Membrane-Based Extraction Joined with Membrane-Based Stripping in a Circulating Arrangement II. Extraction of Organic Acids*

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The paper presents an experimental verification of a simple model for a circulating arrangement of extraction joined with stripping (re-extraction) developed in Part I. Membrane-based extraction connected with simultaneous membrane-based stripping of organic acids was performed. The original derivative equations for both stripping and solute phases were modified into a new form. This form of the model equations reflects the fact that for a system with a very high value of the distribution coefficient, model equations are independent of the arrangement of flow through the contactor. Both butyric acid and 5-methyl-2-pyrazinecarboxylic acid (MPCA) were extracted from aqueous solutions by TOA diluted in alkanes or xylene and successively stripped by an aqueous solution of sodium hydroxide. For both systems time courses of the concentration of carboxylic acids were used. By fitting the experimental time dependences of concentrations of transferred component in the feed, solute and stripping phase containers, the values of model parameters, namely extraction mass transfer coefficient, $K_{\rm e}$, and the stripping mass transfer coefficient, $K_{\rm s}$, were obtained.

In the last two decades several technologies using the hollow fibre contactors (HFC) in membrane-based extraction [1] which can be found in the separation of carboxylic acids [2—8], amino acids [9], drugs [10, 11], metals [7, 12—17], and organic pollutants [6, 10, 13] were established.

Membrane-based extraction was carried out in HFC using microporous membranes to immobilize the interface in the pores without dispersion. Similarly the membrane-based stripping was carried out in HFC using the same effect of immobilization of solute in microporous membranes as in extraction from the membrane. This arrangement provides several advantages over classical extractors, such as a very high contact area per volume, the possibility of operating with a wide range of flow rates, no need of density differences to achieve phase separation, and none or very low emulsification. On the other hand, a higher pressure has to be kept in the aqueous phase to prevent penetration of organic phase through fibre pores.

Therefore, HFC could be used not only to extract the solute but also to concentrate it. For this reason, simultaneous extraction and stripping of carboxylic acids were accomplished using two hollow fibre contactors connected in series. This configuration assured that saturation of the carrier did not occur, as it was continuously regenerated. As a consequence, the carrier quantity could be reduced along with the amount of the solute phase. Thus, the associated operating costs of the process could be diminished, too.

THEORETICAL

In the last years several models of membrane-based extraction joined with membrane-based stripping have been proposed [3, 4, 8, 9, 12, 18—20]. The presented article is linked with our previous works [21, 22] in which a simple model for a circulating arrangement of extraction joined with stripping (re-extraction) was developed.

The Modification of the Proposed Model in Part I [21] for the Case when the Distribution Coefficient for the Stripping Process, $D_{\rm R}$, is of a High Value

In the stripping a chemical binding between the stripping component and the stripping agent on the stripping interface is very frequent. Thus

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$$D_{\rm R} \Rightarrow \infty$$
 (1)

In this case, eqns (33) and (37) presented in Part I for solvent loop and for countercurrent and cocurrent flow arrangement, respectively, are transformed into the following relation

$$c_{S2t} = c_{S1t} \exp\left(-N_S\right) \tag{2}$$

which is independent of the arrangement of the flow of phases through the hollow fibre contactor for membrane-based stripping, and eqns (40) and (41) presented in Part I are simplified to the following form

$$-\frac{\mathrm{d}c_{S0t}}{\mathrm{d}t} = \tau_{S}^{-1} \left\{ c_{S0t} \left[1 - \exp\left(-N_{S} \right) \right] - \frac{\dot{V}_{F}}{V_{S}} \left(c_{F1t} - c_{F2t} \right) \exp\left(-N_{S} \right) \right\}$$
(3)

In eqns (2) and (3) N_S is number of transfer units defined as

$$N_{\rm S} = \frac{K_{\rm s} A_{\rm s}}{\dot{V}_{\rm c}} \tag{4}$$

and

$$\tau_{\rm S} = \frac{V_{\rm S}}{\dot{V}_{\rm S}} \tag{5}$$

is space time (mean residence time).

