### Palaty:

# MASS TRANSFER IN LIQUID IN APPARATUS WITH MOBILE PACKING. APPLICATION OF MODEL OF CASCADE OF IDEAL MIXERS

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The paper deals with the determination of parameters of a cascade of ideal mixers (i.e. their number and mean residence time of liquid in each member) in modelling the liquid flow on a plate with mobile packing. The number of cascade members has been found practically independent of the gas and liquid velocities and the static bed heights in the gas velocity range from 1.0 to  $3.5 \text{ m s}^{-1}$ , liquid velocity range from  $5.36 \cdot 10^{-3}$  to  $12.5 \cdot 10^{-3} \text{ m s}^{-1}$ , and the range of static bed height from  $21 \cdot 10^{-3}$ to  $47 \cdot 10^{-3}$  m. The mean residence time is increased with increasing static bed height and is decreased with increasing velocities of both gas and liquid. The calculated parameters of the model of flow of liquid and the experimental data on desorption of CO<sub>2</sub> from water by a stream of air have been used to calculate the values of mass transfer coefficient in liquid which have been compared with those obtained from the dispersion model. The both approaches have been found to give practically identical results.

The main prerequisite for a correct calculation of the mass transfer coefficient in liquid is the information about the flow of liquid through the system. A number of models have been suggested and elaborated for this purpose, their parameters being obtainable by the transient response technique. The dispersion model<sup>1–8</sup> is preferably used for the quantification of flow of liquid on a plate of an apparatus with mobile packing, other models such as the model of cascade of ideal mixers<sup>9</sup> and the cell model<sup>10,11</sup> being used only rarely.

The aim of the present work is to find, on the basis of experimental data, the parameters of a simple model of cascade of ideal mixers, adopt them in the calculation of mass transfer coefficient in liquid, and compare the latter with the values obtained earlier from the case considering the dispersion flow of liquid on the plate.

# THEORETICAL

The mass balance of the tracer in a cascade of N ideal mixers of the same volume is given by a set of simple differential equations<sup>12</sup>

$$c_0 = c_1 + \tau_{\rm m} (dc_1/d\tau) \qquad j = 1$$
 (1)

$$c_{i-1} = c_i + \tau_{\rm m} ({\rm d} c_i / {\rm d} \tau) \qquad j = 2, 3, \dots, N$$
 (2)

with the initial condition  $\tau = 0$ ,  $c_j = 0$  (j = 1, 2, ..., N). In Eq. (1),  $c_0$  represents the time dependence of concentration of the tracer at the inlet of the first cascade member,  $\tau_m = V_{L,j}/\dot{V}_L$  is the mean residence time of liquid in each cascade member, and  $V_{L,j}$  is the volume of liquid in the respective member. If the liquid fills only a part of volume  $V_j$  of the cascade member, then  $V_{L,j} = \varepsilon_L V_j = \varepsilon_L V_t / N$  ( $\varepsilon_L$  is a dimensionless hold-up of liquid in the layer,  $V_t$  is the total volume of layer). In special cases (e.g. an ideal step change of concentration) there exists an analytical solution to the equation system (1) and (2) (see ref.<sup>12</sup>). If  $c_0$  is a general function of time, the given equation system must be solved numerically.

If the parameters of cascade of ideal mixers, i.e. N and  $\tau_m$ , are available, then the mass transfer coefficients in liquid can be determined in a simple way.

# EXPERIMENTAL

For the determination of hydrodynamic characteristics of the plate with mobile packing and the determination of mass transfer coefficient in liquid the earlier data<sup>13,14</sup> were used. The number *N* of cascade members and the mean residence time of liquid in each member,  $\tau_m$ , were determined from the measurements in unsteady state, where the time dependence of the tracer (KCl solution) concentration at the inlet and outlet of the layer was monitored. The apparatus and method of measurement are described in detail elsewhere<sup>13</sup>. The mass transfer coefficients in liquid were determined from the experimental data obtained for the desorption of CO<sub>2</sub> from water by a stream of air<sup>14</sup>.

