EXTRACTION OF D-TARTARIC ACID WITH ALAMINE 336/LIX 54 MIXTURE

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Extraction equilibria in the system aqueous solution of D-tartaric acid–solution of tertiary amine in diluent at 25 °C have been studied. The amine was Alamine 336, a long-chain tertiary amine with C_8–C_{10} alkyls in 2 : 1 ratio; the diluent was chelating extractant LIX 54, a solution of an alkyl β-diketone in kerosene. The experimental results were correlated using a mathematical model which takes into account chemical reactions involved in acid–amine complex formation and non-specific effects of the nonideality of the system.

Key words: Extractions; Extraction equilibrium; Carboxylic acids; Tertiary amines; Mixed extractants.

Reactive extraction of various carboxylic and hydroxycarboxylic acids from aqueous solutions with tertiary amines has been extensively investigated. As an alternative to classic refining method, solvent extraction with Alamine 336 has been proposed for separation of citric acid from the fermentation broth[1]. Extensive studies on the distribution of propionic, lactic, pyruvic, succinic, fumaric, maleic, malic, itaconic, tartaric, citric, and isocitric acids, products of aerobic fermentation of glucose, in various solvents and extractants were published by Kertes and King[2]. The influence of various diluents on the extraction of acetic, lactic, succinic, malonic, fumaric, and malic acids with commercial extractant Alamine 336 was studied by Tamada *et al.*[3]. Formation of various complexes in the organic phase as well as the effect of temperature and the water coextraction were discussed in other papers of these authors[4,5]. Yang *et al.*[6] studied the influence of pH, amine and diluent concentrations on the extraction of lactic, acetic, propionic, and butyric acids with tertiary amine Alamine 336 and quaternary ammonium salt Aliquat 336.

The desired product in manufacturing of carboxylic acids is free acid and, therefore, stripping into pure water should be the final step of the extraction method applied. Two main conceptions have been proposed[5] for adjusting equilibrium distribution to favour the organic phase in the extraction and aqueous phase in the stripping steps. These are the temperature swing and the diluent swing. The first is based on enthalpy effects in transitions of solute between the phases and can be realized by increasing temperature in the stripping step. The other swing is based on synergistic effects of some diluents resulting in an increase in the extracting power of amine. Thus, when a binary diluent (inert hydrocarbon and active modifier) is used, this conception can be realized by changing the solvent composition.

In the extraction of citric acid with trialkylamine (TAA), the influence of single-component diluents[7] and the effects of temperature and solvent composition of a binary diluent[8] were investigated. In another study extraction of lactic, malic, and citric acids with TAA in the same diluent was compared[9]. Recently, the amine extraction equilibrium in systems D-tartaric acid–solution of TAA in binary diluents (octan-1-ol–heptane, isodecanol–kerosene, and methyl isobutyl ketone (MIBK)–heptane) has been investigated[10].

LIX 54 (Henkel Co.), a mixture of a β-ketone in kerosene, is widely used as chelating extractant for various metals. In the present work it has bee applied as a binary diluent of the amine extractant; the β-diketone acts as a ketonic modifier. As a tertiary amine, Alamine 336 was used. The results of equilibrium measurements at 25 °C were compared with those obtained earlier with other binary diluents[10]. Similar models for data correlation and simulation were used.

THEORETICAL

As in previous studies[9–11], the following simplifying assumptions were made:

1. The dissociation in aqueous phase was neglected.

2. Specific bonding of coextracted water molecules to acid–amine complexes was not taken into account; the measured water content in the organic phase was only used to calculate the composition of organic phase on water-free basis.

3. Physical extraction of acid with the organic phase and the solubility of solvent components in the aqueous phase were neglected.

4. In the aqueous phase the acid activities were assumed equal to their concentrations. Nonideality of the organic phase was taken into account by introducing an empirical function of the acid and amine molality.