In the stripping loop, eqns (46) and (48) presented in Part I are also transformed to a form independent of the arrangement of the flow of phases through HFC for membrane-based stripping

$$\frac{\mathrm{d}c_{\mathrm{R0}t}}{\mathrm{d}t} = \tau_{\mathrm{R}}^{-1} \frac{\dot{V}_{\mathrm{S}}}{V_{\mathrm{R}}} \left[c_{\mathrm{S0}t} + \frac{\dot{V}_{\mathrm{F}}}{\dot{V}_{\mathrm{S}}} \left(c_{\mathrm{F1}t} - c_{\mathrm{F2}t} \right) \right] \cdot \left[1 - \exp\left(-N_{\mathrm{S}} \right) \right]$$
(6)

where space time $\tau_{\rm R}$ is defined as

$$\tau_{\rm R} = \frac{V_{\rm R}}{\dot{V}_{\rm p}} \tag{7}$$

For feed-raffinate loop model equations for ccuntercurrent flow of phases were used

$$-\frac{\mathrm{d}c_{\mathrm{F1}t}}{\mathrm{d}t} = \frac{1 - \mathrm{e}^{-W}}{\tau_{\mathrm{F}}(1 - E_{\mathrm{F}}\mathrm{e}^{-W})} \left(c_{\mathrm{F1}t} - \frac{c_{\mathrm{S0}t}}{D_{\mathrm{F}}}\right)$$
(8)

and

$$c_{\rm F2} = c_{\rm F1} \frac{(1 - E_{\rm F}) \,\mathrm{e}^{-W}}{1 - E_{\rm F} \mathrm{e}^{-W}} + c_{\rm S0} \frac{1 - \mathrm{e}^{-W}}{D_{\rm F} \,(1 - E_{\rm F} \mathrm{e}^{-W})}$$
 (9)

where

$$W = (1 - E_{\rm F}) N_{\rm F} \tag{10}$$

$$E_{\rm F} = \frac{\dot{V}_{\rm F}}{D_{\rm F}\dot{V}_{\rm S}} \tag{11}$$

$$N_K = \frac{e^{A_e}}{\dot{V}_E}$$
 (12)

$$\tau_{\rm F} = \frac{V_{\rm F}}{\dot{V}_{\rm F}} \tag{13}$$

Estimation of the Coefficients $K_{\rm e}$ and $K_{\rm s}$

The value of the coefficient $K_{\rm e}$, required as an initial value by the optimization procedure, can be estimated from the beginning of the time course of the extracted component in the feed reservoir. At the beginning of the extraction process connected with simultaneous re-extraction, the influence of re-extraction on the concentration decrease in the feed phase is small and could be neglected. For extraction in a batch circulating system not joined with stripping, the following relation in Part I was proposed

$$\ln \frac{c_{\rm F1t} - c_{\rm F0} \frac{\bar{E}_{\rm F}}{1 + \bar{E}_{\rm F}}}{\frac{c_{\rm F0}}{1 + \bar{E}_{\rm F}}} = -X\Theta \tag{14}$$

where

$$\bar{E}_{\rm F} = \frac{V_{\rm F}}{D_{\rm F} V_{\rm S}} \tag{15}$$

$$X = \frac{1 - \exp(-W)(1 + \bar{E}_{F})}{1 - E_{F} \exp(-W)}$$
 (16)

The value of the coefficient $K_{\rm s}$ can be estimated from the time corresponding to the maximum on the curve of the time dependence of concentration of the extracted component in the solvent reservoir. For the extraction system with high values of $D_{\rm F}$ ($D_{\rm F} > 100$) and extremely high values of $D_{\rm R}$ the following relation is valid [23]

$$t_{\text{max}} = \frac{\tau_{\text{F}}\tau_{\text{S}}}{Q\tau_{\text{F}} - T\tau_{\text{S}}} \ln \frac{Q\tau_{\text{F}}}{T\tau_{\text{S}}}$$
(17)

where $t_{\rm max}$ is the time corresponding to the maximum concentration of the extracted component in the solvent reservoir, and

$$Q = 1 - \exp(-N_{\rm S}) \tag{18}$$

$$T = 1 - \exp(-N_{\rm F})$$
 (19)

The value of the coefficient K_s obtained from eqn (17) is a good first estimate.

