

APPLICATION OF THE CORRELATION ACCORDING TO ROD AND OF ARRANGED NRTL EQUATION FOR CALCULATION OF LIQUID EXTRACTION

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Ternary equilibrium data in the liquid-liquid system, calculated on thermodynamic basis by use of the arranged NRTL equation, are compared with the distribution coefficients of individual components and concentrations of the extracted component, obtained by the correlation according to Rod. On simple extraction examples, the effect of quality of equilibrium data on the results of chemical engineering calculation of the number of equilibrium stages at countercurrent extraction, is evaluated.

Accuracy of the design calculations of extraction units is significantly affected by the quality of the necessary equilibrium liquid-liquid data. Equilibrium composition l-l data obtained experimentally can be directly used without additional treatment only for graphical extraction calculations (*e.g.* calculation of the number of equilibrium stages, calculation of driving forces for mass transfer *etc.*), moreover only in cases when the system contains three components. In the solution of more complex problems, which is practically impossible without a computer, it is necessary to express the relation between the composition of equilibrium phases analytically. The basis for analytical expression of mutual dependence of composition of equilibrium phases are usually thermodynamic relations based on the dependence of the excess Gibbs energy of solution on composition. Calculation of composition of equilibrium phases is then performed by various numerical procedures based either on the condition of equality of activities of components in equilibrium phases or on minimization of the Gibbs energy^{1,2}. Purely empirical is the correlation proposed by Rod³ based on expression of the dependence of distribution coefficients of components on concentration of the extracted component by use of second or third degree polynomials.

In the first part of this study there is demonstrated to what extent differ the equilibrium l-l concentrations calculated on the thermodynamic basis at the use of the arranged form of the NRTL equation with the ternary correction⁴ from values calculated by use of the correlation according to Rod³. Studied are four ternary systems hydrocarbon-hydrocarbon-solvent: *a*) heptane-toluene-DMFA, $t = 40^\circ\text{C}$, *b*) heptane-cyclohexane-DMFA, $t = 25^\circ\text{C}$, *c*) cyclohexane-benzene-furfural, $t = 25^\circ\text{C}$, *d*) heptane-benzene-DMSO, $t = 40^\circ\text{C}$. The systems *a*), *c*) and *d*) have a closed binodal curve and differ by mutual solubility of the original solvent and extractant. The system *b*) has an open binodal curve *i.e.* two pairs of partially soluble

components. The experimental equilibrium concentrations of four considered systems were given in the last study⁵.

In the second part of this study an attempt is made to demonstrate on two simple examples of calculation of the number of equilibrium stages at countercurrent extraction, the effect of various accuracies of reproducibility of equilibrium data according to the arranged NRTL equation and the correlation by Rod on the result of computation.

THEORETICAL

Correlation According to Rod

The equilibrium distribution coefficient of any component K_i is defined by relation

$$x_i^{\text{II}} = K_i x_i^{\text{I}}; \quad i = 1, 2, 3, \quad (1)$$

where x_i^{II} and x_i^{I} are mole fractions both in the extractive (II) and raffinate (I) phases at equilibrium.

At constant temperature and pressure of the system the distribution coefficients depend only on composition of phases. The dependence of logarithm of distribution coefficients K_i can be expressed in the parametric form with the mole fraction of the extracted component in the extract x_2^{II} as parameter

$$\ln \hat{K}_i = \sum_k a_{ik} (x_2^{\text{II}})^k, \quad (2)$$

where $k = 0, 1 \dots n$.

When the values a_{ik} are known for individual components of the system the calculation of equilibrium concentrations for the in advance selected x_2^{II} is simple. From Eq. (2) are calculated values of distribution coefficients K_1, K_2 and K_3 . By use of Eqs (1) and conditions

$$\sum_i \hat{x}_i^{\text{I}} = 1; \quad \sum_i \hat{x}_i^{\text{II}} = 1 \quad (3)$$

result for calculation of composition of the raffinate phase the next relations

$$\hat{x}_2^{\text{I}} = \hat{x}_2^{\text{II}} / \hat{K}_2, \quad (\hat{x}_2^{\text{II}} = x_2^{\text{II}}) \quad (4a)$$

$$\hat{x}_1^{\text{I}} = [1 - \hat{K}_3 + (\hat{K}_3 - \hat{K}_2) \hat{x}_2^{\text{I}}] / (\hat{K}_1 - \hat{K}_3) \quad (4b)$$

$$\hat{x}_3^{\text{I}} = [1 - \hat{K}_2 + (\hat{K}_2 - \hat{K}_1) \hat{x}_1^{\text{I}}] / (\hat{K}_3 - \hat{K}_2). \quad (4c)$$

The mole fractions of components of the extract phase x_2^{II} are calculated by use of Eq. (1).

