

Influence of temperature on the liquid–liquid equilibria containing two pairs of partially miscible liquids Water + furfural + 1-butanol ternary system ¹

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Abstract

In order to show the influence of the temperature on the liquid–liquid equilibria for water + furfural + 1-butanol ternary system, the solubilities and equilibrium data starting at 298.15 K have been measured. Between 298.15 K and 376.6 ± 0.5 K, binodal band type curves are present, but above this last value, two separate binodal curves appear. The solubility data for water + furfural binary system is also reported. From the solubility curve, the upper critical solution temperature (UCST) was interpolated: 396 K and mass fraction of water equal to 0.49. When the temperature is decreased below the lower CST, the ternary system presents two binodal curves expanding toward each other until they meet with a common plait point at the lowest point on the ridge (saddle point, or col), which is situated at 376.6 ± 0.5 K with a ternary composition: $w_1 = 0.64$; $w_2 = 0.21$ and $w_3 = 0.15$. Tie-line data were satisfactorily correlated by the Othmer and Tobias method on a mass fraction basis, and their plait point coordinates were estimated when two separate binodal curves were present. From experimental distribution coefficients and selectivities, 1-butanol appears to be a good solvent for extraction purposes, particularly when the process is carried out at room temperature. © 1998 Elsevier Science B.V. All rights reserved.

Keywords: Chemical equilibria; Ternary system; Liquid–liquid equilibria; Water; Furfural; 1-butanol

1. Introduction

In previous studies [1,2], we obtained the liquid–liquid equilibria for (water + 4-hydroxy-4-methyl-2-pentanone + benzyl alcohol) and (water + nitromethane + 1-butanol) ternary systems up to their

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corresponding upper critical solution temperature (UCST) and clarified the effects of temperature on the binodal curves and on their distribution behaviour.

As part of a continuing study of the effect of temperature on liquid–liquid equilibria (LLE), this work reports extensive data for the ternary system: water + furfural + 1-butanol (W + F + B), and one partially miscible binary system: water + furfural (W + F) from room temperature (298.15 K) up to the highest critical solution temperature of the two partially miscible binaries for the ternary system (403.15 K). No data about this ternary system was found in the literature, but solubility data for both binaries were previously published [2–4], and our experimental results are in agreement with those. Additionally, the capability of 1-butanol for separating furfural from aqueous solutions was analyzed by using experimental distribution coefficients and selectivities at different temperatures.

An equilateral triangular prism was used in order to show binaries and ternary solubility–temperature relationships and the two binary UCST. Solubilities or binodal curves at 298.15, 323.15, 348.15, 373.15, 376.15, 383.15 and 398.15 K for the (W + F + B) ternary system were obtained in order to construct this prism, while equilibrium data were only obtained at 298.15, 323.15, 348.15, 373.15 and 383.15 K with the same objective.

Tie-line data were satisfactorily correlated by the Othmer and Tobias method [5] and the plait point coordinates were estimated from it when two separate binodal curves were present.

2. Experimental

2.1. Materials

Furfural (2-furaldehyde, Riedel de Haën, R.G.) was fractionally distilled at reduced pressure under nitrogen atmosphere, and only the colourless middle fraction was collected. After its purification, a

Table 1

Experimental values of density (ρ) and refractive index (n_D) of pure components at 298.15 K, and mutual solubilities of the partially miscible binary system: water(1) + furfural(2)

Compound	ρ (kg m ⁻³)		n_D	
	Experimental	Literature	Experimental	Literature
W	997.1	997.0474 ^a	1.3328	1.3325029 ^a
F	1159.8	1154.5 ^a	1.5232	1.52345 ^a
B	805.8	805.75 ^a	1.3974	1.39741 ^a

Mutual solubility (mass fraction percentage)	W in solvent		Solvent in W		
	T (K)		T (K)		
	Experimental	Literature	Experimental	Literature	
	298.15	6.0	5.0 ^b	8.0	8.1 ^b
	323.15	7.0	7.3 ^c	9.3	9.7 ^c
	348.15	10.75	10.8 ^c	13.00	12.1 ^c
	373.15	17.25	17.2 ^c	19.50	19.5 ^c
	376.15	18.50	19.0 ^c	21.00	20.8 ^c
	383.15	21.50	24.0 ^c	25.00	24.0

^aJ.A. Riddick, W.B. Bunger, T.K. Sakano, Organic Solvents, 4th edn., Wiley, New York, 1986, pp. 74, 198 and 708.