Mass Transfer System

The overall reaction of the formation of a complex between acid (HA) (in our case butyric acid (BA) or 5-methyl-2-pyrazinecarboxylic acid (MPCA)) and carrier (P), e.g. trioctylamine (TOA) on the feedmembrane interface can be written as

$$n{\rm HA} + \overline{{\rm P}} \Leftrightarrow \overline{{\rm HA}_n {\rm P}}$$
 (A)

where bars denote species dissolved in the membrane phase. Experimental equilibrium concentration data

were used for the determination of the distribution coefficient $D_{\mathbf{F}}$.

The overall reaction for the complex decomposition on the membrane stripping solution interface is

$$\overline{\text{HA}_n P} + n\text{OH}^- \rightarrow n\text{A}^- + n\text{H}_2\text{O} + \overline{P}$$
 (B)

A value of the distribution coefficient $D_{\rm R}$ can be considered as extremely high $(D_{\rm R} \to \infty)$.

EXPERIMENTAL

Butyric acid (BA, Merck) with a purity >99 mass % was used. The density and molar mass of pure BA were 959 kg m⁻³ (at 30 °C) and 88.1 kg kmol⁻¹, respectively.

5-Methyl-2-pyrazinecarboxylic acid (MPCA) was received from the Research Institute of Organic Synthesis (Pardubice, Czech Republic) in a crystalline form with purity higher than 99 mass %. Molar mass of MPCA is $138.14~\mathrm{g}~\mathrm{mol}^{-1}$. Density of MPCA at 25 °C was $1403~\mathrm{kg}~\mathrm{m}^{-3}$.

n-Alkanes. A dodecane fraction of n-alkanes (Slovnaft Bratislava, Slovakia) was used as diluent. The composition estimated by GC in mass % and mean molar mass were 5.4 mass % C_{10} , 40.6 mass % C_{11} , 40.4 mass % C_{12} , and 13.1 mass % C_{13} , and 164.2 kg kmol⁻¹, respectively. Density and kinematic viscosity at 30 °C were $\rho = 741.4$ kg m⁻³ and $v = 1.596 \times 10^{-6}$ m² s⁻¹.

Xylene, technical quality (Slovnaft Bratislava, Slovakia), was used as diluent. The mean molar mass was 94.15 kg kmol⁻¹. Density and kinematic viscosity at 30 °C were $\rho = 859.9$ kg m⁻³ and $v = 0.651 \times 10^{-6}$ m² s⁻¹.

Trioctylamine (TOA, Fluka) with a purity >95 mass % was used as received. The density and molar mass of pure TOA was 804.3 kg m⁻³ at 30 °C and 353.7 kg km⁻¹, respectively.

Feed: either an aqueous solution of butyric acid, $c_{\rm F0}=0.48$ —1.2 kmol m⁻³, pH_{F2} = 2.4—2.6 (without adjustment of pH) or an aqueous solution of MPCA, $c_{\rm F0}\approx 0.1$ kmol m⁻³ in an aqueous solution of Na₂SO₄ of the concentration 1 kmol m⁻³, pH_{F0} = 1.

Solvent: 0.4 kmol m⁻³ TOA in n-alkanes, density and kinematic viscosity at 30 °C were $\rho=751.9$ kg m⁻³ and $\upsilon=2.026\times10^{-6}$ m² s⁻¹.

For MPCA 0.4 kmol m⁻³ TOA in xylene was used. Density and kinematic viscosity at 30 °C were $\rho = 847.9 \text{ kg m}^{-3}$ and $v = 0.84 \times 10^{-6} \text{ m}^2 \text{ s}^{-1}$.

Stripping solution: aqueous solution of NaOH with a concentration 1.5—3.0 kmol m⁻³.

Temperature: 30°C.

For simultaneous extraction and stripping two hollow fibre modules Liqui-Cel Extra-Flow 2.5 inch \times 8 inch (Hoechst-Celanese, USA) with a cross-flow of phases Celgard X-30 were used. In these modules microporous polypropylene hollow fibres were assembled.

Table 1. Characteristics of the Hollow-Fibre Module and Membrane Celgard X-30

Module	Hollow fibres Celgard X-30		
Diameter/cm	6.3	Internal diameter/µm	240
Length of module/cm	20	Wall thickness/ μ m	30
Effective fibre length/cm	15	Porosity/%	40
Number of fibres	10000	Effective pore size/ μ m	0.03
Inner effective area/m ²	0.56	Tortuosity	2.0
Specific area/(m ² m ⁻³)	2930		

The characteristics of modules and fibres are presented in Table 1.