For calculation of coefficients a_{ik} in Eq. (2) has been used the objective function in the form

$$F(a_{ik}) = \sum_i \sum_l w_{il} \left(\frac{K_{il} - \hat{K}_{il}}{K_{il}} \right)^2 \quad (5)$$

for $i = 1, 2, 3$; $l = 1, 2, \dots, m$. For all i and l has been considered the statistical weight $w_{il} = 1$.

The objective function (5) has been minimized by the Newton-Raphson method for which has been obtained by the usual procedure the system of equations

$$\sum_i \sum_k A_{iks} \delta a_{ik} = B_{is}, \quad (6)$$

where

$$A_{iks} = \sum_l \left(\frac{1}{K_{il}} \right)^2 \left(\frac{\partial \hat{K}_{il}}{\partial a_{ik}} \right)_{a_{k \neq s}} \left(\frac{\partial \hat{K}_{il}}{\partial a_{is}} \right)_{a_{k \neq s}} \quad (7)$$

$$B_{is} = \sum_l \left(\frac{1}{K_{il}} \right)^2 (K_{il} - \hat{K}_{il}) \left(\frac{\partial \hat{K}_{il}}{\partial a_{is}} \right)_{a_{k \neq s}} \quad (8)$$

$$\delta a_{ik} = a_{ik} - a_{ik0}. \quad (9)$$

The systems of linear equations (6) are for each component i mutually independent and each of them is solved independently. The starting values of coefficients in polynomials (2) — a_{ik0} are calculated by the method of linear regression by use of the logarithmic form of the objective function (5)

$$F'(a_{ik}) = \sum_i \sum_l (\ln K_{il} - \ln \hat{K}_{il})^2,$$

In Table I are given values of a_{ik} calculated for four studied systems. The numbers of coefficients in the polynomial (2) were for individual components different. For the component 2 has been used the equation with four coefficients while for the components 1 and 3 the relation (2) has been used with three or four a_{ik} coefficients.

Calculated equilibrium concentrations were evaluated on basis of residuals of mole fractions

$$F_x = 100 \sqrt{[\sum_i \sum_j (x_{ij1} - \hat{x}_{ij1})^2 / 6m]} \quad (10)$$

$$l = 1, 2, \dots, m; \quad j = \text{I, II}; \quad i = 1, 2, 3$$

and also by relative residuals of distribution coefficients of the extracted component

$$F_{K_2} = 100 \sqrt{\left[\sum_1 \left(\frac{K_{21} - \hat{K}_{21}}{K_{21}} \right)^2 \frac{1}{m} \right]}. \quad (11)$$

The values F_x and F_{K_2} are given in Table III. The results are evaluated in the following part of this study.

Calculation by Use of the Arranged NRTL Equation

Improved prediction of ternary l-l equilibrium data, by introduction of the universal correction into the NRTL equation and also methods of adjustment of binary

TABLE I
Coefficients a_{ik} in Eq. (2)

System	t °C	i	a_{i0}	a_{i1}	a_{i2}	a_{i3}
a) heptane toluene DMFA ^a	40	1	-2.032	7.162	-14.08	263.0
			-2.001	4.268	39.38	—
		2	-0.4176	6.377	-67.78	295.4
			2.048	-8.064	9.803	-215.0
3	2.002	-6.762	-21.38	—		
	b) heptane cyclohexane DMFA	1	-2.399	3.544	-2.826	5.431
-2.398			3.455	-1.445	—	
2		-1.882	-0.4090	20.93	-69.66	
		2.535	-1.493	-5.672	33.61	
3	2.543	-2.160	3.941	—		
	c) cyclohexane benzene furfural	1	-1.932	5.831	-17.78	103.7
-1.932			3.599	13.24	—	
2		-0.5862	8.991	-65.58	173.8	
		2.807	-10.78	38.95	-173.9	
3	2.770	-8.219	-3.217	—		
	d) heptane benzene DMSO ^b	1	-4.267	6.028	0.4053	6.862
-4.262			5.697	3.436	—	
2		-0.7652	1.920	-1.835	-2.419	
		4.896	-8.067	11.22	-47.06	
3	4.867	-5.807	-9.654	—		