#### TREATMENT OF RESULTS

The parameters of the cascade of ideal mixers, N and  $\tau_m$ , were determined on the basis of the original procedure developed for the purpose of the present study, which procedure consists of the numerical integration of the set of simple differential equations (1) and (2) and subsequent optimization procedure. The number of the equations (1) and (2) is identical with that of the cascade members and must always be an integer. The optimization procedure used, however, takes the  $N_{opt}$  parameter as a real number which need not necessarily be integer. Therefore, the program for numeric solution of Eqs (1) and (2) was assembled in such a way as to ensure the following procedure. In the cases in which the given step of the optimization procedure gave the  $N_{opt}$  value as a real noninteger number, the number N of Eqs (1) and (2) for further iteration was equal to the integer part of  $N_{opt}$  plus 1. The decimal part determined the fraction of liquid volume and (hence) of the mean residence time of liquid in the last member of cascade with respect to the volume and  $\tau_{\rm m}$  in the preceding member, respectively. So, e.g., for  $N_{\rm opt} = 2.3$  the subsequent iteration solves three equations (1) and (2), the  $\tau_{\rm m}$  value in the third equation being 30% of that in the first and/or second member. In a simpler case the optimized parameter  $N_{\rm opt}$  could be considered to be an integer number. Such a procedure, however, would result in greater differences between the model and experiment for smaller numbers of cascade members.

The procedure adopted can be summarized in the following steps:

1. The initial estimate of  $N_{opt}$  and  $\tau_m$  parameters. (In all the experiments  $N_{opt}^{(0)} = 5$ ,  $\tau_m^{(0)} = 1.0$  s.)

2. Numerical integration of the differential equations (1) and (2) by the Runge–Kutta method of the 4th order with the integration step of 0.1 s. This step provides the calculated values of concentration of the tracer at the outlet of the last member of cascade,  $c_{N,\text{calc}}^i$ 

3. Calculation of the objective function defined by the equation

$$F(N_{\text{opt}}, \tau_{\text{m}}) = \sum_{i=1}^{n} (c_{N,\text{exp}}^{i} - c_{N,\text{calc}}^{i})^{2}, \qquad (3)$$

where *n* is the number of experimental data in the given time series (see ref.<sup>13</sup>).

4. Carrying out one optimization step of the objective function (3) and obtaining the corrected values of  $N_{opt}$  and  $\tau_m$  parameters. For this purpose, the method by Nelder and Mead<sup>15</sup> was used.

5. The procedure in the steps 2. - 4. was repeated until reaching the minimum of the objective function (3).

Figure 1 gives typical time dependences of concentration of tracer at the inlet and outlet of the layer (both experimental and optimized values). The comparison of calcu-



#### Fig. 1

Time dependence of concentration of tracer: 1 experimental values (inlet), 2 experimental values (outlet), 3 calculated values (outlet)

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lated and experimental concentrations of tracer at the outlet of layer shows that there is and adequate agreement between experiment and the model. At some points of the response curves, greater deviations are observed between the experimental and calculated (from the model adopted) concentration values of the tracer. The breaks at the experimental response curve could be due to the existence of several regions of liquid with different residence times of liquid in these regions. Due to the considerable irregularities in the three-phase fluid layer on the plate, the response curves obtained in repeated experiments at the same hydrodynamic conditions are not identical, which prevents the identification of the above-mentioned regions of liquid.

The mass transfer coefficients in liquid,  $k_L a$ , were determined for the known N and  $\tau_m$  parameters from Eqs (4) which were derived, for the purposes of the present work, from the mass balance of component A (CO<sub>2</sub>) in the desorption and from the definition of mean residence time of liquid in the individual members of cascade under the presumption of negligible resistance to the mass transfer in gas phase

$$c_{A,j-1} - c_{Aj} = k_{L}a\tau_{m}H_{A}((p_{Aj}'' - p_{Aj}')/\ln((c_{Aj} - H_{A}p_{Aj}')/(c_{Aj} - H_{A}p_{Aj}'')))$$
(4)  

$$j = 1, 2, 3, \dots, N.$$

The partial pressures of component A at the inlet and outlet of each member of cascade can be determined from the partial pressure of component A in the entering gas stream  $(p'_{AN})$  and in the gas stream leaving the layer  $(p''_{A1})$  and from the number N of cascade members:  $p'_{A,j-1} = p''_{Aj} = (p'_{AN} - p''_{A1})/N$  (j = 2).



