EFFECT OF LIQUID AXIAL MIXING ON LOCAL k_La VALUES IN INDIVIDUAL STAGES OF MULTIPLE-IMPELLER VESSEL

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The effect of the liquid phase axial mixing on "local" k_La values, measured by dynamic pressure method (DPM), in individual stages of multiple-impeller vessel is evaluated. The data measured by the DPM in our previous papers are corrected for this effect. Various calculation methods for k_La in the upper stages based on single-impeller vessel correlations are compared.

Key words: Mixing; Mass transfer; Axial dispersion; Multiple impeller.

The mixing power dissipated in individual stages of multiple-impeller vessel is very nonuniform as a result of different gas volumes passed through the respective impeller regions. Each stage is associated with an agitator in the vessel. The mass transfer correlations suggested in literature do not reflect the nonuniformity in spite of the fact that the differences in the power dissipated in individual stages reach 50% (see, *e.g.*, refs[1,2]). They give a single k_La value for the whole device. These values are usually measured[3,4] in the middle of the vessel height and are correlated with the mean power dissipated in the vessel so that they have no relation to the local values in individual stages. Parameters of the correlations are scale-dependent and cannot be applied to devices of different sizes without a risk.

The main reason why the inhomogeneous character of dispersions was not considered consisted in the fact that a method of the local k_La values measurement in individual stages was lacking. Such a method has recently been presented by Moucha *et al.*[5]. It is the dynamic pressure method (DPM) which employs a small change in the total pressure in the system to generate the concentration change simultaneously in the whole dispersed gas. This special way of introducing the concentration disturbance into the system suppresses the effect of the gas phase axial mixing on the coefficient measured. As a result, the evaluation of k_La using a simple mass transfer model with ideal mixing in the gas phase leads to reasonably small differences from the real values characteristic for individual stages as shown by Linek and Sinkule[6]. As it will be shown below, this is not the case of the effect of the liquid phase axial mixing which have to be taken into account.

The values measured by the DPM in individual stages of the vessel of inner diameter $T = 0.19$ m stirred with four Rushton turbines led us[5] to the conclusion that the interfacial mass transfer in multiple-impeller vessels with the same agitators on a common shaft can be described by two k_La values directly measured by the DPM: one for the bottom stage (associated with bottom agitator) and the other for all the upper stages. In the upper stages, the values were up to 1.4 times higher than those in the bottom stage. Differences between the values in the upper stages were considerably smaller and they were regarded as a scatter of the experimental data. Further experiments have shown, however, that the differences in k_La in upper stages are systematic. This is due to the axial dispersion of liquid from the bottom stage, which is less saturated with oxygen than the liquid in the upper stages owing to lower k_La in the bottom stage. This is not taken into account in the DPM evaluation as the responses of oxygen probes in individual stages are evaluated separately using a simple mass transfer model with ideal mixing in both phases. This error distorted the k_La values presented in our earlier papers[5,7] and also the conclusions deduced from them[8].

The aim of this paper is to re-evaluate the k_La data measured previously[5,7] by the DPM in individual stages of four-impeller vessel of inner diameter $T = 0.19$ m. The re-evaluation will be performed by a new procedure which takes the axial mixing of the liquid phase into account. The recalculation method for k_La in upper stages from correlations obtained in a single-impeller vessel is proposed which is based on the re-evalued k_La data.

THEORETICAL

Gas–Liquid Mass Transfer Model

A model of plug flow with axial dispersion D_E in the gas and a cascade of N perfectly mixed stages with exchange flows Q between adjacent cells for the liquid was used to describe the gas–liquid mass transfer in the vessel with N impellers. The component balances and total balance equations for gas are ($i = O_2$ or N_2, k denotes stage numbered from the bottom)

$$\varepsilon_G \frac{\partial p_{Gi}}{\partial t} + \frac{\partial (v_G p_{Gi})}{\partial l} - \varepsilon_G D_E \frac{\partial^2 p_{Gi}}{\partial l^2} + m_i (1 - \varepsilon_G)(k_L a_k)_i \, (p_{Gi} - p_{Li}) = 0 \qquad (1)$$

$$P(t)\frac{\partial v_G}{\partial l} + \varepsilon_G \frac{dP(t)}{dt} + (1 - \varepsilon_G)(k_L a_k)_{O_2}[m_{O_2}(p_{GO_2} - p_{LO_2}) + m_{N_2} A (p_{GN_2} - p_{LN_2})] = 0 \qquad (2)$$

with boundary and initial conditions

$$
\begin{gathered}
l = 0: \quad v_G p_{Gi} - \varepsilon_G D_E \frac{\partial p_{Gi}}{\partial l} = p(\infty) z_i v_{G0} \\
v_G = v_{G0} P(\infty)/P(t) \\
l = H: \quad \frac{\partial p_{Gi}}{\partial l} = 0 \\
t = 0: \quad p_{Gi} = P(0) z_i \ .
\end{gathered} \tag{3}
$$