^a DMFA Dimethylformamide, ^b DMSO dimethyl sulfoxide.

and ternary parameters and of their determination have been considered in recent studies^{4,5}. Similarly were discussed methods of reverse calculation of equilibrium composition.

In the calculation of activity coefficient of an arbitrary component in the ternary solution, the relation has been used

$$\ln \gamma_i = \ln \gamma_{ib} + A_i \ln \gamma_i \quad (12)$$

in which the first right-hand side term is given by the classical form of the NRTL equation with binary parameters τ_{ij} and α_{ij} . The second term of this equation – the ternary correction is related to ternary interactions and the relation has been derived for it⁵

$$A_i \ln \gamma_i = x_2 x_3 [E_1 x_1 (2 - 3x_1) + E_2 x_2 (1 - 3x_1) + E_3 x_3 (1 - 3x_1)]. \quad (13)$$

The ternary correction (13) is written for the component 1. For the remaining components Eq. (13) is arranged by cyclical exchange of indices.

TABLE II
Binary and ternary parameters of the arranged NRTL equation for studied systems

Parameter	Values of parameters for system			
	a) heptane(2)- toluene(2)- DMFA(3) <i>t</i> = 40°C	b) heptane(1)- cyclohexane(2)- DMFA(3) <i>t</i> = 25°C	c) cyclohexane(1)- benzene(2)- furfural(3) <i>t</i> = 25°C	d) heptane(1)- benzene(2)- DMSO(3) <i>t</i> = 40°C
τ_{12}	0.0878	-0.8646	0.0031	-0.4936
τ_{21}	0.3614	1.0198	0.5589	1.1863
α_{12}	0.4097	0.0664	0.2341	0.2499
τ_{13}	1.7395	2.2541	2.6582	4.1180
τ_{31}	1.7190	2.1387	0.7223	3.6368
α_{13}	0.2850	0.3560	0.1600	0.3500
τ_{23}	1.1255	2.3153	1.1164	1.2671
τ_{32}	-0.1286	1.5494	0.4213	0.5790
α_{23}	0.4001	0.3660	0.8698	0.6422
E_1	-1.4341	0.0705	-2.7785	3.7388
E_2	-3.9436	-0.2070	-3.4147	-3.4221
E_3	-0.8059	0.0524	-0.8602	3.3155

For calculation of equilibrium concentrations for four studied systems the values of parameters given in Table II have been used. The accuracy of reproducibility of equilibrium l-l data determined by the isoactivity method has been evaluated on basis of residuals of mole fractions according to Eq. (10) and also of residuals of distribution coefficients K_2 according to Eq. (11). The values F_x and F_{K_2} are given in Table III. All the given data are taken from papers^{4,5}.

It can be seen from data of Table III, that the equilibrium l-l mole fractions calculated on the thermodynamic basis by use of the NRTL equation, have the standard deviation from the experimental data of about $2 \cdot 10^{-3}$ so that the calculation might be considered as very accurate. Nevertheless, by use of the correlation according to Rod the values F_x and F_{K_2} have been obtained, which are up by one order of magnitude, in average about five times smaller especially at the use of the third degree polynomials for all three components. This depicts high flexibility of the equations proposed by Rod.