The applied model[10] consists of two parts. The chemical part of this model takes into account expressions for total acid and amine concentrations in the organic phase and the equilibrium constants of formation of the acid–amine complexes considered:

$$\overline{m_a} = \sum_{k=1}^{K} i_k \left(\overline{m_{ij}}\right)_k \tag{1}$$

$$\overline{m_e^0} = \overline{m_e} + \sum_{k=1}^{K} j_k \left(\overline{m_{ij}}\right)_k \tag{2}$$

$$\beta_{ij} = \overline{m_{ij}} \,\overline{\gamma_{ij}} \Big/ \overline{m_a}^i \,\overline{m_e}^j \,\overline{\gamma_e}^j = \beta_{ij}^I \,\overline{\gamma_{ij}} \Big/ \overline{\gamma_e}^j \ . \tag{3}$$

Combining Eqs (*1*) and (*2*) with Eq. (*3*) we obtain:

$$\overline{m_a} = \sum_{k=1}^{K} i_k (\beta_{ij}^I m_a^i \,\overline{m_e}^j)_k \tag{4}$$

$$\overline{m_e^0} = \overline{m_e} + \sum_{k=1}^{K} j_k (\beta_{ij}^I m_a^i \,\overline{m_e}^j)_k \ . \tag{5}$$

In these expressions m is the molality, subscripts "a" and "e" refer to acid and amine, respectively, and the bar denotes the organic phase; $\overline{m_e^0}$ is the total molality of amine, $(\overline{m_{ij}})_k$ is the molality of the k-th complex, and K is the number of complexes considered. The molalities are related to water and the binary diluent, respectively. In Eqs (*1*)–(*5*) i_k, j_k are the numbers of acid and amine molecules in the k-th complex; i, j are the numbers of acid and amine molecules in all types of acid–amine complexes considered. In Eq. (*3*), β_{ij} and β_{ij}^I are the thermodynamic and conditioned equilibrium constants of formation of complex with i molecules of acid and j molecules of amine; $\overline{\gamma_{ij}}$ and $\overline{\gamma_e}$ are the activity coefficients of acid and amine in the organic phase.

In the second physical part of the model, non-specific interactions of the species in the organic phase are taken into account. The ratio of activity co-

efficients in Eq. (*3*) is expressed by an exponential function of the total amine and acid molalities and of the mass fraction of modifier in the diluent, $\overline{m_{\mathrm{e}}^0}$, $\overline{m_{\mathrm{a}}}$, and $\overline{x}$, respectively. Thus, the conditioned equilibrium constant β_{ij}^{I} can be expressed as:

$$\beta_{ij}^{\mathrm{I}} = \beta_{ij} \exp\left(A_{ij} \overline{m_{\mathrm{e}}^0} + B_{ij} \overline{x} + C_{ij} \overline{m_{\mathrm{a}}} \right) . \tag{6}$$

Here β_{ij}, A_{ij}, B_{ij}, and C_{ij} are the model parameters. Accordingly, the number of model parameters is 12. Since LIX 54 was used as supplied the fraction of modifier in the binary diluent, $\overline{x}$, was not varied and the influence of diluent composition onto the extraction of D-tartaric acid cannot be followed. Arbitrary values were ascribed to the parameters B_{ij} and the terms were included into the thermodynamic equilibrium constants β_{ij} (Table II). Thus, in the present work the number of model parameters is 9.

For correlation of data and evaluation of parameters, the optimization algorithm and procedure were the same as described earlier[9–11]. The weighed sum of squares of relative deviations between calculated and experimental values of acid molality in the organic phase was used as objective function:

$$S = \sum_{q=1}^{N} W_q \left[(1 - m_{\mathrm{a}_{\mathrm{calc}}} / m_{\mathrm{a}_{\mathrm{exp}}})^2 + \left(1 - \overline{m}_{\mathrm{a}_{\mathrm{calc}}} / \overline{m}_{\mathrm{a}_{\mathrm{exp}}} \right)^2 \right] N / \sum_{q=1}^{N} W_q . \tag{7}$$

The weights are used in the optimization process both for reducing the influence of individual points suspect of large experimental error and for an interactive way of finding global optimum. Since the optimization algorithm involves an internal iteration procedure for optimizing the value of independent variable of each experimental point separately, a common weight for both co-ordinates was used in Eq. (*7*). Here N is the number of experimental points and W_q are the weights ascribed to the individual points.

EXPERIMENTAL

Alamine 336 (Henkel Co.) is mixture of long-chain tertiary amines with C_8–C_{10} alkyl groups (the C_8 chains predominating in a molar ratio of 2 : 1) and with the average molecular mass equal to 393 g/mol. LIX 54 (Henkel Co.), a 35% mixture of a β-diketone in a low-aromatic kerosene[12] was used as binary diluent. On the basis of the given[12] structural formulae its determined by us average molecular mass was 244 g/mol.

D-Tartaric acid (Lachema Co.) and other reagents used were of analytical grade purity.