FIG. 2

Dependence of optimized parameter  $N_{opt}$  on gas velocity for  $h_0 = 47 \cdot 10^{-3} \text{ m}$ :  $\bigcirc u_L = 5.36 \cdot 10^{-3} \text{ m s}^{-1}$ ,  $\blacktriangle u_L = 8.93 \cdot 10^{-3} \text{ m s}^{-1}$ ,  $\triangle u_L = 12.5 \cdot 10^{-3} \text{ m s}^{-1}$ ; ------ the value obtained by regression

# **RESULTS AND DISCUSSION**

Figure 2 presents the dependence of the optimized parameter  $N_{opt}$  upon the gas velocity at three velocities of liquid, viz. 5.36 .  $10^{-3}$ , 8.93 .  $10^{-3}$ , and 12.5 .  $10^{-3}$  m s<sup>-1</sup> with the static bed height  $h_0 = 47$  .  $10^{-3}$ . From the figure it is obvious that the value of parameter  $N_{opt}$  is practically independent of the velocity of both gas and liquid in the interval of gas and liquid velocities investigated. The analysis of all the results obtained showed that in the investigated intervals of gas and liquid velocities and static bed heights no statistically significant dependence of the  $N_{opt}$  parameter upon these quantities could be identified. A probable cause can be the earlier-mentioned irregularity in the layer. In the gas velocity interval of 1.0 - 3.5 m s<sup>-1</sup> the value of  $N_{opt}$  parameter lies in the interval from 1.2 to 2.3, the average value being  $1.73 \pm 0.36$ . Hence at the given conditions the flow of liquid on a plate with mobile packing can be modelled by a cascade of two ideal mixers in which the relative magnitude of the second step is 73%.

A comparison with results of other authors is made difficult by the fact that in the field of modelling of liquid flow on a plate with mobile packing only one paper<sup>9</sup> adopting the model of a cascade of ideal mixers has been published so far. The analysis of the F-curves obtained from experimental measurements with a radioactive tracer (0.001% solution of <sup>24</sup>NaCl) showed that the flow of liquid in the apparatus with two plates with mobile packing (the diameter of spherical particles 19.6 .  $10^{-3}$  m, density 266 kg m<sup>-3</sup>) could be modelled appropriately by a cascade of two ideal mixers of the same magnitude with a pre-region of laminar flow. The conclusions of the paper<sup>9</sup>, which differ from those presented here in the number of cascade members per one plate, i.e. 1 member per 1 plate, are valid for the interval of gas velocities from 1.88 to 2.60 m s<sup>-1</sup> and liquid velocities from 13 .  $10^{-3}$  to 21 .  $10^{-3}$  m s<sup>-1</sup> with a constant static bed height of 0.29 m on each plate.

The values of mean residence time  $\tau_{mt}$  of liquid on the plate with mobile packing determined from the mean residence times in the individual cascade members are in average by about 2.6% (the maximum increase is ca 6%) greater than those calculated from the same experimental data with the help of the dispersion model<sup>13</sup>. Except for the arrangement with  $u_L = 5.36 \cdot 10^{-3} \text{ m s}^{-1}$  and  $h_0 = 21 \cdot 10^{-3} \text{ m}$ , where the mean residence time of liquid on plate slightly increases with increasing gas velocity, the mean residence time of liquid on plate with mobile packing shows a decrease with increasing gas velocity as well as with increasing liquid velocity. In contrast to the effect of gas and liquid velocities upon  $\tau_{mt}$ , an increase in the static bed height results in an increase in the residence time of liquid on plate. These conclusions agree with those of ref.<sup>13</sup>.