In this connection it is very interesting to study whether the results of computation of the extractor can be significantly affected by a higher accuracy of equilibrium data by use of the equations according to Rod, with regard to the reached accuracy by use of the NRTL equation. An attempt has been made to give an answer to this

TABLE III

Comparison of reproducibility of equilibrium data l-l by use of residuals F_x and F_{K_2}

System	Arranged NRTL Eq.		Correlation according to Rod	
	F_x	F_{K_2}	F_x	F_{K_2}
a) heptane-toluene-DMFA, $t = 40^\circ\text{C}$	0.253	5.64	0.0531 ^a 0.193 ^b	0.981
b) heptane-cyclohexane-DMFA, $t = 25^\circ\text{C}$	0.155	2.62	0.0923 ^a 0.0935 ^b	0.635
c) cyclohexane-benzene-furfural, $t = 25^\circ\text{C}$	0.146	6.66	0.0254 ^a 0.153 ^b	0.198
d) heptane-benzene-DMSO, $t = 40^\circ\text{C}$	0.139	2.59	0.0866 ^a 0.0942 ^b	1.17

^a Polynomial(2) of the third degree was used for all three compounds of the system, ^b for component 2 has been used the polynomial(2) of the third degree and for components 1 and 3 polynomial of the second degree.

question in the following part of this study on basis of two examples of calculations of equilibrium stages of the countercurrent extractor.

Basic Relations in Calculation of the Number of Equilibrium Stages in Countercurrent Extraction

A simple countercurrent extraction operation is demonstrated in Fig. 1. Graphical procedure of calculation of equilibrium stages for a system with three components is so familiar, that it is not necessary to discuss it in detail. In the analytical solution of the problem the composition of material streams from stage to stage is determined numerically, while calculation of equilibrium phases of the extract and raffinate is iterative.

Basic relations for calculation of streams leaving the system are equations of the overall material balance and component balances, for which agreement with Fig. 1 holds

$$R_0 + E_{n+1} = R_n + E_1 \quad (14)$$

$$R_0 x_{i,0} + E_{n+1} y_{i,n+1} = R_n x_{i,n} + E_1 y_{i,1} ; \quad i = 1, 2, 3 . \quad (15)$$

Eqs (14) and (15) can be formally written

$$R_0 + E_{n+1} = S \quad (16a)$$

$$R_n + E_1 = S \quad (16b)$$

$$R_0 x_{i,0} + E_{n+1} y_{i,n+1} = S x_{i,S} \quad (17a)$$

$$R_n x_{i,n} + E_1 y_{i,1} = S x_{i,S} \quad (17b)$$

while for each material stream hold the conditions

$$\sum_i x_i = 1 ; \quad \sum_i y_i = 1 . \quad (18)$$

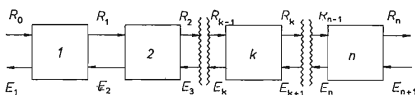


FIG. 1

Countercurrent multistage extraction

When S in Eq. (17b) is expressed by use of Eq. (16b) and so arranged equation is written for components $i = 2$ and $i = 3$, by their combination the relation is obtained

$$\frac{y_{3,1} - x_{3,S}}{y_{2,1} - x_{2,S}} = \frac{x_{3,S} - x_{3,n}}{x_{2,S} - x_{2,n}} \quad (19)$$

It is suitable to denote the right-hand side of Eq. (19) as l_1 , i.e.

$$l_1 = (x_{3,S} - x_{3,n}) / (x_{2,S} - x_{2,n}) \quad (20)$$

and $x_{3,1}$ to express from condition (18) so that the arranged form of Eq. (19) is obtained

$$y_{2,1} = (1 + l_1 x_{2,S} - x_{3,S} - y_{1,1}) / (1 + l_1) \quad (21)$$

Eq. (21) is used in the iteration calculation of composition of the extract stream E_1 , which is explained in the following part.

Another arrangement of Eqs (14) and (15) leads to the definition of the so-called fictive material stream Z , for which there holds

$$R_0 - E_1 = R_n - E_{n+1} = Z \quad (22)$$

$$R_0 x_{i,0} - E_1 y_{i,1} = R_n x_{i,n} - E_{n+1} y_{i,n+1} = Z x_{i,Z} \quad (23)$$

By use of the fictive stream Z it is possible to express the relation among the raffinate leaving any stage with the extract which enters the same stage. From the material balances of individual sections of the system results

$$R_0 - E_1 = R_1 - E_2 = \dots = R_k - R_{k+1} = \dots = R_n - E_{n+1} = Z \quad (24)$$