^bAverage values among several data from Ref. [3].

^cInterpolated from Ref. [3].

chromatographic analysis showed that its purity was greater than 99.5 mass%. This same analysis was also made with dark furfural in order to determine the concentration of its impurities which appear when it is heated, because they could produce important deviations respect to the real equilibrium data. The experimental results showed that furfural always had greater than 99 mass% purity. 1-Butanol (Merck) was used without further purification because its purity was similar to that of purified furfural. This degree of purity was considered to be adequate since no abnormalities were observed in the experimental coexistence temperature–composition curves for the binaries [6]. Bi-distilled water was used for the determination of equilibrium data.

Table 1 shows densities and refractive indices of the pure components together with those reported in the literature for comparison. Mutual solubilities are also included.

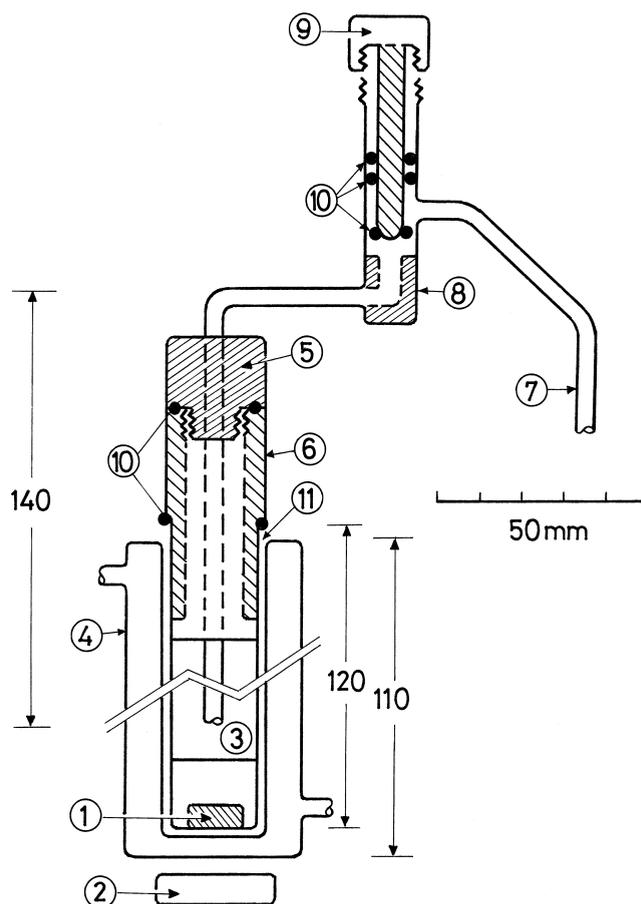


Fig. 1. Line drawing of the experimental device for tie line determination at high temperature. (1) Magnetic stirring bar, (2) magnetic stirrer, (3) Pyrex glass cell, (4) jacket for circulating the isothermal fluid, (5) stainless steel threaded connection, (6) Teflon threaded connection to 5 and Pyrex glass cell, (7) stainless steel tube (o.d. 1/8"), (8) stainless steel valve seat, (9) Teflon valve, (10) Viton O-ring and (11) annular space filled with polyethylene glycol 400.

2.2. Procedure

The solubility curve for the partially miscible binary system (W + F) was determined as previously reported [2]. The samples were brought into thermal equilibrium in a thermostatic bath at their

Table 2
Experimental temperature–composition data for the water(1)+furfural(2) binary system

wt.%,	<i>T</i> (K)
91.8	293.75 ^a
91.8	298.55 ^a
91.7	304.95 ^a
91.2	308.65 ^a
90.8	313.75 ^a
90.3	318.35 ^a
90.9	323.05 ^a
90.05	328.1 ^b
89.95	332.6 ^b
88.70	337.7 ^b
88.50	341.7 ^b
88.25	348.1 ^b
80.40	370.5 ^b
81.80	367.7 ^b
75.25	386.5 ^b
68.45	391.7 ^b
64.10	391.4 ^b
63.10	392.9 ^b
54.95	393.2 ^b
58.85	394.7 ^b
49.80	394.5 ^b
46.45	395.7 ^b
45.65	396.4 ^b
40.35	396.6 ^b
34.90	393.5 ^b
30.80	392.1 ^b
22.10	383.7 ^b
18.15	375.8 ^b
8.50	341.7 ^b
8.55	337.6 ^b
8.25	332.6 ^b
7.70	328.2 ^b
7.3	323.05 ^a
6.7	318.25 ^a
6.3	312.75 ^a
7.0	308.05 ^a
6.7	303.15
5.8	298.35 ^a
49 ^c	396 ^c

^aObtained with the titration method.