The concentration of BA and MPCA in the aqueous phases was determined by an isotachophoretic analyzer EA 100 (VILLA, Slovakia) using an HCl—histidine solution with pH = 6 as leading electrolyte. Concentration of BA in the organic phase was determined also isotachophoretically after stripping of acids to the solution of NaOH.

Procedure

The scheme of the extraction and stripping units is presented in Fig. 1. The feed solution circulates from the reservoir through the fibres and back into the reservoir. The tube side Reynolds numbers range from 0.5 to 16. The solute circulates in the shell side of the modules, countercurrently in the extraction module and cocurrently in the stripping module. The stripping solution circulates from the reservoir through the fibres of the stripping contactor and back into the reservoir.

The volume of the feed was approximately 5 dm³. The volume of the feed phase in HFC and connected pipes was approximately 0.12 dm³. Thus, the ratio of the volume of phase in HFC to the volume of phase in the reservoir was less than 0.03.

Note: The volume of the feed phase in the filter vessel and in connecting pipes before the extraction module was added to the volume of the feed reservoir. The volume of connected pipes at the exit of the contactor was considered negligibly small. The same procedure was used for the stripping module.

For the membrane-based stripping a volume of the stripping phase of 1.0—1.2 dm³ was used. The ratio of the volume of the stripping phase in HFC to the volume of the stripping phase in the reservoir was approximately 0.12. The volume of the solute used for the membrane-based extraction joined with membrane-based stripping was around 0.8 dm³. The volume of the solute hold-up in the extraction HFC and in the stripping HFC was around 0.4 dm³. Sampling of the phases as well as concentration variation of the transferred component caused a change of volumes of the phases. In all calculations the average volume of the phase in the reservoirs was assumed. The flow of

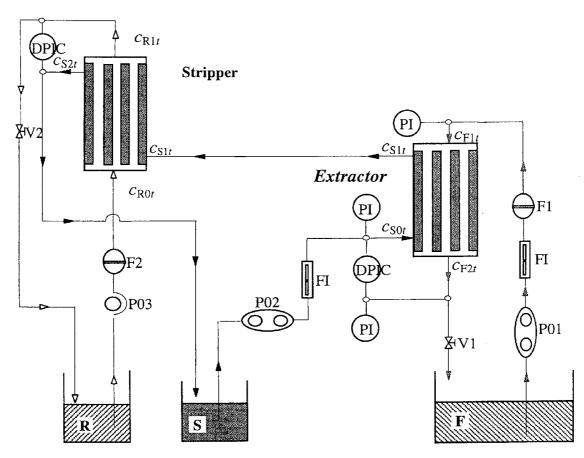


Fig. 1. Scheme of the extraction and stripping unit. F - feed reservoir, S - solvent reservoir, R - stripping reservoir, FI - flow indication (rotameters), PI - pressure indication, DPIC - difference pressure indication and control, F1, F2 - sintered glass filters, V1, V2 - metering valves for the adjustment of pressures of the aqueous phases, P01, P02 - gear pumps, P03 - peristaltic pump.

phases through contactors during the experiment was kept approximately constant.

For evaluation of the concentration variation with time of experiment the start of experiment has been defined. In this study, the moment when the stripping contactor began to be filled with the membrane phase was considered as zero time of experiment. This definition was satisfying not only for the feed reservoir but also for the stripping reservoir. The stripping contactor worked insufficiently during the short starting period in which the module was filled with solute. However, the influence of this transition period on the whole experiment was negligible. For the solute reservoir as well as the outlet of the solute from the stripping contactor the chosen start of counting time of the experiment caused some time delay of approximately 60 s (depending on the solute flow rate).

The pressure of the aqueous phase was maintained higher than the pressure of the organic phase (about 60 kPa) as the hydrophobic fibres were preferentially wetted by the organic phase. The pressure was adjusted using the valves at the outlet of the modules. The pressure and pressure differences were measured by piezoelectric sensors (Omega, USA) connected to a control unit.