For illustration, Fig. 3 gives the dependence of mean residence time of liquid on plate for the static bed height  $h_0 = 47 \cdot 10^{-3}$  m, the parameter of lines being the liquid velocities 5.36  $\cdot 10^{-3}$ , 8.93  $\cdot 10^{-3}$ , and 12.5  $\cdot 10^{-3}$  m s<sup>-1</sup>. Table I presents the constants  $a_0$  and  $a_1$  of Eq. (5) used to fit the experimental data.

$$\tau_{\rm mt} = a_0 + a_1 u_{\rm G}, \qquad u_{\rm L} = \text{ const.} \qquad h_0 = \text{ const.} \qquad (5)$$

The mass transfer coefficients  $k_L a$  were determined for the same intervals of liquid and gas velocities and static bed heights as the parameters of the model of cascade of ideal mixers. With respect to the average value of the optimized parameter  $N_{opt}$  the coefficients were obtained by solving two equations (4) by the Newton–Raphson method, the mean residence time of liquid on plate calculated from Eq. (5) being divided into the two cascade members in such a way as to make the mean residence time of liquid in the second member equal to 73% of the  $\tau_m$  value in the first member. The mass transfer coefficients obtained by the procedure given represent a product of the mass transfer coefficient in liquid and the specific interfacial area defined as a quotient of the interfacial area and volume of liquid hold-up on plate. For obtaining,

$u_{\rm L} \cdot 10^3$ m s <sup>-1</sup>	$h_0 = 21 \cdot 10^{-3} \text{ m}$		$h_0 = 32 \cdot 10^{-3} \text{ m}$		$h_0 = 47 \cdot 10^{-3} \text{ m}$	
	$a_0$	$a_1$	$a_0$	$a_1$	$a_0$	$a_1$
5.36	5.168	0.065	6.791	-0.361	9.420	-0.327
8.93	4.647	-0.280	5.434	-0.277	8.111	-0.605
12.50	3.191	-0.133	4.570	-0.136	7.204	-0.530



FIG. 3

TABLE I

Constants  $a_0$  (s),  $a_1$  (s<sup>2</sup> m<sup>-1</sup>) of Eq. (5)

Dependence of mean residence time of liquid on plate on gas velocity for  $h_0 = 47 \cdot 10^{-3} \text{ m}$ :  $\bigcirc u_L = 5.36 \cdot 10^{-3} \text{ m s}^{-1}$ ,  $\blacktriangle u_L = 8.93 \cdot 10^{-3} \text{ m s}^{-1}$ ,  $\bigtriangleup u_L = 12.5 \cdot 10^{-3} \text{ m s}^{-1}$ 

from the  $k_L a$  coefficient, the mass transfer coefficient  $k_L a'$  in which the interfacial area is referred to the volume of contact space, i.e. the space enclosed between the plate and the mist eliminator, one must know the hold-up of liquid. This quantity can be assessed from the volume flow rate of liquid and total mean residence time of liquid. Mutual interconversions of the mass transfer coefficients in liquid can be carried out using Eq. (6) where  $z_a = 0.45$  m is the height of contact space (see ref.<sup>14</sup>)

$$k_{\rm L}a' = (k_{\rm L}a \, u_{\rm L} \, \tau_{\rm mt})/z_{\rm a} \quad . \tag{6}$$

Table II presents the values of mass transfer coefficient in liquid  $k_{\rm L}^{\rm c}a'$  calculated on the basis of the model of the cascade of ideal mixers and the value  $k_{\rm L}^{\rm d}a'$  calculated from the same experimental data by the procedure given in ref.<sup>14</sup> using the dispersion flow of liquid. The comparison of the values given in this table shows that both procedures lead to practically the same results. Also for other combinations of liquid velocities and static bed heights the deviations between the two coefficients are less than 2%.

The dispersion model of flow of liquid contains the same number of parameters as the model of the cascade of ideal mixers, i.e. 2. However, their determination (i.e. the determination of mean residence time of liquid and of the diffusion Peclet number) by

I ADLL II				
Comparison of mass	transfer coefficients	s in liquid calculated	on the basis of dispersion	model and
model of the cascade	e of ideal mixers for	$u_{\rm L} = 8.93 \ . \ 10^{-3} \ {\rm m \ s}^{-3}$	$^{-1}$ and $h_0 = 32 \cdot 10^{-3}$ m	