The partial pressure of the gas components meets the condition:

$$p_{GO_2} + p_{GN_2} = P(t) \ . \tag{4}$$

The balance equations for component *i* in the liquid are

$$Q(p_{Li,k+1} - 2p_{Li,k} + p_{Li,k-1}) + (k_L a_k)_i V_L (\overline{p_{Gi,k}} - p_{Li,k}) = V_L \frac{dp_{Li,k}}{dt} \tag{5}$$

with initial condition

$$t = 0: \quad p_{Li,k} = P(0) z_i \ , \tag{6}$$

where $k = 1, 2, \ldots, N$ and $p_{Li,0} = p_{Li,1}$, $p_{Li,N+1} = p_{Li,N}$. The average partial pressure of component *i* in the stage *k* is defined as

$$\overline{p_{Gi,k}} = \frac{N}{H} \int_{\frac{k-1}{N}H}^{\frac{k}{N}H} p_{Gi} \, dl \ , \quad k = 1, 2, \ldots, N \ . \tag{7}$$

The nitrogen mass transfer coefficient $(k_L a_k)_{N_2}$ was eliminated using the following relation

$$\frac{(k_L a_k)_{N_2}}{(k_L a_k)_{O_2}} = \left(\frac{D_{N_2}}{D_{O_2}} \right)^{1/2} = A \tag{8}$$

which follows from the penetration theory. In further text, the oxygen mass transfer coefficient is used only, and the subscript O_2 is omitted (*i.e.*, $k_L a_k = (k_L a_k)_{O_2}$.

Oxygen mass transfer coefficient in the bottom stage $k_L a_1$ differs from that in the upper stages $k_L a_{2-N}$

$$k_L a_k = \begin{cases} k_L a_1 & \text{for } k = 1 \text{ or } 1 < H/N \\ k_L a_{2-N} & \text{for } k > 1 \text{ or } 1 > H/N \ . \end{cases} \tag{9}$$

Modelling of the Liquid Phase Axial Mixing Effect on k_La Measured by DPM

The effect of the liquid axial mixing was evaluated as follows: Normalized oxygen concentration-time profiles in individual stages (= $[p_{LO_2,k}(t)/z_{O_2} - P(0)]/[P(\infty) - P(0)]$) were calculated from Eqs (*1*) to (*9*) as responses to the step change in total pressure from the value $P(0)$ to $P(\infty)$. The profiles were calculated for various values of parameters ν_{G0}, V_L, z_i, m_i, A, D_E, ε_{Gk}, Q, k_La_1, k_La_{2-4}. They were taken the same as in the experiments performed previously[5,7] in the vessel agitated with four Rushton turbines. Volumetric mass transfer coefficients $k_La_k^{DPM}$ were calculated from the profiles by the same evaluation method as previously[5,7] applied on real experimental profiles. The method, being described in detail elsewhere[9], consists in fitting the time profiles by the simple mass transfer model with ideal mixing in both phases. For example, this model reduces to simple exponential for pure oxygen absorption (z_{O_2} = 1)

$$\frac{p_{LO_2,k}(t) - P(0)}{P(\infty) - P(0)} = 1 - \exp\,(-k_La_k^{DPM}t)\ . \qquad (10)$$

The differences between the calculated values k_La^{DPM} and the values k_La used in the time-profile calculations (Eqs (*1*) to (*9*)) are just due to the axial mixing in both phases. Axial dispersion in the liquid phase is characterized by the exchange flux Q and in the gas phase by the axial dispersion coefficient D_E. For illustration, the differences for the bottom stage ($k_La_1^{DPM} - k_La_1$) are plotted in Fig. 1 as a function of the time constant ratio: the time constant of the interfacial mass transfer ($1/k_La$) and that of the flushing