The desired ratios of Alamine 336 in LIX54 were prepared by weighing. Before use, the mixtures were treated successively with aqueous solutions of hydrochloric acid, sodium hydroxide, and several times with distilled water.

The phases were contacted in separatory funnels in a thermostatted bath shaker at 25 °C for 1 h.

Both organic and aqueous phases were analyzed. The concentration of tartaric acid was determined by potentiometric titration with sodium methanolate solution in methanol–dimethylformamide 3 : 1 mixture and in water, for organic and aqueous phases, respectively. The amine in the organic phase was titrated with perchloric acid in anhydrous acetic acid. The concentration of water in the organic phase was determined using the Karl–Fischer method. The concentrations of both the amine and the acid in the organic phase were expressed in mol/kg of binary diluent. The maximum overall relative error of the measured concentrations was less than 5%.

RESULTS AND DISCUSSION

Experimental Results

The measured equilibrium concentrations of amine in the diluent and concentrations of acid in aqueous and organic phases are presented in Table I. These results are also depicted as points in Figs 1 and 2. In these figures the content of acid in the organic phase is expressed as loading of extractant $Z = \overline{m_a} / \overline{m_e^0}$.

Similarly as for the systems examined in the previous work[10], the present system shows overloading of amine at high acid concentration in the aqueous phase. Thus maximal value $Z = 1.2$ mol of acid/mol of amine was obtained at $\overline{m_e^0} \div 1.0$ mol amine/kg diluent; $m_a = 6.16$ mol acid/kg water and

Fig. 1

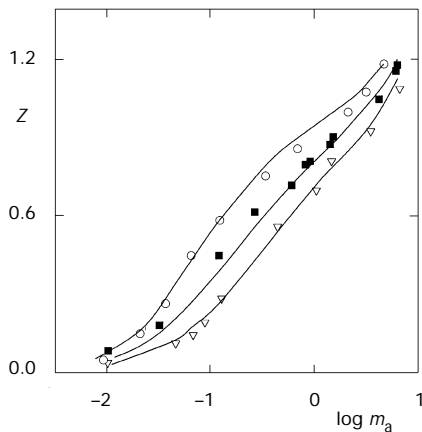Extractant loading Z in dependence on acid concentration in the aqueous phase m_a (in mol of acid/kg of water), $\overline{x} = 0.35$ kg modifier/kg binary diluent (model variant I). Points: experimental data; lines: calculated data. ∇, $\overline{m_e^0} = 0.3$ mol amine/kg diluent; ■, $\overline{m_e^0} = 1.0$ mol amine/kg diluent; ○, $\overline{m_e^0} = 2.0$ mol amine/kg diluent

TABLE I
Equilibrium data for the system aqueous solution of D-tartaric acid–solution of Alamine 336 in LIX 54, $\overline{x}$ = 0.35 kg modifier/kg diluent; 25 °C, N = 32

$\overline{m}_e^0$, mol amine/kg diluent	m_a, mol acid/kg water	$\overline{m}_a$, mol acid/kg diluent
0.294	0.010	0.013
0.292	0.046	0.034
0.292	0.068	0.044
0.293	0.088	0.058
0.291	0.147	0.084
0.291	0.439	0.163
0.290	1.035	0.203
0.291	1.447	0.236
0.288	3.434	0.267
0.281	6.507	0.306
1.050	0.010	0.094
1.050	0.032	0.195
1.023	0.120	0.463
1.051	0.265	0.649
1.017	0.599	0.733
1.050	0.813	0.840
1.043	0.900	0.847
1.040	1.401	0.912
1.040	1.499	0.943
1.053	4.122	1.107
1.050	5.978	1.216
1.014	6.164	1.197
1.963	0.009	0.105
1.973	0.021	0.304
1.989	0.037	0.536
1.973	0.065	0.892
1.994	0.123	1.168
1.970	0.336	1.489
1.955	0.681	1.684
1.947	2.079	1.947
1.938	3.106	2.088
1.946	4.595	2.304

$\overline{m_e^0} \div 2.0$ mol amine/kg diluent; m_a = 4.59 mol acid/kg water. This degree of overloading is similar to that reached[10] in systems with alcoholic modifiers (octan-1-ol and isodecanol) and in the systems with MIBK, at $\overline{x}$ = 0.4 kg of modifier/kg of binary diluent. In the latter case, however, this seems to have been caused by the upper limit of aqueous acid concentration, at which third phase appears since at $\overline{x}$ = 0.7 and 1.0 kg of modifier/kg of binary diluent $Z \geq 2$ mol of acid/mol of amine has been obtained. Hence, whereas in the system with MIBK it was necessary to include formation of the 3 : 1 complex in the model, in the present system formation of the 2 : 1 and/or 3 : 1 complexes can be assumed.