On basis of balance of the first up to a general stage k , it is possible in a similar way as in derivation of Eqs (19) or (21) to derive, for calculation of composition of the extract stream E_{k+1} , the relations

$$\frac{y_{3,k+1} - x_{3,Z}}{y_{2,k+1} - x_{2,Z}} = \frac{x_{3,k} - x_{3,Z}}{x_{2,k} - x_{2,Z}} \quad (25)$$

or after introduction

$$l_2 = (x_{3,k} - x_{3,Z}) / (x_{2,k} - x_{2,Z}) \quad (26)$$

the relation is obtained

$$y_{2,k+1} = \frac{1 + I_2 x_{2,Z} - x_{3,Z} - y_{1,k+1}}{1 + I_2} \quad (27)$$

CALCULATIONS

Algorithm of Calculation

A short calculation algorithm of the number of equilibrium stages is described, when the correlation equilibrium composition according to Rod is applied. At the use of the arranged NRTL equation the algorithm differs only in the method of calculation of equilibrium, by which one of the selectable mole fractions (usually $x_2 \equiv x_2^I$ or $y_2 \equiv y_2^{II}$) can be chosen according to situation. The information on the amount and composition of the treated material R_0 and of the solvent E_{n+1} are known and from the fixed requirement on the separation degree is known the concentration $x_{2,n}$.

1) Composition of the raffinate product R_n is calculated. For the selected value y_2 and by use of relations (2) and (4) the raffinate composition is calculated. When x_2 of this raffinate is not equal to the required $x_{2,n}$ the calculation is repeated. In each next iteration is used the value y_2 which resulted from the equation $y_2 = K_2 x_2$, when for x_2 and K_2 is substituted the value from the last iteration.

2) By use of Eqs (16a) and (17a) are calculated the amount and composition of the mixture S and from Eq. (20) the value I_1 . The equilibrium composition of two liquid phases is calculated by use of equations of the Rod correlation for the chosen value $y_{2,1}$. According to concentrations of the extract phase is checked whether the condition given by Eq. (19) is satisfied. When the value of the left-hand side of Eq. (19) is not equal to I_1 , the calculation must be repeated. In each next iteration the value $y_{2,1}$ is used, calculated according to Eq. (21) in which for $y_{1,1}$ is substituted the value from the last iteration. After completion of the iteration calculation, composition of the extract stream E_1 and composition of the raffinate stream R_1 are known.

3) Calculation proceeds by determination of flow rates of streams leaving the system E_1 and R_n by use of relations (16b) and (17b). By solution of Eqs (22) and (23) the flow rates and compositions of fictive streams Z are calculated.

4) Composition of the streams leaving the second or in general the k -th stage is calculated. First of all I_2 is calculated from Eq. (26) and for the chosen value y_2 the composition of equilibrium phases is calculated. When the so calculated composition E_2 or E_{k+1} does not satisfy Eq. (25) the calculation is repeated. In the iteration procedure y_2 is chosen according to relation (27) while for y_1 is substituted in the first iteration the value calculated in the preceding stage and in each following the value y_1 from the preceding iteration.

5) The calculation is returned to the step 4. Calculation of the number of stages is terminated, when x_2 of the raffinate stream has the value equal or smaller than $x_{2,n}$.

Example of Calculation

According to the described algorithm, the multisage countercurrent extraction has been calculated for one ternary system with the closed and one system with the open binodal curve.

Example 1. System heptane(1)–benzene(2)–dimethyl sulfoxid (DMSO) (3), $t = 40^\circ\text{C}$. Extraction process conditions: the feed is equimolar mixture of components (1) and (2). The used solvent is pure component 3. To 1 mol of feed R_0 corresponds 1.2 mol of solvent E_{n+1} . Degree

of extraction is fixed by the mole fraction of the extracted component in the final raffinate, $x_{2,n} = 0.01$.

Example 2. System heptane(1)-cyclohexane(2)-dimethylformamide (DMFA) (3), $t = 25^\circ\text{C}$. Extraction process conditions: composition of the feed is given by $x_{1,0} = x_{2,0} = 0.5$. The solvent does not contain components 1 and 2. Relatively high consumption of the solvent $E_{n+1}/R_0 = 6.40$. Extraction degree is determined by the value $x_{2,n} = 0.01$.

