	0.000
		T/K = 323.15 K			
0.955	0.000	0.045	0.196	0.000	0.804
0.957	0.000	0.043	0.182	0.077	0.741
0.959	0.001	0.040	0.167	0.135	0.698
0.961	0.001	0.038	0.160	0.199	0.641
0.967	0.001	0.032	0.133	0.338	0.529
0.969	0.002	0.029	0.104	0.470	0.426
0.971	0.002	0.027	0.086	0.577	0.337
0.972	0.002	0.026	0.063	0.681	0.256
0.980	0.003	0.017	0.040	0.824	0.136
0.994	0.003	0.003	0.021	0.923	0.056
0.996	0.004	0.000	0.018	0.982	0.000

■ RESULTS AND DISCUSSION

Liquid−Liquid Equilibrium. Tie Lines and Data Correlation Using NRTL and UNIQUAC. The LLE data for the binary systems of water + chloroform and water + acetylacetone at (283.15 to 343.15) K are listed in Table 2 as well as plotted in Figures 1 and 2 with the literature values.[17] The solubility of chloroform in the aqueous phase was negligible, and water solubility in chloroform was less than 0.02 mole in the measured temperature ranges, whereas the solubility of water in acetylacetone was increased significantly by increasing the temperature, and the solubility of acetylacetone in the aqueous phase was slightly increased within the measured temperature ranges. The experimental LLE data showed good agreement with the literature values for the system water + chloroform, while the system water + acetylacetone showed a small difference in the acetylacetone-rich phase.

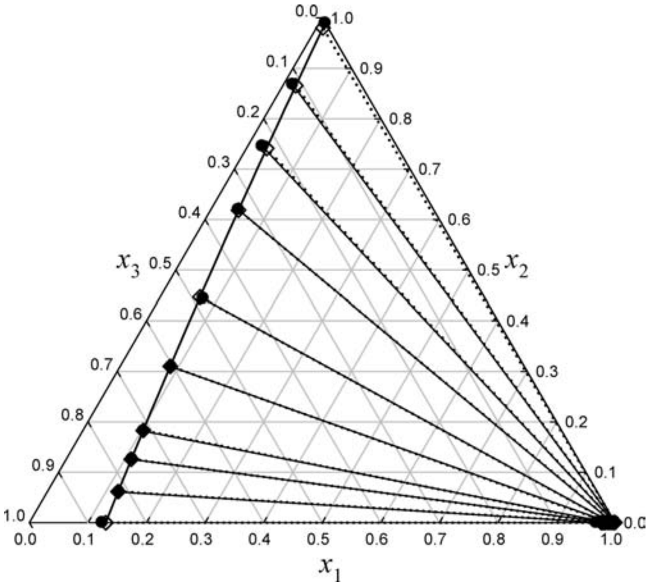

Figure 3. LLE for the ternary system of water (1) + chloroform (2) + acetylacetone (3) at 298.15 K. ●, experimental data; ◇, calculated data using the UNIQUAC model; dotted tie lines and binodal curves were calculated from the UNIQUAC model.

The binary LLE data were correlated with the NRTL and the UNIQUAC models. The adjustable binary parameters of the NRTL and the UNIQUAC models are listed in Table 3, along with the root-mean-square deviations (rmsd's). The UNIQUAC model produced slightly better correlation results with 0.04 and 0.17 % mean deviations of the water mole fraction than the NRTL model. The temperature dependency for the model interaction energy parameters was mentioned below in Table 3.

The measured ternary LLE data for the water + chloroform + acetylacetone mixture at (298.15 and 323.15) K and atmospheric pressure are given in Table 4 and plotted in the form of Gibbs triangles in Figures 3 and 4. As shown in Figure 3, chloroform was distributed much more in the organic phase than the aqueous phase as we expected. However this phenomenon was decreased at higher temperature, 323.15 K, since the solubility of water in acetylacetone. These ternary systems are classified as Treybal type II[19] since they have two partially miscible binaries. The experimental ternary LLE data were also correlated using the NRTL and UNIQUAC models. The parameters for the NRTL and UNIQUAC equations are expressed as

$$\text{NRTL} \qquad \tau_{12} = \frac{g_{12} - g_{22}}{RT} \qquad (1)$$

$$\text{UNIQUAC} \qquad \tau_{12} = \exp\left(-\frac{u_{12} - u_{22}}{RT}\right) \qquad (2)$$