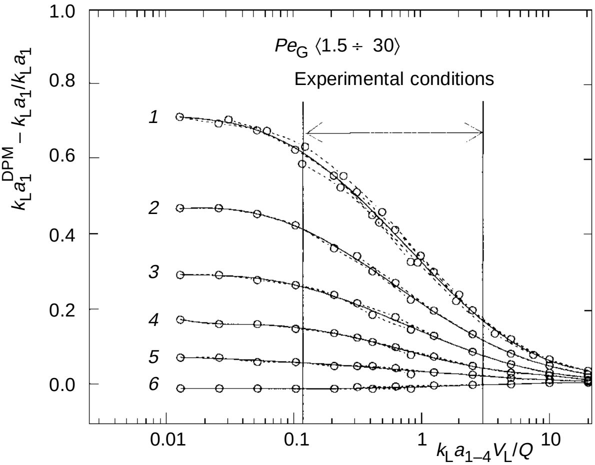

FIG. 1
Effect of liquid axial mixing on the local values $k_La_1^{DPM}$ measured by the dynamic pressure method in the bottom stage. m Numerical simulation for different D_E values. k_La_1/k_La_{2-4} = 0.5 (1); 0.6 (*2*); 0.7 (*3*); 0.8 (*4*); 0.9 (*5*), 1.0 (*6*)

out of the liquid in the stage by the exchange flow (V_L/Q). The differences are nearly independent of the mixing of gas (see Fig. 1, points calculated for Peclet numbers from 1.5 to 30) but they strongly depend on the liquid axial mixing. For example, if the flushing out is five times more rapid than the saturation of the batch ($k_La_{1-4}V_L/Q = 0.2$), the $k_La_1^{DPM}$ can be overestimated by 60% compared with the true local value k_La_1. If the flushing is five times slower than the saturation ($k_La_{1-4}V_L/Q = 5$), the overestimation of $k_La_1^{DPM}$ is lower and makes only 10%.

A similar figure can be drawn for the differences in upper stages ($k_La_2^{DPM} - k_La_2$) which are lower and of opposite sign than that for the bottom stage.

New Method of the Experimental Data Treatment to Account for Axial Mixing in the Liquid Phase

An attempt was made to determine k_La_1 and k_La_{2-4} simultaneously by fitting the calculated liquid concentrations in individual stages from model (*1*) to (*9*) to experimental ones, keeping for the hydrodynamic parameters (D_E and Q) values obtained by independent tracer experiments[7]. The simultaneous determination of the two parameters was unsuccessful as they are interdependent. This is illustrated in Table I where the results of the evaluation of some experiments are presented. The values of k_La_1 are unreasonably low or even negative being counterbalanced by the high values of k_La_{2-4}. The simultaneous determination of the local parameters is thus impossible. From Table I, we can see, however, that the average k_La_{1-4} and $(k_La_{1-4})^{DPM}$ values are roughly the same. Further analysis of the results has shown that the mean values k_La_{1-4} of the simultaneously evaluated k_La_i values differ only slightly from the mean values $(k_La_{1-4})^{DPM}$ of the separately evaluated $k_La_i^{DPM}$ (using the model of perfectly mixed phases) in the whole extent of experimental conditions (see Fig. 2). This finding is expressed by the following expression:

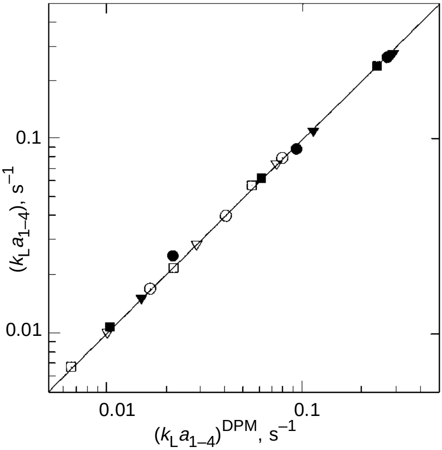

FIG. 2

Comparison of averages of the k_La local values evaluated by the simultaneous determination, k_La_{1-4}, with the averages evaluated by the dynamic pressure method $(k_La_{1-4})^{DPM}$. v_{G0}, mm s^{-1}: 0.5 M Na_2SO_4: ■ 2.12, ▼ 4.24, ● 8.48; water: ❐ 2.12, ∇ 4.24, ❍ 8.48

$$\frac{1}{N}\sum_{k=1}^{N} k_\mathrm{L}a_k \equiv k_\mathrm{L}a_{1-N} = (k_\mathrm{L}a_{1-N})^\mathrm{DPM} \equiv \frac{1}{N}\sum_{k=1}^{N} k_\mathrm{L}a_k^\mathrm{DPM} \quad . \qquad (11)$$