Sintered glass filters filtered the streams prior entering the contactors, especially those passing through the fibre lumen. The feed and organic phases were fed by tooth pumps (Verder, USA) and the stripping solution by a peristaltic pump. The flow rate of the feed solution and the organic phase was measured at the modules inlet using rotameters.

At the beginning of measurements, both contactors were at first filled with aqueous phases and their pressure was adjusted. Only then the solvent pumping started.

Results of these experiments were used not only for the verification of the proposed simple model. Another purpose of these experiments was evaluation of coefficients of the mass transfer, $K_{\rm e}$ and $K_{\rm s}$, by using macroscopical model [24]. So, not only samples from reservoirs were taken, but also the samples of raffinate $(c_{\rm F2}t)$, loaded solvent $(c_{\rm S1}t)$, solvent after the stripper $(c_{\rm S2}t)$ and in the feed $(c_{\rm F1}t)$, and samples from stripping reservoirs $(c_{\rm R0}t)$ and rarely from the solvent reservoir $(c_{\rm S0}t)$ were collected for the analysis of concentration of the transferred component as a function of time.

RESULTS AND DISCUSSION

In the case of butyric acid extraction, the polynomial correlation between the distribution coefficient, $D_{\rm F}$, and the solute concentration of butyric acid was

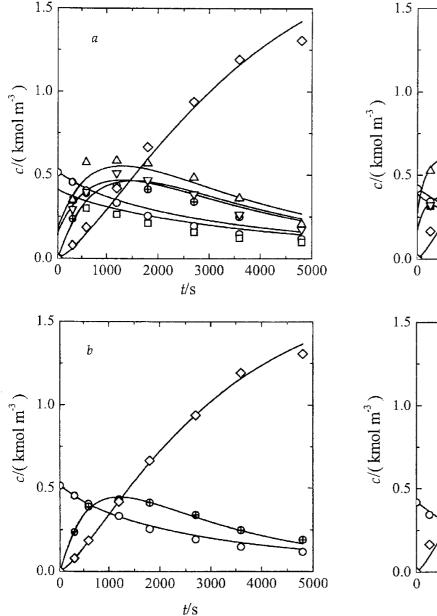


Fig. 2. Change of BA concentration with time in the membrane-based extraction – stripping system with circulating arrangement. Best fit of a) the whole set of experimental data ($\dot{V}_{\rm F}=8.0\times10^{-6}~{\rm m}^3~{\rm s}^{-1}$, $\dot{V}_{\rm S}=4.2\times10^{-6}~{\rm m}^3~{\rm s}^{-1}$, $\dot{V}_{\rm R}=2.0\times10^{-6}~{\rm m}^3~{\rm s}^{-1}$. Optimization results: $K_{\rm e}=4.30\times10^{-6}~{\rm m}~{\rm s}^{-1}$, $K_{\rm s}=1.38\times10^{-6}~{\rm m}~{\rm s}^{-1}$) and b) the concentration measured in the three reservoirs ($K_{\rm e}=5.98\times10^{-6}~{\rm m}~{\rm s}^{-1}$, $K_{\rm s}=1.79\times10^{-6}~{\rm m}~{\rm s}^{-1}$). Experimental concentration of the acid in 0 feed reservoir, \Box raffinate, \triangle solute from extraction module, ∇ solute from stripping module, \oplus solute reservoir, \Diamond stripping reservoir.

used [25]

$$D_{\rm F} = 0.70 + 11.35C_{\rm S} - 17.56(C_{\rm S})^2 + 19.70(C_{\rm S})^3 - 14.01(C_{\rm S})^4 + 3.89(C_{\rm S})^5$$
 (20)

For extraction of MPCA the analogical correlation had the form [6, 26]