According to Tamada *et al.*[3] and Bízek *et al.*[8], if a complex with more than one amine molecule is present in the organic phase, Z should increase with increasing amine concentration. A marked growth of Z with growing $\overline{m_e^0}$ can be detected in the range $-1.3 < \log m_a < 0$, when comparing the curves in Figs 1 or 2. This supports assuming predominant formation of the 1 : 2 complex in this region. At medium acid concentrations, formation of the 1 : 1 complex should be the dominant reaction.

In the present experiments, no third phase formation was observed, even at the highest acid and amine concentrations in the organic phase. Apparently, the fraction of the β-diketone in LIX 54, $\overline{x}$ = 0.35 kg of modifier/kg of binary diluent, is high enough to prevent this. When comparing the acid distribution in the present system at low and medium concentrations in aqueous phase with that in the system with MIBK (ref.[10]) at $\overline{x}$ = 0.4 kg of modifier/kg of binary diluent, about equal distributions with both ketone modifiers can be observed.
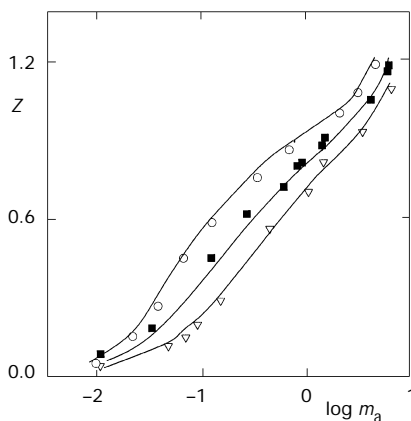

FIG. 2
Extractant loading Z in dependence on acid concentration in the aqueous phase m_a (in mol acid/kg water), $\overline{x}$ = 0.35 kg modifier/kg binary diluent (model variant II). Points: experimental data; lines: calculated data. ▽, $\overline{m_e^0}$ = 0.3 mol amine/kg diluent; ■, $\overline{m_e^0}$ = 1.0 mol amine/kg diluent; ○, $\overline{m_e^0}$ = 2.0 mol amine/kg diluent

Data Correlation and Simulation

In accord with the above reasoning, a mathematical description of the present system should contain formation of the 1 : 1 and 1 : 2 complexes as processes governing the low- and medium-concentration ranges, and formation of the 2 : 1 or 3 : 1 complexes responsible for overloading. As the algorithm used allows to include only three parallel chemical reactions, two model variants have been used for correlation of data. Variant I includes complexes 1 : 1, 1 : 2, and 2 : 1 (i = 1; 1; 2 and j = 1; 2; 1), variant II complexes 1 : 1, 1 : 2, and 3 : 1 (i = 1; 1; 3 and j = 1; 2; 1). The particular forms of Eqs (*3*)–(*5*) can be rewritten as:

Variant I:

$$\overline{m_{a}} = \beta_{11}^{I} m_{a} \overline{m_{e}} + \beta_{12}^{I} m_{a} \overline{m_{e}}^{2} + 2 \beta_{21}^{I} m_{a}^{2} \overline{m_{e}} \tag{8}$$

$$\overline{m_{e}^{0}} = \overline{m_{e}} + \beta_{11}^{I} m_{a} \overline{m_{e}} + 2 \beta_{12}^{I} m_{a} \overline{m_{e}}^{2} + \beta_{21}^{I} m_{a}^{2} \overline{m_{e}} \tag{9}$$

Variant II:

$$\overline{m_{a}} = \beta_{11}^{I} m_{a} \overline{m_{e}} + \beta_{12}^{I} m_{a} \overline{m_{e}}^{2} + 3 \beta_{31}^{I} m_{a}^{3} \overline{m_{e}} \tag{10}$$

$$\overline{m_{e}^{0}} = \overline{m_{e}} + \beta_{11}^{I} m_{a} \overline{m_{e}} + 2 \beta_{12}^{I} m_{a} \overline{m_{e}}^{2} + \beta_{31}^{I} m_{a}^{3} \overline{m_{e}} \tag{11}$$

and

$$\beta_{11}^{I} = \overline{m_{11}} / m_{a} \overline{m_{e}} ; \quad \beta_{12}^{I} = \overline{m_{12}} / m_{a} \overline{m_{e}}^{2} ;$$
$$\beta_{21}^{I} = \overline{m_{21}} / m_{a}^{2} \overline{m_{e}} ; \quad \beta_{31}^{I} = \overline{m_{31}} / m_{a}^{3} \overline{m_{e}} . \tag{12}$$