The new method of evaluation of corrected local values $k_\mathrm{L}a_1$ and $k_\mathrm{L}a_{2-N}$ from the values $k_\mathrm{L}a_i^\mathrm{DPM}$ is based on this relation (*11*). The main assumption of the method is that the values $k_\mathrm{L}a_1^\mathrm{DPM}$ determined in a single-impeller vessel filled with liquid to the height equal to the vessel diameter ($L/T = 1$) are the true local values in the bottom stage even in a multiple-impeller vessel, *i.e*, $k_\mathrm{L}a_1 = k_\mathrm{L}a_1^\mathrm{DPM}$ at $L/T = 1$. The values in the upper stages $k_\mathrm{L}a_{2-N}$, are calculated from the mean value $(k_\mathrm{L}a_{1-N})^\mathrm{DPM}$ using Eq. (*11*) in the form

$$k_\mathrm{L}a_{2-N} = \frac{N(k_\mathrm{L}a_{1-N})^\mathrm{DPM} - k_\mathrm{L}a_1^\mathrm{DPM}(L/T = 1)}{N-1} \quad . \qquad (12)$$

TABLE I
Comparison of the simultaneously fitted values ($k_\mathrm{L}a_1$, $k_\mathrm{L}a_{2-N}$), the DPM-measured values ($k_\mathrm{L}a_1^\mathrm{DPM}$) and the mean values ($(k_\mathrm{L}a_{1-4})^\mathrm{DPM}$, $k_\mathrm{L}a_{1-4}$). The data measured at $f = 5.5\ \mathrm{s}^{-1}$ in 0.5 M Na_2SO_4 solution

k	$k_\mathrm{L}a_k^\mathrm{DPM}$, $10^{-3}\ \mathrm{s}^{-1}$	k	$k_\mathrm{L}a_k$, $10^{-3}\ \mathrm{s}^{-1}$
	$v_{G0} = 2.12\ \mathrm{mm\ s}^{-1}$		
1	7.5	1	–3.17
2	10.2	2–4	15.5
3	10.4		
4	13.3		
1–4	10.4	1–4	10.8
	$v_{G0} = 4.24\ \mathrm{mm\ s}^{-1}$		
1	12.7	1	0.308
2	14.7	2–4	20.1
3	15.0		
4	17.5		
1–4	15.0	1–4	15.1
	$v_{G0} = 8.48\ \mathrm{mm\ s}^{-1}$		
1	12.8	1	–10.0
2	18.1	2–4	33.0
3	24.9		
4	30.9		
1–4	21.7	1–4	22.3

The mean value $(k_L a_{1-N})^{DPM}$ is calculated from separately evaluated values $k_L a_i^{DPM}$ (i = 1, 2, …, N).

RESULTS AND DISCUSSION

In our previous papers[5,8], methods of $k_L a$ evaluation in individual stages of multiple-impeller vessels were presented. The methods are based on correlations derived from single-impeller data. For obvious reasons, single-impeller data are more frequent in the literature than multiple-impleller ones. Linek *et al.*[8] proposed a correlation in which the total specific power dissipated in stages was used, *i.e.*, the sum of that due to the agitator and the power dissipated by rising bubbles of aeration gas. The correlation fitted to the corrected $k_L a$ data from this work resulted in the following forms:

$$k_L a_k = 3.79 \;.\; 10^{-3}\;\; (e_{k,\mathrm{agit}} + v_{G0}\rho_L g)^{0.706}\,(v_{Gk})^{0.443}$$

$$(k = 1,\; 2\text{–}N) \qquad (13)$$

for coalescing batch (water) and

$$k_L a_k = 1.11 \;.\; 10^{-4}\;\; (e_{k,\mathrm{agit}} + v_{G0}\rho_L g)^{1.34}\,(v_{Gk})^{0.356}$$

$$(k = 1,\; 2\text{–}N) \qquad (14)$$

for noncoalescing batch (0.5 M Na_2SO_4), where v_{Gk} is the superficial velocity of the gas flowing through the impeller region in the stage k. In the bottom stage, it is the superficial velocity of the input gas v_{G0} if the gas is introduced below the bottom agitator and enters its mixing region practically in full amount. In the upper stages, the superficial gas velocity in the impeller region v_{G2-4} was estimated (Moucha *et al.*[5]) from the power difference between the upper and bottom agitators following the concept of Lu and Yao[2].