^bObtained with the sealed-tube technique.

^cUCST obtained by graphical interpolation from Fig. 2.

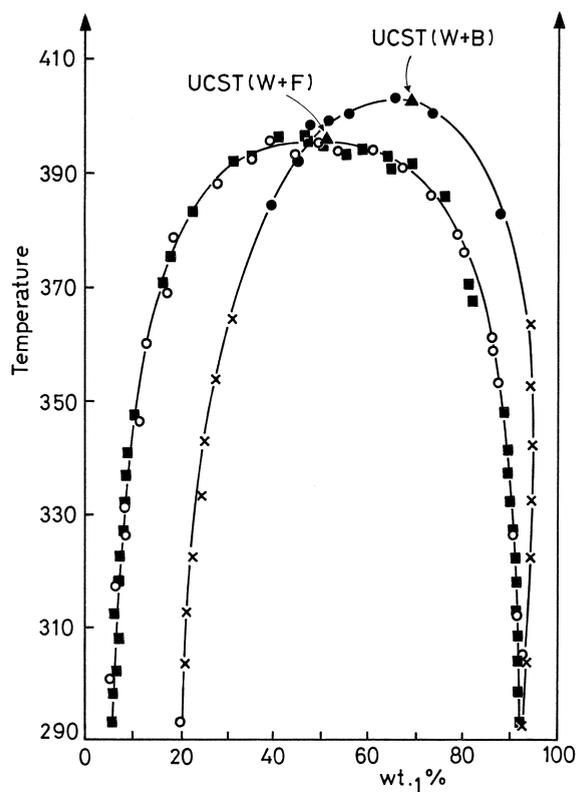


Fig. 2. Experimental coexistence temperature–composition curves for water + furfural: (■), this paper and (○), from Ref. [3]; and water + 1-butanol: (●), from Ref. [2] and (x), from Ref. [4]. ▲, Binaries UCSTs.

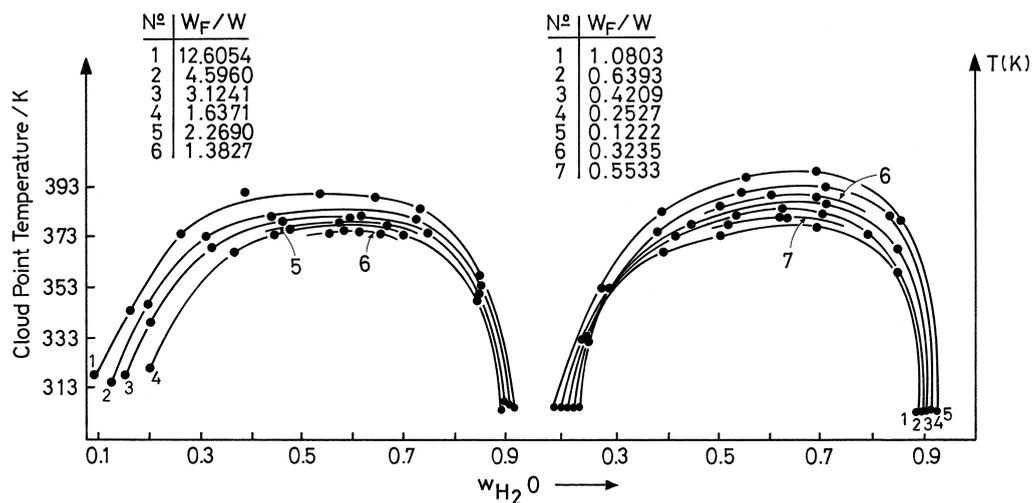


Fig. 3. Correlation of cloud-point data for the water(1) + furfural(2) + 1-butanol(3) ternary system. Constant mass fraction ratios of furfural to 1-butanol (w_F/w_B).