The optimized values of parameters for the two model variants are listed in Table II and corresponding simulation curves are presented in Figs 1 and 2. The relative standard deviation of measured and calculated data, s^{r}, was used as a measure of fit of the correlation:

$$s^r = \left\{ \frac{1}{2N-n} \sum_{q=1}^{N} \left[\left(1 - m_{a_{calc}} / m_{a_{exp}} \right)^2 + \left(1 - \overline{m}_{a_{calc}} / \overline{m}_{a_{exp}} \right)^2 \right]_q \right\}^{1/2}. \quad (13)$$

Here $n = 9$ is the number of parameters evaluated.

No significant improvement of the standard deviation was obtained with model variant II. However, a comparison of Figs 1 and 2 shows a somewhat better fit of model variant II at medium and high acid concentrations, mainly for the series $m_e^0 = 0.3$ mol amine/kg diluent and 2.0 mol amine/kg diluent. This confirms the importance of including the formation of the 3 : 1 complex in the organic phase.

It is interesting to note that for both model variants, the values of A_{11}, expressing the non-specific effects of amine on loading, are negative. This phenomenon can be explained assuming that non-specific effects may partly compensate the positive specific effect of the 1 : 1 complex formation on overall loading.

Figures 3 and 4 show the contributions of partial extractant loadings of individual acid–amine complexes, Z_{ij}, to the overall extractant loading Z. These curves have been simulated using the model variant II and respective

TABLE II
Parameter values of model variants I and II

$i : j$	β_{ij}, (kg/mol)$^{i+j-1}$	A_{ij}, kg diluent/mol amine	C_{ij}, kg water/mol acid
	Model variant I		
	$N = 32$; $n = 9$; $s^r = 8.524 \cdot 10^{-2}$		
1 : 1	$2.124 \cdot 10^0$	$-2.393 \cdot 10^{-2}$	$1.759 \cdot 10^0$
1 : 2	$5.283 \cdot 10^{-3}$	$2.687 \cdot 10^0$	$-2.167 \cdot 10^{-1}$
2 : 1	$5.073 \cdot 10^0$	$-3.426 \cdot 10^{-1}$	$2.068 \cdot 10^0$
	Model variant II		
	$N = 32$; $n = 9$; $s^r = 8.485 \cdot 10^{-2}$		
1 : 1	$2.139 \cdot 10^0$	$-3.970 \cdot 10^{-2}$	$1.908 \cdot 10^0$
1 : 2	$5.202 \cdot 10^0$	$-4.071 \cdot 10^{-1}$	$2.481 \cdot 10^0$
3 : 1	$4.621 \cdot 10^{-3}$	$7.331 \cdot 10^{-1}$	$1.702 \cdot 10^0$

parameters values from Table II. The data for Z_{ij} are calculated with the simulation model variant II basing on the relations:

$$Z_{ij} = i\left(\overline{m_{ij}}\right) / \overline{m_e^0} \tag{14}$$

and (for $i = 1; 1; 3$ and $j = 1; 2; 1$)

$$Z = Z_{11} + Z_{12} + Z_{31} . \tag{15}$$

The graphs show an important role of the 1 : 2 acid–amine complex at $m_a < 0.5$ mol acid/kg water. The effect becomes greater with increasing con-
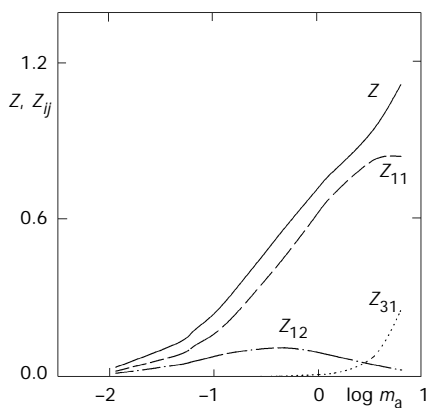