Figure 3 shows that the mean deviations of corrected experimental and calculated $k_L a_i$ values are even greater (–18.5 and –20% for water and 0.5 M Na_2SO_4, respectively) than the mean deviations of the original uncorrected $k_L a_i^{DPM}$ data (–20 and –5%, see Linek *et al.*[8], Fig. 1). In Figs 3 and 4, k = 2–2 and k = 2–3 represents the $k_L a$ data measured in upper sections in the vessel with two and three impellers, respectively. The deviations are consequencies of an improper prediction of $k_L a_{2-N}$ data from the correlation based on the single-impeller data. A better fit of the $k_L a_{2-N}$ data was obtained with the correlation which includes a term proportional to the part of gas passed through the impeller regions. This term is a relative decrease in the gassed and ungassed power dissipated by the impeller, $e_{k,\mathrm{agit}}/e_0$. The correlation resulted in the form

$$k_\mathrm{L}a_k = 5.56\,.\,10^{-4}\ (e_{k,\mathrm{agit}} + v_{G0}\rho_\mathrm{L}g)^{0.73} \left(\frac{e_{k,\mathrm{agit}}}{e_0}\right)^{-0.653} (v_{G0})^{0.202} \quad (k = 1,\ 2\text{–}N) \qquad (15)$$

for coalescing batch (water) and

$$k_\mathrm{L}a_k = 4.96\,.\,10^{-5}\ (e_{k,\mathrm{agit}} + v_{G0}\rho_\mathrm{L}g)^{1.34} \left(\frac{e_{k,\mathrm{agit}}}{e_0}\right)^{-0.351} (v_{G0})^{0.251} \quad (k = 1,\ 2\text{–}N) \qquad (16)$$

for noncoalescing batch (0.5 M Na_2SO_4). The correlations were evaluated by fitting the experimental data, $k_\mathrm{L}a_1^\mathrm{DPM}$, measured by the dynamic pressure method in a single-impeller vessel at *L/T* = 1. The values of gassed power dissipated by agitators in upper sections $e_{2-N,\mathrm{agit}}$ were calculated from the correlations presented in a previous paper[7].

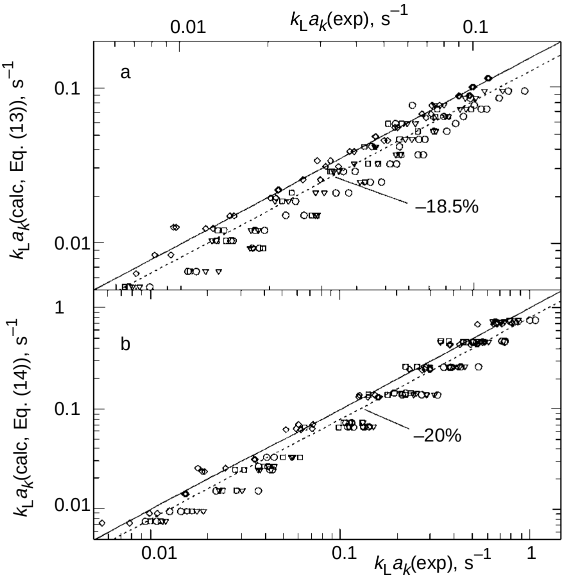

FIG. 3

Comparison of experimental $k_\mathrm{L}a$ values with those calculated from correlations (*13*) and (*14*) in all stages. a Water, standard deviation 18%; b 0.5 M Na_2SO_4, standard deviation 22%; *k*: ✧ 1, ❍ 2–2, ∇ 2–3, ❒ 2–4

The ungassed power of turbine agitators e_0 was calculated from the power number $P_0 = 4.6$ measured by Linek *et al.*[7]. Mean differences between k_La_i values calculated from the correlations and those measured experimentally as well as standard deviations are given in Fig. 4.