Table 3
Binodal curves at several temperatures for the ternary system water(1) + furfural(2) + 1-butanol(3)

(a) Band type curves

Water-rich phase		Organic-rich phase	
wt. ₁ %	wt. ₂ %	wt. ₁ %	wt. ₂ %
<i>298.15 K</i>			
92.0	8.0	6.7	89.3
91.3	5.8	10.2	83.7
90.0	3.9	11.4	77.3
92.3	1.7	11.9	72.1
92.0	0.0	13.6	66.9
		15.4	60.4
		16.0	56.9
		16.7	54.4
		18.9	46.7
		19.6	43.6
		21.1	40.2
		19.8	39.9
		19.3	36.0
		21.6	35.5
		21.7	30.0
		21.8	29.8
		21.9	27.0
		23.0	24.1
		22.5	20.9
		21.9	18.2
		23.5	16.5
		21.8	15.2
		22.2	12.2
		20.9	9.6
		20.0	5.0
		20.3	0.0
<i>323.15 K</i>			
90.8	9.3	7.0	93.0
90.0	9.3	11.0	82.5
89.5	8.6	14.3	70.4
88.8	8.6	16.8	63.4
88.5	7.1	20.8	49.2
88.8	5.8	22.8	40.1
89.5	4.1	23.3	29.9
90.3	2.9	23.8	22.6
90.8	1.9	23.3	15.5
92.0	0.9	20.5	8.7
93.5	0.0	22.5	0.0
<i>348.15 K</i>			
87.0	13.0	10.8	89.3
86.5	12.5	17.8	76.2
86.3	11.3	20.3	65.5
85.3	11.2	23.3	58.4

Table 3 (continued)

(a) Band type curves

Water-rich phase		Organic-rich phase	
wt. ₁ %	wt. ₂ %	wt. ₁ %	wt. ₂ %
<i>348.15 K</i>			
84.5	9.6	27.5	45.0
86.5	7.0	28.8	37.0
87.5	4.9	28.3	28.0
88.0	3.6	27.5	21.5
89.5	2.1	27.3	14.7
90.8	1.0	25.5	8.1
93.5	0.0	26.0	0.0
<i>373.15 K</i>			
80.5	19.5	17.3	82.8
79.0	19.5	25.5	69.0
77.5	18.5	30.8	56.9
76.0	18.3	35.5	49.1
74.0	18.1	39.0	42.3
73.0	16.8	42.5	35.7
72.5	16.0	48.5	29.9
77.0	12.0	46.5	27.8
79.8	7.9	41.5	22.8
83.3	5.0	38.8	18.1
86.0	2.8	36.8	12.8
87.3	1.4	34.0	7.2
90.5	0.0	34.0	0.0
<i>376.15 K</i>			
79.0	21.0	18.5	81.5
77.8	20.6	27.0	67.6
75.8	19.9	33.0	55.0
73.8	20.0	39.5	46.0
70.5	20.5	43.5	39.2
67.0	20.5	49.3	31.5
65.0	20.0	57.5	24.7
72.0	14.5	54.0	23.9
77.3	8.9	45.5	21.3
79.3	7.4	44.3	19.9
81.8	5.4	41.5	17.3
85.3	3.0	38.8	12.4
86.5	1.5	36.8	6.9
90.0	0.0	35.5	0.0

(b) Two separate binodal curves

Water-rich phase		Furfural-rich phase		Water-rich phase		Butanol-rich phase	
wt. ₁ %	wt. ₂ %	wt. ₁ %	wt. ₂ %	wt. ₁ %	wt. ₂ %	wt. ₁ %	wt. ₂ %
<i>383.15 K</i>							
75.0	25.0	21.5	78.5	67.0 ^a	12.0 ^a	67.0 ^a	12.0 ^a
73.5	24.6	32.0	63.0	76.0	7.1	52.0	14.2

Table 3 (continued)

(c) Only one binodal curve

Water-rich phase		Furfural-rich phase		Water-rich phase		Butanol-rich phase	
wt. ₁ %	wt. ₂ %	wt. ₁ %	wt. ₂ %	wt. ₁ %	wt. ₂ %	wt. ₁ %	wt. ₂ %
383.15 K							
64.8	29.0	52.0	39.4	82.2	3.6	43.8	11.3
64.0 ^b	29.0 ^b	64.0 ^b	29.0 ^b	84.0	1.7	39.5	6.6
				88.5	0.0	38.5	0.0
398.15 K							
				71.8	3.1	58.0	4.6
				78.0	0.0	51.5	0.0