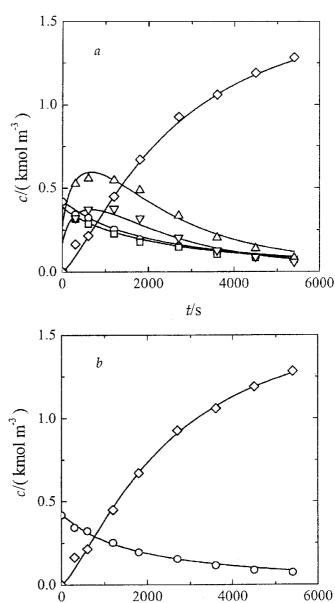


Fig. 3. Change of BA concentration with time in the membrane-based extraction – stripping system with circulating arrangement. Best fit of a) the whole set of experimental data, except of data for the solute reservoir $(\dot{V}_{\rm F}=20.53\times 10^{-6}~{\rm m}^3~{\rm s}^{-1},\,\dot{V}_{\rm S}=2.22\times 10^{-6}~{\rm m}^3~{\rm s}^{-1},\,\dot{V}_{\rm R}=1.97\times 10^{-6}~{\rm m}^3~{\rm s}^{-1}.$ Optimization results: $K_{\rm e}=7.52\times 10^{-6}~{\rm m}~{\rm s}^{-1},\,K_{\rm S}=1.91\times 10^{-6}~{\rm m}~{\rm s}^{-1})$ and b) the concentration measured in the feed and stripping reservoirs ($K_{\rm e}=9.38\times 10^{-6}~{\rm m}~{\rm s}^{-1},\,K_{\rm s}=1.66\times 10^{-6}~{\rm m}~{\rm s}^{-1})$. The meaning of the symbols is the same as in Fig. 2.

t/s

$$D_{\rm F} = 6.679 - 7.313C_{\rm S} \tag{21}$$

In eqns (20) and (21) $C_{\rm S}$ is defined as the ratio

$$C_{\rm S} = \frac{c_{\rm S}}{c_{\rm F0}} \tag{22}$$

for $c_{\rm F0} = 1 \text{ kmol m}^{-3}$.

If eqn (1) is valid, for membrane-based stripping accompanied with chemical binding between the

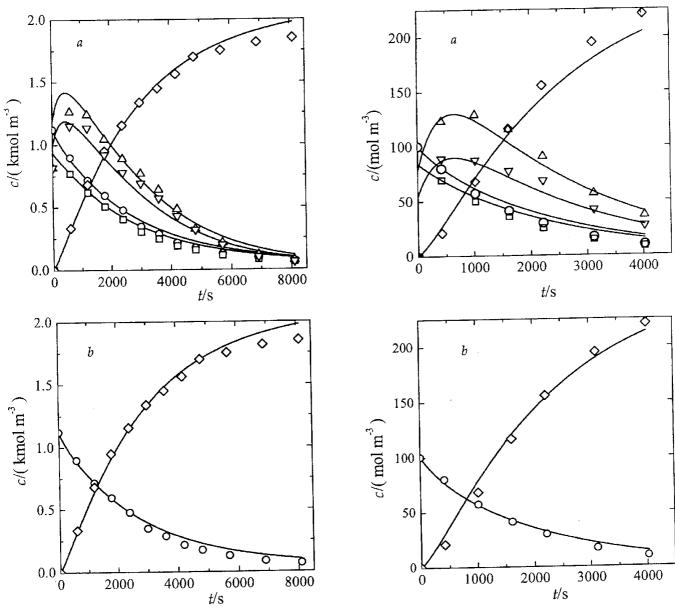


Fig. 4. Change of BA concentration with time in the membrane-based extraction – stripping system with circulating arrangement. Best fit of a) the whole set of experimental data, except of data for the solute reservoir ($\dot{V}_{\rm P}=12.84\times10^{-6}~{\rm m}^3~{\rm s}^{-1},\,\dot{V}_{\rm S}=6.22\times10^{-6}~{\rm m}^3~{\rm s}^{-1},\,\dot{V}_{\rm R}=1.97\times10^{-6}~{\rm m}^3~{\rm s}^{-1}.$ Optimization results: $K_{\rm e}=5.25\times10^{-6}~{\rm m}~{\rm s}^{-1},\,K_{\rm S}=1.50\times10^{-6}~{\rm m}~{\rm s}^{-1})$ and b) the concentration measured in the feed and stripping reservoirs ($K_{\rm e}=5.25\times10^{-6}~{\rm m}~{\rm s}^{-1},\,K_{\rm S}=1.50\times10^{-6}~{\rm m}~{\rm s}^{-1}$). The meaning of the symbols is the same as in Fig. 2.