FIG. 3
Simulation of the share of individual complexes in extractant loading with model variant II. System: D-tartaric acid–Alamine 336 in LIX 54, $\overline{x} = 0.35$ kg modifier/kg binary diluent; $\overline{m_e^0} = 0.3$ mol amine/kg diluent
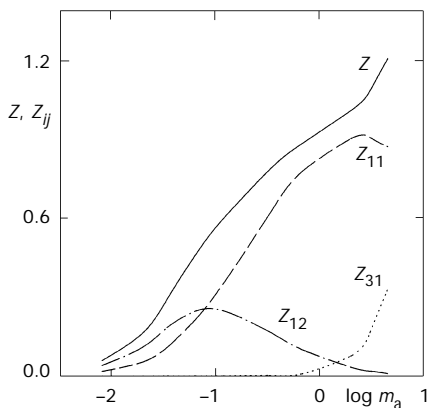

FIG. 4
Simulation of the share of individual complexes in extractant loading with model variant II. System: D-tartaric acid–Alamine 336 in LIX 54, $\overline{x} = 0.35$ kg modifier/kg binary diluent; $\overline{m_e^0} = 2.0$ mol amine/kg diluent

centration of amine in the solvent. Consistently with the assumption made before, the 1 : 1 complex is predominant in the range of medium acid molalities. At $m_a > 2$ mol acid/kg water, where the overloading occurs, the contribution of the 3 : 1 complex in Z value is greater.

CONCLUSIONS

1. Extraction equilibria in system aqueous solution of D-tartaric acid–solution of Alamine 336 in LIX 54 have been studied. A moderate overloading of amine at high acid concentrations was observed. The β-diketone in LIX 54 has been found a powerful enough modifier to prevent a third phase formation. Its effect on the acid extraction is similar to that observed in the system with MIBK (ref.[10]).

2. The extractant LIX 54 used as amine diluent promotes the acid extraction in the whole range investigated. its active component, a β-diketone, has a similar effect on extraction as MIBK studied earlier[10]. The growth of amine loading with increasing amine content indicates a significant role of the 1 : 2 complex at low and medium acid concentrations. With respect to the overloading observed at high acid concentrations formation of the 2 : 1 and/or 3 : 1 complexes has been assumed.

3. Two variants of a mathematical model, including the 1 : 1, 1 : 2 and 2 : 1 or 3 : 1 complexes, have been used for correlation and simulation of data. A somewhat better fit of data has been found with the second variant, mainly at high acid concentrations.

SYMBOLS

A_{ij}	constant in Eq. (*6*), kg diluent/mol amine
B_{ij}	constant in Eq. (*6*), kg diluent/kg modifier
C_{ij}	constant in Eq. (*6*), kg water/mol acid
K	number of complexes in the model variants
$\underline{m_a}$	molality of acid in aqueous phase, mol acid/kg water
$\overline{m_a}$	total molality of acid in organic phase, mol acid/kg diluent
$\overline{m_e}$	molality of free amine in organic phase, mol amine/kg diluent
$\overline{m_e^0}$	total molality of amine in organic phase, mol amine/kg diluent
$\overline{m_{ij}}$	molality of the $i : j$ complex in organic phase, mol/kg diluent
n	number of model parameters, Eq. (*13*)
N	number of experimental points, Eq. (*7*)
s^i	relative standard deviation, Eq. (*13*)
S	weighed sum of squares of relative deviations, Eq. (*7*)
W_q	weight of square of q-th individual point, Eq. (*7*)
$\bar{x}$	mass fraction of modifier in diluent, kg modifier/kg binary diluent
Z	loading of extractant, mol acid/mol amine

Z_{ij}	partial loading of extractant of the $i:j$ complex, mol acid/mol amine
β_{ij}^{I}	conditioned overall extraction equilibrium constant of the $i:j$ complex, $(kg/mol)^{i+j-1}$, Eq. (*3*)
β_{ij}	thermodynamic overall extraction equilibrium constant of the $i:j$ complex, $(kg/mol)^{i+j-1}$, Eq. (*3*)
$\overline{\gamma_e}$	activity coefficient of free amine in organic phase
γ_{ij}	activity coefficient of the $i:j$ complex in organic phase

Subscripts

calc	calculated values
exp	experimental values
a	acid
e	amine
i	number of acid molecules in the $i:j$ complex
j	number of amine molecules in the $i:j$ complex
q	individual experimental point

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