^aEstimated plait point from Eq. (1).^bEstimated plait point from Eq. (2).

equilibrium vapor pressure (using water up to 323.15 K and polyethylene glycol 400 above this temperature), and subjected to changes in temperature with heating rates of about 0.1 K min⁻¹. The change in solubility of the (W + F) binary system with externally applied pressure was ignored because it is very small [7], for our range of vapor pressure which was always lower than 3 atm at the highest temperature. Repeated measurements of the temperatures at which the opalescence of each sample disappeared gave the same value within ± 0.1 K. Thus, the transition temperatures reported here are the mean of several determinations for each of the compositions studied. The uncertainties in the observed values were determined by the accuracy of the observation of the transition points for the titration method and the sealed-tube technique with an accuracy in the temperature and composition of ± 0.05 K and ± 0.001 of mass fraction for the first method, and ± 0.1 K and ± 0.0005 of mass fraction for the second one.

The binodal curve and tie-line data for the ternary system at 298.15 K were obtained as described previously with an estimated accuracy in composition and temperature within ± 0.0005 and ± 0.05 K, respectively [8]. The two end points of the tie-lines were obtained by using a calibration curve: density against mass fraction of F as earlier [1], where the densities were measured with a vibrating tube densimeter KEM DA300 with an accuracy of ± 0.1 kg m⁻³. The binodal curves for the ternary system at 323.15, 348.15, 373.15, 376.15, 383.15 and 398.15 K were constructed by graphical interpolation of the cloud-point data. Several binary solutions with different known mass fraction ratios of F to B ($w_F/w_B = \text{constant}$) were prepared, each one divided into five or six aliquots and different volumes of water added to them in order to make the heterogeneous ternary mixtures. The temperature at which cloudiness disappears in these ternary mixtures of known composition [9] was measured. These points were recorded as the cloud-point temperatures (± 0.1 K) and plotted against the mass fraction of water (± 0.001 of mass fraction).

All ternary measurements were carried out in a Pyrex glass cell with a spherical female ground joint hermetically closed with a male one, which was equipped with a magnetic stirrer and a jacket for circulating the isothermal liquids at the desired temperature within ± 0.05 K.

Tie-lines at temperatures higher than 298.15 K were obtained using the same procedure described earlier [2], based in the Briggs and Comings method [9], but in this case, two experimental devices to

withdraw the upper phase were used. For 232.15 and 348.15 K, we used a thermostatic stainless steel syringe attached to the cell [2,10], while for 373.15 and 383.15 K, a device based on the well-known siphon technique was used. Fig. 1 shows a line drawing of this last experimental device for tie-line determination at the two higher temperatures for this ternary system.

The sample from the upper phase was allowed to reach a new equilibrium at 298.15 K, and the compositions of the newly formed conjugate phases were determined through their densities with the help of the calibration curve (density against mass fraction of F) and the distribution curve (mass fraction of F in the extract phase against mass fraction of F in the raffinate one) [1,10]. This tie-line intersects the higher temperature binodal curve at a point representing the composition of the upper phase removed at the higher temperature. A line extending from this point through the overall composition of the original mixture to the other part of the binodal curve is a tie-line at the higher temperature. Taking into account the accumulative errors in all graphic interpolations included in this