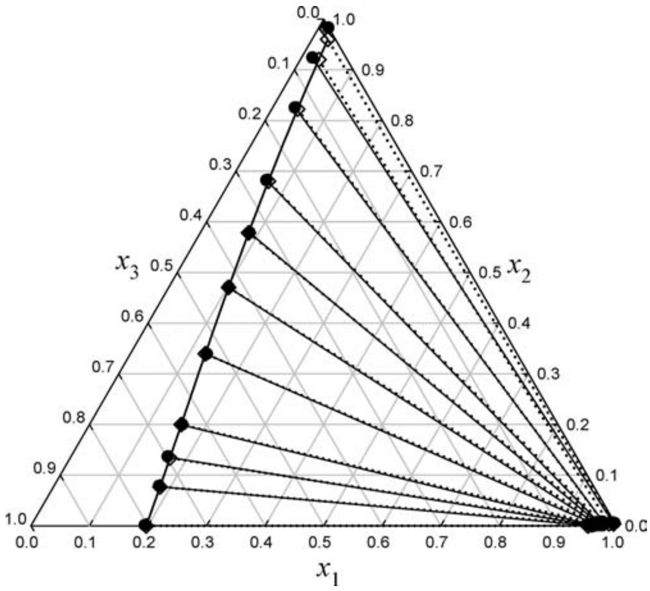

Figure 4. LLE for the ternary system of water (1) + chloroform (2) + acetylacetone (3) at 323.15 K. ●, experimental data; ◊, calculated data using the UNIQUAC model; dotted tie lines and binodal curves were calculated from the UNIQUAC model.

The corresponding sets of binary parameters of both models were determined by minimizing the differences between the experimental and calculated mole fractions for each component over all the measured ternary LLE data. The objective function (OF) used was

$$\mathrm{OF} = \min \sum_i \sum_j \sum_k (x_{ij_k} - x^{\mathrm{c}}_{ij_k})^2 \tag{3}$$

where x and x^{c} are the experimental and calculated mole fractions, respectively. The subscripts i, j, and k denote the component, phase, and tie line, respectively. The optimality of the parameters can be evaluated according to the mean deviation in the compositions of coexisting phases.[20]

The optimized binary parameters are listed in Table 5, with the rmsd values between the experimental and calculated data defined as:

$$\mathrm{rmsd} = \left[\frac{\sum_i \sum_j \sum_k (xi_{j_k} - x^{\mathrm{c}}_{ij_k})^2}{2mN}\right]^{1/2} \tag{4}$$

where m is the number of components and N is the number of tie lines. The dotted lines in Figures 3 and 4 are calculated tie lines using the UNIQUAC model. The experimental and calculated LLE data agreed relatively well as shown in the figures. The parameters of two G^{E} models for ternary systems and mean deviations are given in Table 5. Their rmsd between the determined and the calculated equilibrium compositions are about 0.6 %. In addition, binary experimental data for the systems water + chloroform and water + acetylacetone are compared with calculated data using binary correlated parameters in Table 5. The comparison results were agreed very well within 0.29 % rmsd for the system water + chloroform. However, there was a relatively large deviation of 4 % rmsd for the system water + acetylacetone. Its cause may be from the relatively strong temperature dependency of organic phase of this system, since the binary parameters (from experimental ternary data) were correlated without considering the temperature dependency.

Table 5. Correlated NRTL and UNIQUAC Model Parameters for the Ternary System Water (1) + Chloroform (2) + Acetylacetone (3) at (298.15 and 323.15) K

	NRTL ($\mathrm{J \cdot mol^{-1}}$)			
i–j	$g_{12} - g_{22}$	$g_{21} - g_{11}$	α	rmsd (mole %)
		T/K = 298.15 K		
1–2	11678.676	5550.011	0.20	0.59
2–3	−1566.358	−1694.892		
1–3	8252.310	1714.513		
		T/K = 323.15 K		
1–2	11526.530	4902.267	0.20	0.56
2–3	6210.558	−803.199		
1–3	7913.431	1316.938		
	UNIQUAC ($\mathrm{J \cdot mol^{-1}}$)			
i–j	$u_{12} - u_{22}$	$u_{21} - u_{11}$		rmsd (mole %)
		T/K = 298.15 K		
1–2	3807.895	5357.708		0.38
2–3	3439.668	−3710.289		
1–3	−164.567	4530.964		
		T/K = 323.15 K		
1–2	2326.340	4815.053		0.46
2–3	1599.946	248.572		
1–3	−538.789	4994.802		