Fig. 5. Change of MPCA concentration with time in the membrane-based extraction – stripping system with circulating arrangement. Best fit of a) the whole set of experimental data, except of data for the solute reservoir ($\dot{V}_{\rm F}=11.50\times 10^{-6}~{\rm m}^3~{\rm s}^{-1},\,V_{\rm S}=2.07\times 10^{-6}~{\rm m}^3~{\rm s}^{-1},\,\dot{V}_{\rm R}=2.35\times 10^{-6}~{\rm m}^3~{\rm s}^{-1}.$ Optimization results. $K_{\rm e}=4.29\times 10^{-6}~{\rm m}~{\rm s}^{-1},\,K_{\rm s}=1.69\times 10^{-6}~{\rm m}~{\rm s}^{-1})$ and b) the concentration measured in the feed and stripping reservoirs ($K_{\rm e}=5.02\times 10^{-6}~{\rm m}~{\rm s}^{-1},\,K_{\rm s}=1.40\times 10^{-6}~{\rm m}~{\rm s}^{-1}$). The meaning of the symbols is the same as in Fig. 2.

transferred component and the stripping agent the influence of arrangement of the flow of phases in the stripping HFC is negligible. Although the flow through shell side of the contactors was rather cross flow, the countercurrent flow through extraction HFC was satisfying approximation.

Three methods for the evaluation of experimentally measured data were used. In the first method, all measured time dependences of the concentration of the transferred component were taken into account.

That means that time dependences of the concentration of the transferred component in the reservoirs as well as in the raffinate and in the solute at the exit of contactors (extraction HFC and stripping HFC) were considered. The second method fitted the time dependences of the concentrations of the transferred component in reservoirs of the feed, the solute, and the stripping phase. The third method, which was fully sufficient for the proposed model, involved the time dependences of the concentration of the transferred

Table 2. Statistical Output of the Presented Experiments

Fig.		$c_{\mathrm{F1}t}$	$c_{\mathbf{F2}t}$	$c_{\mathrm{S0}t}$	$c_{\mathrm{S1}t}$	$c_{\mathrm{S}2t}$	$c_{\mathrm{R0}t}$	Data set	$\frac{K_{\rm e}/10^{-6}}{{ m m s}^{-1}}$	$K_{\rm s}/10^{-6}$ m s ⁻¹	MSC
2 <i>a</i>	c	0.998	0.359	0.942	0.981	0.988	0.951	0.962	4.30 ± 0.36	1.38 ± 0.07	2.34
	cod	0.961	0.791	0.851	0.856	0.856	0.982	0.911			
2b	c	0.999		0.996			0.999	0.999	5.98 ± 0.17	1.79 ± 0.04	5.61
	cod	0.998		0.99			0.997	0.997			
3a	c	0.998	0.307		0.91	0.912	0.999	0.968	7.52 ± 0.77	1.91 ± 0.14	2.61
	cod	0.986	0.742		0.79	0.802	0.997	0.906			
3b	c	0.997					0.999	0.999	9.38 ± 1.95	1.66 ± 0.24	5.83
	cod	0.992					0.997	0.988			
4a	c	0.999	0.481		0.968	0.981	0.998	0.971	5.25 ± 0.48	1.50 ± 0.07	2.51
	cod	0.988	0.397		0.91	0.961	0.991	0.924			
4b	С	0.999					0.998	0.998	5.25 ± 0.86	1.50 ± 0.21	5.06
	cod	0.988					0.992	0.994			
5a	c	0.999	0.263		0.684	0.663	0.998	0.921	4.29 ± 0.53	1.69 ± 0.18	1.56
	cod	0.987	0.915		0.44	0.435	0.992	0.812			
5b	С	0.998					0.998	0.918	5.02 ± 1.04	1.40 ± 0.28	4.91
	cod	0.991					0.993	0.994			

c - correlation, cod - coefficient of determination, MSC - model selection criterion.

component only in two reservoirs, those of the feed and the stripping phase.

The experimental data of BA and their fit by the model are graphically presented in Figs. 2—4. Optimization of the overall mass transfer coefficients, $K_{\rm e}$ and $K_{\rm s}$, was performed using the standard program Scientist from MicroMath Inc. Satisfactory coincidence between experimental and calculated time courses of the concentration for the transferred component was obtained.