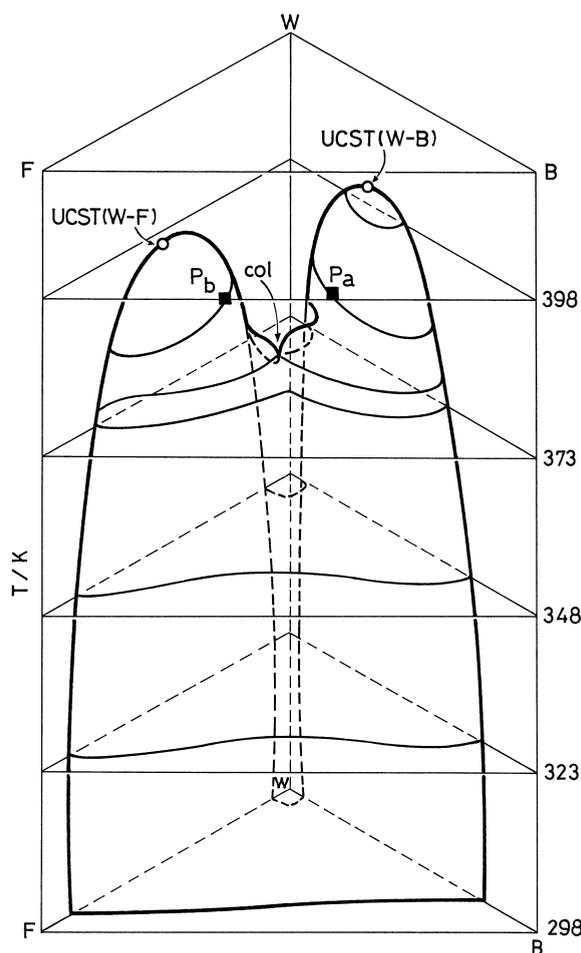


Fig. 4. Schematic equilateral triangular prism for the water(1)+furfural(2)+1-butanol(3) ternary system, showing both binary UCSTs and the col position. Plait points (P_a and P_b) are also shown.

method, the errors of these solubilities and equilibrium compositions were estimated to be within ± 0.001 and ± 0.005 of mass fraction, respectively.

3. Results and discussion

The experimental values for the solubility curve of the (W + F) binary system are given in Table 2, and shown in Fig. 2, together with bibliographic data for comparison. The corresponding UCST is 396 K with $w_1 = 0.49$ obtained by graphic interpolation from Fig. 2 can also be seen.

The curves showing the cloud-point temperatures vs. the mass fraction of water are shown in Fig. 3. Taking into account that the mass fraction ratio between F and B is known for each curve and that the sum of the mass fractions of all components is equal to 1, the ternary compositions of the binodal curves at 323.15, 348.15, 373.15, 376.15, 383.15 and 398.15 K can be obtained from these curves. They are listed in Table 3.

The binodal surface in the solid diagram inside the equilateral triangular prism is a ridge across it connecting the solubility curves of the two partially miscible binary systems. If the ridge is concave upward, an isotherm may cut-off two separate binodal curves as can be seen in Fig. 4 above 377 K, approximately. Points P_a and P_b are plait points at the indicated temperatures, whose compositions were obtained with the Othmer and Tobias' correlation.

Fig. 5 shows the isotherm projected onto the base of the prism, where binodal band type curves can be observed below 377 K and two separate binodal curves above it. These last curves tend to expand

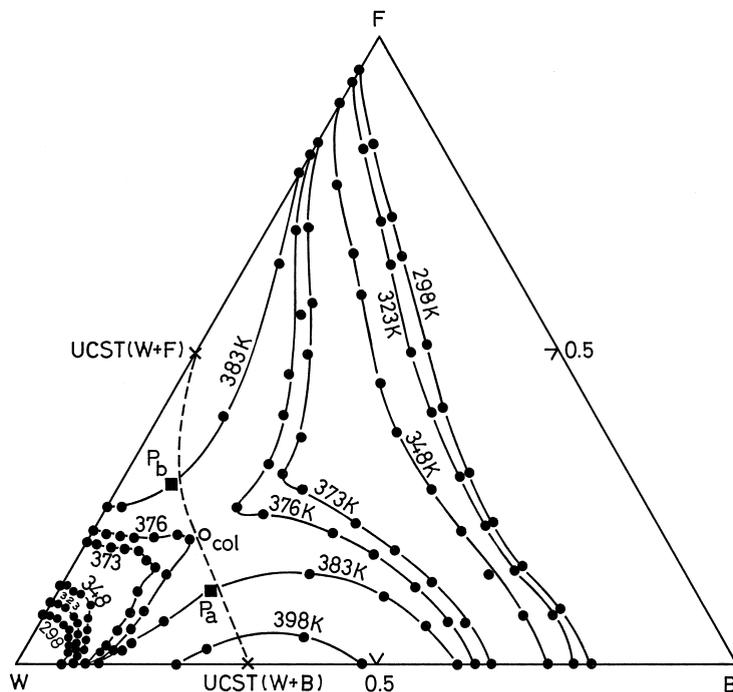


Fig. 5. Isothermal projected onto the base of the prism for the system water(1) + furfural(2) + 1-butanol(3), showing the ridge (dashed line) connecting all plait points and passing by the col, and both binary UCSTs.