Table 6. SLE Data for the Binary System Chloroform + Acetylacetone

x_1	T/K	x_1	T/K
0.0000	249.57	0.6926	197.40
0.1001	245.61	0.7100	194.03
0.1990	240.98	0.7879	195.02
0.2900	235.50	0.8100	197.18
0.4100	226.95	0.8999	204.24
0.4999	219.14	0.9498	207.19
0.6001	208.65	1.0000	209.27
0.6510	202.85		

Solid–Liquid Equilibrium. For the SLE in the binary system with the pure solid phase of component i and the saturated solution of i as the second phase, one can derive the following relation between mole fraction of component i (x_i) and temperature (T):

$$\ln(\gamma_i x_i) = -\frac{\Delta H_{\mathrm{fus},\,i}}{RT}\left[1 - \frac{T}{T_{\mathrm{fus},\,i}}\right] \tag{5}$$

where x_i is the mole fraction in the liquid phase, γ_i the activity coefficient in the liquid phase, $\Delta H_{\mathrm{fus},i}$ the molar enthalpy of fusion, $T_{\mathrm{fus},i}$ the melting temperature, T the absolute temperature, and R is the universal gas constant.

The results of the determined SLE data for the binary system of chloroform + acetylacetone are listed in Table 6 and plotted in Figure 5. This binary system has a single eutectic point as shown in the figure. The calculated G^{E} model parameters are given together with the rmsd in Table 7. The rmsd was calculated from eq 6.

$$\text{rmsd} = \sqrt{\frac{1}{n}\sum_{i=1}^{n}(T_{\text{exp}} - T_{\text{cal}})^2} \qquad (6)$$

where n is the number of experimental points; T_{cal} is the melting temperature calculated by G^E model, and T_{exp} represents the experimental melting temperature. NRTL and UNIQUAC models were used for the correlation of this SLE data. For the NRTL model, the constant of proportionality like the nonrandomness parameter ($\alpha = 0.31$) was taken into account in the calculations.[21] The NRTL model gave better correlation results than the UNIQUAC model for the measured system, and the rmsd of the equilibrium temperature from eq 6 was less than 0.40 K. The interpolated eutectic point for the chloroform + acetylacetone system at 191.07 K was given by NRTL model as $x_1 = 0.7325$.

Densities and Refractivity. The ρ and n_D for the binary system chloroform + acetylacetone were measured using a digital vibrating U-tube densimeter and a precision digital refractometer. The binary V^E values were then derived using eq 7 from the measured ρ.

$$V^E(\text{cm}^3\cdot\text{mol}^{-1}) = \frac{\sum_i x_i M_i}{\rho_m} - \sum_i\left(\frac{x_i M_i}{\rho_i}\right) \qquad (7)$$

where M_i is the molecular weight of a pure component in mixture.

The values of V^E are listed in Table 8 and plotted in Figure 6.

In the case of V^E, the system of chloroform + acetylacetone showed a negative deviation throughout the entire range of compositions. The binary V^E data correlated well with the Redlich–Kister polynomial of the following form.

$$V^E/\text{cm}^3\cdot\text{mol}^{-1} = x_1 x_2 \sum_{i=1}^{n} A_i(x_1 - x_2)^{i-1} \qquad (8)$$

Table 9 shows the correlated Redlich–Kister parameters with the standard deviation. The solid line in Figure 6 illustrates the calculated V^E with the correlated Redlich–Kister parameters. Besides, V^E data were compared with reported values.[16] As

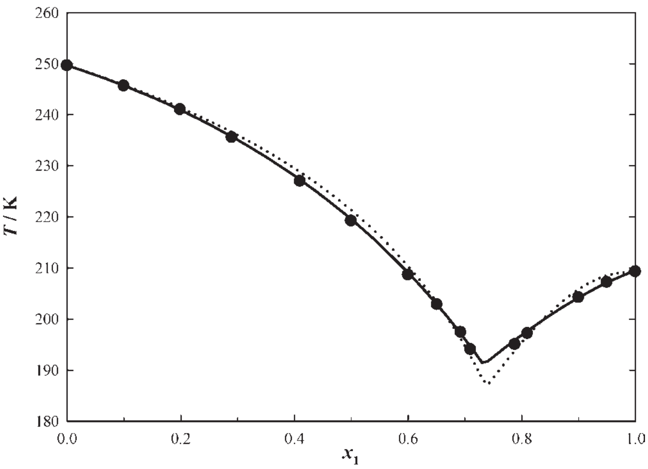

Figure 5. SLE for the system of chloroform (1) + acetylacetone (2). ●, experimental data; —, calculated line from NRTL model; dotted tie line was calculated from UNIQUAC model.