<i>u</i> <sub>G</sub>	$k_{\rm L}^{\rm c}a'$ . 10 <sup>2</sup>	$k_{\rm L}^{\rm d}a'$ . 10 <sup>2</sup>	$\delta^a$
$m s^{-1}$	$m s^{-1}$	$m s^{-1}$	%
1.0	2 020	2.025	_0.25
1.0	1 992	1 998	-0.25
1.0	2.175	2 180	-0.23
1.5	2.166	2.172	-0.28
2.0	2.545	2.546	-0.04
2.0	2.543	2.544	-0.04
2.5	2.971	2.962	0.30
2.5	2.894	2.888	0.21
3.0	3.193	3.178	0.47
3.0	3.212	3.196	0.50
3.5	3.514	3.486	0.80
3.5	3.482	3.455	0.78

 $^a$   $\delta = (k_{\rm L}^{\rm c}a'-k_{\rm L}^{\rm d}a')/k_{\rm L}^{\rm c}a'$  . 100 .

TABLE II

the transient response technique with a general input signal is more complex than the determination of parameters of the model of the cascade of ideal mixers because a partial differential equation must be solved. Also the calculation proper of the mass transfer coefficient in liquid considering the dispersion flow necessitates solving a more complex nonlinear equation (see e.g. ref.<sup>14</sup>). Hence from this point of view it is more suitable to determine the basic characteristics of mass transfer in an apparatus with mobile packing with the help of the model of the cascade of ideal mixers.

The dependence of the mass transfer coefficient in liquid upon the gas velocity at constant velocity of liquid and constant static bed height is linear, the conclusions about the effects of other quantities on the mass transfer coefficient being identical with those published<sup>14</sup>. For illustration, Fig. 4 presents the dependences  $k_{\rm L}a' = f(u_{\rm G})$  for  $h_0 = 32 \cdot 10^{-3}$  m; the straight lines given were obtained by the linear regression of the data sets for  $u_{\rm L} = \text{const}$  and  $h_0 = \text{const}$ .



Fig. 4

Dependence of mass transfer coefficient in liquid on gas velocity for  $h_0 = 32 \cdot 10^{-3} \text{ m}$ :  $\bigcirc u_L = 5.36 \cdot 10^{-3} \text{ m s}^{-1}$ ,  $\blacktriangle u_L = 8.93 \cdot 10^{-3} \text{ m s}^{-1}$ ,  $\bigtriangleup u_L = 12.5 \cdot 10^{-3} \text{ m s}^{-1}$ 

## SYMBOLS

- *a* specific interfacial area (referred to the volume of hold-up),  $m^{-1}$
- $a_0$  constant in Eq. (5), s
- $a_1$  constant in Eq. (5), s<sup>2</sup> m<sup>-1</sup>
- a' specific interfacial area (referred to the volume of absorption zone), m<sup>-1</sup>
- c molar concentration of tracer or desorbed component, kmol m<sup>-3</sup>
- *H* Henry's constant, kmol  $m^{-3} Pa^{-1}$
- $h_0$  static bed height, m
- $k_{\rm L}$  mass transfer coefficient in liquid, m s<sup>-1</sup>
- *N* number of cascade members
- *p* partial pressure, Pa

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и	velocity, m s <sup>-1</sup>
V	volume, m <sup>3</sup>
$V_{\rm t}$	volume of layer, m <sup>3</sup>
V	volume flow rate, m <sup>3</sup> s <sup>-1</sup>
δ	deviation, %
ε <sub>L</sub>	dimensionless hold-up of liquid in layer
τ	time, s
τ <sub>m</sub>	mean residence time of liquid in each cascade member, s

mean residence time of liquid on plate,

$$\tau_{\rm mt} = \sum_{j=1}^N \tau_{\rm mj} \,, \,\, {\rm s}$$

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 $\tau_{\text{mt}}$ 

Α	referred to component A (CO <sub>2</sub> )
c	referred to the model of cascade of ideal mixers
calc	calculated value
d	referred to the dispersion model
exp	experimental value
G	referred to gas
L	referred to liquid
t	total
0	referred to inlet into the first cascade member
,	inlet of gas into the <i>j</i> -th cascade member
"	outlet of gas from the <i>j</i> -th cascade member

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