toward each other until they touch at a common plait point, when the temperature is decreased. This is a saddle point (also called ‘col’) which is situated at 376.6 ± 0.5 K with a ternary composition: $w_1 = 0.64$; $w_2 = 0.21$ and $w_3 = 0.15$ for this system, obtained graphically from Fig. 5 from the

Table 4

Equilibrium data at several temperatures for the ternary system water(1) + furfural(2) + 1-butanol(3)

(a) Band type curves

Water-rich phase		Organic-rich phase	
wt. _{1R} %	wt. _{2R} %	wt. _{1E} %	wt. _{2E} %
<i>298.15 K</i>			
92.5	7.5	10.0	83.0
91.5	7.0	12.5	72.0
91.5	6.5	15.5	60.0
91.0	6.0	18.0	50.0
90.5	5.0	21.5	38.5
90.0	4.5	22.0	31.5
90.5	3.5	22.5	22.0
91.5	2.0	23.0	14.5
92.5	1.0	20.0	7.0
<i>323.15 K</i>			
90.0	9.5	13.0	77.0
89.5	8.5	16.5	64.5
88.0	7.5	19.5	55.0
88.5	6.0	21.0	48.0
89.0	6.0	23.0	34.0
89.0	5.5	24.0	30.5
89.0	5.0	23.5	19.5
90.5	3.0	22.5	14.0
90.5	2.5	21.0	7.5
<i>348.15 K</i>			
87.0	12.0	19.0	72.0
86.0	11.5	21.5	66.0
86.0	11.0	26.0	50.0
86.0	7.5	28.0	39.5
86.0	7.0	28.5	34.0
86.5	7.0	28.0	29.0
86.5	6.5	27.5	20.5
87.0	6.0	27.0	14.0
89.5	2.0	26.0	8.0
<i>373.15 K</i>			
77.5	19.0	28.0	64.0
76.5	18.5	33.0	52.5
74.5	18.0	39.0	41.5
74.0	18.0	43.0	35.0
82.0	6.0	40.0	21.0
83.0	5.0	36.5	14.0
88.0	1.5	34.0	5.0

Table 4 (continued)

(b) Two separate binodal curves

Water-rich phase		Furfural-rich phase		Water-rich phase		Butanol-rich phase	
wt. _{1R} %	wt. _{2R} %	wt. _{1E} %	wt. _{2E} %	wt. _{1R} %	wt. _{2R} %	wt. _{1E} %	wt. _{2E} %
383.15 K							
75.0	24.5	28.0	69.0	78.0	6.0	46.5	13.5
74.0	24.5	34.0	61.0	79.5	5.0	42.0	11.0
69.5	26.0	48.0	44.0	81.5	4.0	39.0	6.0

Parameters from Eq. (1): $a = 0.3460$; $b = -0.6093$; $r^2 = 0.9974$.Parameters from Eq. (2): $a = 0.3411$; $b = -0.3867$; $r^2 = 0.9955$.

intersection of the asymptotes of the two hyperbola-like curves at 376.6 K [11]. The ridge is indicated by a curve (dashed line, in Fig. 5) connecting all plait points and both binary UCST. The high non-ideality of this ternary system is probably the responsible of the pronounced curvature for this curve in opposition to the behaviour observed in other systems [2,11]. Below the col isotherm, there are band type curves, but between it and the lower UCST of the binaries, there are two separate binodal curves. Above the lower UCST, only one binodal curve is present, while the higher UCST was not surpassed. Consequently, the ternary system can be classified as type 1 in the Treybal's classification [7] in this last situation.