Table 8. Densities, Excess Molar Volumes, and Refractivity for the Measured Binary System at 298.15 K

		ρ	V^E	
system	x_1	$\text{g}\cdot\text{cm}^{-3}$	$\text{cm}^3\cdot\text{mol}^{-1}$	n_D
chloroform (1) + acetylacetone (2)	0.0498	0.9885	−0.037	1.4508
	0.0996	1.0092	−0.058	1.4497
	0.2007	1.0526	−0.085	1.4496
	0.3002	1.0972	−0.110	1.4494
	0.4002	1.1443	−0.131	1.4492
	0.4999	1.1936	−0.156	1.4487
	0.5997	1.2454	−0.175	1.4482
	0.7007	1.3004	−0.183	1.4475
	0.7997	1.3568	−0.164	1.4465
	0.8990	1.4156	−0.105	1.4450
	0.9494	1.4463	−0.049	1.4442

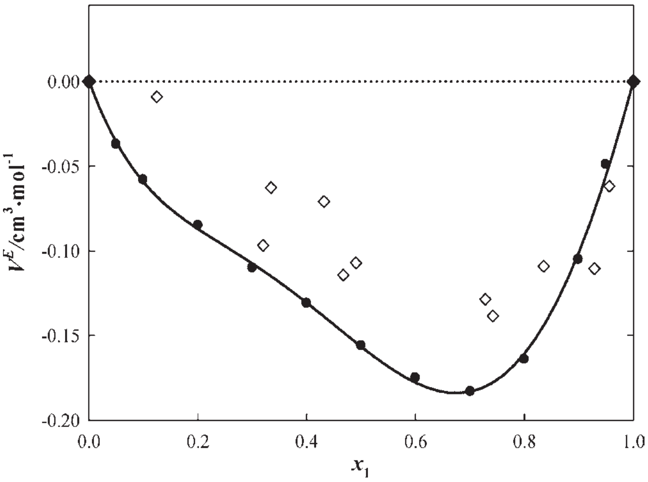

Figure 6. V^E for the system of chloroform (1) + acetylacetone (2) at 298.15 K. ●, experimental data; ◇, data from ref 16; —, calculated data using the Redlich–Kister polynomial.

Table 7. Correlated NRTL and UNIQUAC Model Parameters and rmsd for SLE Data

	NRTL ($\text{J}\cdot\text{mol}^{-1}$)			
	$g_{12} - g_{22}$	$g_{21} - g_{11}$	α	rmsd (K)
chloroform (1) + acetylacetone (2)	288.02	−2300.25	0.31	0.39
	UNIQUAC ($\text{J}\cdot\text{mol}^{-1}$)			
	$u_{12} - u_{22}$	$u_{21} - u_{11}$		rmsd (K)
chloroform (1) + acetylacetone (2)	5100.80	−2358.14		1.16

Table 9. Fitted Redlich−Kister Parameters with the Standard Deviation for the System Chloroform + Acetylacetone at 298.15 K

system	excess property	A_1	A_2	A_3	A_4	σ_{st}
chloroform (1) + acetylacetone (2)	V^E	−0.626	−0.505	−0.410	0.339	0.004

shown in the figure, the literature values scattered significantly compare to our experimental data.

CONCLUSIONS

The solubility of chloroform in water was negligible and vice versa was less than 2 % in the measured temperature ranges, whereas the solubility of water in acetylacetone was increased significantly by increasing the temperature, and the solubility of acetylacetone in the aqueous phase was slightly increased within the measured temperature ranges. The ternary LLE data for the system of water + chloroform + acetylacetone are of Treybal type II, having two partially miscible binaries. The extractant of molybdenum, acetylacetone, was less soluble than ca. 3.5 % at 298.15 K and ca. 4.5 % at 323.15 K. Therefore, the ternary LLE data correlated well with the UNIQUAC model, with a rmsd of about 0.5 in mole percent.

The solid−liquid equilibrium region for the binary system chloroform + acetylacetone has a single eutectic point. The binary SLE data correlated well with the NRTL model, with a rmsd less than 0.4 K.

The thermodynamic quantities of V^E and refractivity for the system chloroform + acetylacetone at 298.15 K were investigated, and V^E showed negative values throughout the entire range of composition. These data correlated very well with the Redlich−Kister polynomial.

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