RESULTS AND DISCUSSION

Results of the calculated number of equilibrium stages for both examples are given in Table IV. They are classified according to the used methods of calculation of the equilibrium composition. The correlation according to Rod has been used in two modifications, when in the case *a*) the polynomial by Eq. (2) has been of the third degree for each component, while in the case *b*) for the components (1) and (3) the second degree polynomial has been used. Beside the analytical calculation of the number of stages is in Table IV also given the number of stages determined graphically. In the graphical calculation experimental equilibrium data have been used.

Relation between the calculated number of stages in Table IV and quality of equilibrium dependences given by the magnitude of residuals in Table III is obvious. Small differences in residuals F_x at both modifications of the correlation according to Rod have not appeared in the total number of equilibrium stages. It can be seen, that usually suffices when the concentration dependence of distribution coefficients of components 1 and 3 is expressed by the second degree polynomial. For about 10% lower number of stages has been calculated by the modified NRTL equation which has been caused by bigger differences between the calculated and measured K_2 values.

The 10% differences in the calculation of the number of stages can be considered as adequate *i.e.* not too significant. In agreement with usances and possibilities

TABLE IV

Calculated number of equilibrium stages in the countercurrent extraction

Example	Calculated number of stages			
	graphical	Rod correlation		NRTL equation
		<i>a</i>)	<i>b</i>)	
1 (system <i>d</i>)	11.8	11.4	11.4	10.6
2 (system <i>b</i>)	12.7	12.5	12.5	11.6

in the chemical engineering calculations the limit of calculation accuracy of the number of stages is fixed as $\pm 20\%$. On the basis of results of demonstrated examples and data given in Table III and some linear extrapolation it is obvious that the values F_x at this accuracy should be equal to 0.3, values F_{K_2} to about 5. But these are requirements too severe for the thermodynamic calculation and *e.g.* for numerous systems which are given in the study of Sørensen and coworkers² these requirements are not met.

Though it is not possible to make any far-reaching conclusions on the basis of a small number of examples, it is obvious that in the extractor calculations it is necessary to pay considerable attention both to the equilibrium I-I data and accuracy of their thermodynamic treatment. A very good reproducibility of equilibrium data at a high speed of calculation is reached by use of the correlation according to Rod. It is true that the used equations are purely empirical and of single use, while thermodynamic relations can be used also in calculation of the rectification part of the separation process.

LIST OF SYMBOLS

a_{ik}	coefficient in Eq. (2)
E_1, E_2, E_3	ternary parameters of the arranged NRTL equation
E_1	mole flow rate of extract from the 1st stage
E_{k+1}	mole flow rate of extract from the stage $k + 1$
E_{n+1}	mole flow rate of solvent entering the system
$F(a_{ik})$	objective function
F_x, F_{K_2}	residual of equilibrium fractions or distribution coefficients for the component 2
K_i	experimental equilibrium distribution coefficient
\hat{K}_i, K_i	calculated equilibrium distribution coefficient
l_1, l_2	quantities given by Eqs (20) or (26)
m	number of experimental conodes
n	degree of polynomial
R_0	mole flow rate of feed
R_k	mole flow rate of raffinate from the k th stage
R_n	mole flow rate of raffinate from the last stage
S	mole flow rate of mixture given by Eq. (16)
x_i	mole fraction of component i in the raffinate stream
x_i^I, x_i^{II}	experimental equilibrium mole fraction of component i in the raffinate or extract phase
$\hat{x}_i^I, \hat{x}_i^{II}$	calculated equilibrium mole fraction
y_i	mole fraction of component in the extract stream
Z	mole flow rate of fictive mixture given by Eq. (22)
α_{ij}	binary parameter NRTL equation in the system of components $i-j$
γ_i	activity coefficient of component in the ternary solution
γ_{ib}	activity coefficient of component of ternary solution determined by use of binary parameters
Δ_i	ternary correction
τ_{ij}	binary parameter of NRTL equation in the system of components $i-j$

Subscripts

i	arbitrary component of the system
j	phase
k	ordinal number of coefficient in Eq. (2)
l	ordinal number of experimental point
1, 2, 3	symbols used for components of the ternary system
1, 2, ..., n	symbols used for stages in countercurrent extraction

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