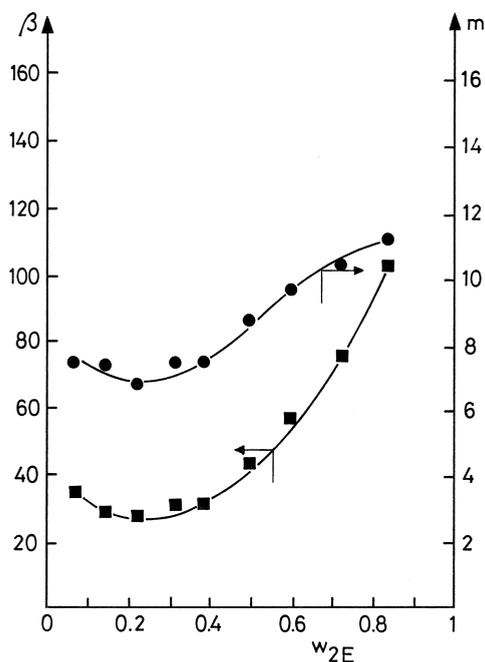


Fig. 6. Distribution coefficients (m) and selectivities (β) against mass fraction of furfural in the extract phase (w_{2E}) for the system water(1)+furfural(2)+1-butanol(3) at 298.15 K.

Table 4 shows tie-line data for the ternary system at 298.15, 323.15, 348.15, 373.15 and 383.15 K.

Whenever two binodal curves were present, the tie-lines were satisfactorily correlated by the method of Othmer and Tobias [5]. Their empirical equation can represent all tie-line data with adequate precision both for the pair partially miscible: water(1)-1-butanol(3):

$$\log[(1 - W_{1R})/W_{1R}] = a \log[(1 - W_{3E})/W_{3E}] + b \quad (1)$$

and for the pair partially miscible: water(1)-furfural(2):

$$\log[(1 - W_{1R})/W_{1R}] = a' \log[(1 - W_{2E})/W_{2E}] + b'. \quad (2)$$

In our case, the best values of the parameters of Eq. (1) are: $a = 0.3460$ and $b = -0.6093$ with $r^2 = 0.9974$, while the corresponding for Eq. (2) are: $a' = 0.3411$ and $b' = -0.3867$ with $r^2 = 0.9955$. The values of these parameters were obtained with the least squares method.

The effect of aqueous furfural concentration on its distribution coefficient, m , and solvent selectivity, β , is an important factor in solvent selection. The dependence of both extraction properties with the mass fraction of furfural in the extract phase (w_{2E}) at 298.15 K (band type curve) is shown in Fig. 6. As can be seen, the distribution coefficients are always greater than unity and the selectivities have sufficiently high values as to consider 1-butanol as a potential solvent for extraction purposes, particularly at room temperature at which these extraction properties have the highest values.

4. Conclusions

Equilibrium diagrams for the binary mixture (water + furfural) and for the ternary system (water + furfural + 1-butanol) as a function of temperature from 298.15 K and up to their CST were experimentally obtained. For this last system, an equilateral triangular prism was constructed in order to show the dependence of the equilibrium data with temperature, both binary UCSTs, and the position of the saddle point (situated at 376.6 ± 0.5 K with a ternary composition: $w_1 = 0.64$, $w_2 = 0.21$ and $w_3 = 0.15$). From experimental results, we can conclude that binodal band type curves are present between 298.15 K and 376.6 ± 0.5 K, but above this last value, two separate binodal curves appear. Their tie-lines were satisfactorily correlated by the Othmer and Tobias method [5] and the plait point coordinates were estimated.

A new experimental device for tie-line determination at high temperatures was developed, which is adequate for the withdrawal of samples from ternary systems in equilibrium conditions, using the well-known siphon principle.

The capability of 1-butanol as an extractive agent was evaluated from distribution coefficients and selectivities. Taking into account the high values of both extractive properties for this ternary system, 1-butanol appears to be a good solvent to extract furfural from aqueous solutions particularly at room temperature when these properties have the highest values.

5. List of symbols

a, b	parameters of Eq. (1)
a', b'	parameters of Eq. (2)
B	1-butanol (component 3)

F	furfural (component 2)
m	experimental furfural distribution coefficient ($= w_{2E}/w_{2R}$)
n_D	refractive index for sodium D line
P_{a-b}	plait points in Fig. 4
r	linear correlation coefficient of Eqs. (1) and (2)
T	temperature (K)
UCST	upper critical solution temperature
W	water (component 1)
w	mass fraction
wt. _{ij} %	percent mass fraction of component i in the phase j
Subscripts	
i	components
j	phase (E, extract phase and R, raffinate phase)
Greek	
β	selectivity ($= mw_{1R}/w_{1E}$)
ρ	density (kg m^{-3})

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