Fig. 2a shows the results of fitting the complete set of measured BA concentrations; meanwhile, in Fig. 2b the fit of the BA concentration in the three reservoirs is presented.

Figs. 3 and 4 represent the fit of the BA concentrations obtained for experiments with different initial conditions. In these experiments, different concentrations of BA in the feed and solute phases were used in order to test the concentration sensitivity of distribution coefficient, $D_{\rm F}$. Figs. 3a and 4a show the time dependences of measured BA in the feed and stripping reservoirs and also at the solute outlet from contactors and in the raffinate. Figures denominated b present the results of fitting BA concentrations in the feed and stripping reservoirs.

In Figs. 5a and 5b the time variation of experimental MPCA concentrations in the feed and stripping reservoirs and also at the solute outlets from contactors and in the raffinate was fitted according to the second and third method, respectively.

Note: In Figs. 2—5 solid lines are calculated values of the BA or MPCA concentrations from the proposed model equations.

If in addition to the concentration of the transferred component in the reservoirs also concentrations of this component at the exit of the contactors, in the raffinate from the extraction HFC, and in the solute phase at the exit from the extraction HFC and the

stripping HFC were taken into account, the calculated values of coefficients $K_{\rm e}$ and $K_{\rm s}$ were, in generally, different from the values of coefficients $K_{\rm e}$ and $K_{\rm s}$ calculated only from concentrations in the reservoirs. The reason of this phenomenon could be in using a large quantity of data measured using the samples taken from tubing. Experimental data obtained using samples of the solute and raffinate taken from connecting pipes could be more influenced by the fluctuations of the flow of phases and some inhomogeneities. Statistical output of the presented experiments is shown in Table 2.

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SYMBOLS

$A_{\mathbf{e}}$	interfacial area	m^2
c	molar concentration	${ m kmol}~{ m m}^{-3}$
c_{F0}	initial concentration of the extracte	ed com-
	ponent in the feed	${ m kmol}~{ m m}^{-3}$
C	dimensionless concentration	
D	diffusion coefficient	${ m m^2 \ s^{-1}}$
$D_{ m F}$	distribution coefficient in the feed-	$\operatorname{-solvent}$
	system	
$D_{\mathbf{R}}$	distribution coefficient in the so	lvent
	stripping solution system	
$E_{\mathbf{F}}$	quantity defined by eqn (11)	
$ar{E}_{ extbf{F}}$	quantity defined by eqn (15)	
K	overall mass transfer coefficient	${ m m~s^{-1}}$
$N_{ m F}$	number of transfer units for the ext	raction
	process, eqn (12)	
$N_{ m S}$	number of transfer units for the st	ripping
	process, eqn (4)	
Q	quantity defined by eqn (18)	
t	time	S
T	quantity defined by eqn (19)	

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V	volume of the reservoir		m^3
\dot{V}	volumetric flow of the liquid	m^3	s^{-1}
W	quantity defined by eqn (10)		
X	quantity defined by eqn (16)		
Θ	dimensionless time $(t/\tau_{\rm F})$		
$ au_{ extbf{F}}$	space-time of the feed circuit		s
$ au_{ m S}$	space-time of the solute circuit		s
$ au_{ m R}$	space-time of the stripping phase circu	it	s

Indices

е

F

1	ınner
EC	extraction contactor
SC	stripping contactor
R	stripping phase
S	stripping
S	solvent (organic phase)
t	time-dependent
0	marking of the place
1	marking of the place
2	marking of the place

extraction

feed phase

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ERRATUM

Membrane-Based Extraction Joined with Membrane-Based Stripping in a Circulating Arrangement. I

M. VAJDA

In Chem. Pap. 56 (5) 288—294 (2002) on page 293 instead of eqn (17) in the left column, line 8 from above, should be

$$-\frac{\mathrm{d}c_{\mathrm{F1}t}}{\mathrm{d}t} = \frac{1 - \mathrm{e}^{-W}}{\tau_{\mathrm{F}}(1 - E_{\mathrm{F}}\mathrm{e}^{-W})} \left(c_{\mathrm{F1}t} - \frac{c_{\mathrm{S0}t}}{D_{\mathrm{F}}} \right) \tag{15}$$