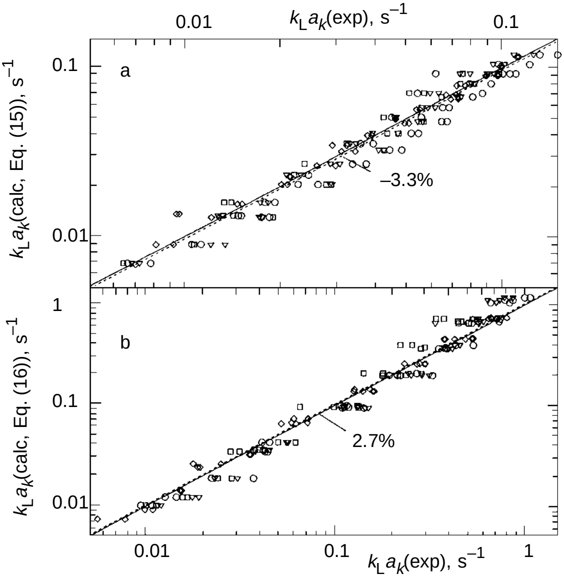

FIG. 4

Comparison of experimental k_La values with those calculated from correlations (*15*) and (*16*) in all stages. a Water, standard deviation 15%; b 0.5 M Na_2SO_4, standard deviation 28%; *k*: ✧ 1, ❍ 2–2, ∇ 2–3, ❐ 2–4

CONCLUSION

Axial mixing of liquid in multiple-impeller vessels affects significantly the results of the DPM measurements, which vary from the k_La values characteristic for individual stages. A new method of the DPM data treatment, taking into account axial mixing of the liquid phase, is based on Eq. (*11*). Correlations of the coefficients measured in a single-impeller vessel are presented, which can be used even for calculation of the coefficients in the upper stages with sufficient accuracy. The procedure presented here is now being verified using the data measured in larger vessel (T = 0.29 m, so far unpublished). If the verification is succesfull, it will be possible to predict seriously an average coefficient k_La_{1-N} for multiple-impeller vessels using the mass transfer data measured in single-impeller vessels only.

SYMBOLS

A	nitrogen to oxygen mass transfer coefficient ratio (see Eq. (*5*))
D_E	coefficient of axial dispersion, $m^2 s^{-1}$
e_0	ungassed power dissipated by agitator, $W m^{-3}$
$e_{k,agit}$	gassed specific power dissipated by agitator in stage k, $W m^{-3}$
f	agitator frequency, s^{-1}
g	acceleration due to gravity, $m s^{-2}$
H	dispersion height, m
k_La_k	volumetric oxygen mass transfer coefficient per unit volume of liquid in stage k, s^{-1}
k_La_{2-N}	average volumetric oxygen mass transfer coefficient per unit volume of liquid in upper stages, s^{-1}
$k_La_k^{DPM}$	volumetric oxygen mass transfer coefficient per unit volume of liquid in stage k, measured by DPM, s^{-1}
l	vertical coordinate, m
L	liquid height in vessel, m
m_i	distribution coefficient of component i
N	number of impellers on shaft
p_{Gi}, p_{Li}	partial pressure of component i in gas and liquid, Pa
Pe_G	Peclet number, $Pe_G = Hv_{G0}/(D_E\varepsilon_G)$
$P(t)$	total pressure in vessel, Pa
$P(\infty)$	final steady-state total pressure in vessel, Pa
$P(0)$	initial steady-state total pressure in vessel, Pa
Q	exchange flow rate, $m^3 s^{-1}$
t	time, s
T	vessel diameter, m
v_{Gk}	superficial velocity of the gas flowing through the impeller in stage k, $m s^{-1}$
v_{G0}	superficial velocity of input gas, $m s^{-1}$
V_L	volume of liquid in each stage, m^3
z_i	volume fraction of component i input gas ($z_{O_2} = 0.21$, $z_{N_2} = 0.79$)
ε_G	gas holdup
ρ_L	liquid batch density, $kg m^{-3}$

Subscripts

i	value related to oxygen ($i = O_2$) or nitrogen ($i = N_2$)
k	value related to the k-th stage of a vessel, numbered from bottom

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REFERENCES

1. Nocentini M., Magelli F., Pasquali G., Fajner D.: *Chem. Eng. J*. **1988**, *37*, 53.
2. Lu Wei-Ming, Yao Chia-Lin: *J. Chin. Inst. Chem. Eng*. **1993**, *23*, 173.
3. Machon V., Vlcek J., Hudcova V.: *Proceedings of the 6th European Conference on Mixing, Pavia 24–26 May 1988 (BHRA Cranfield),* p. 351. AIDIC, Bedford, U.K. 1988.
4. Nocentini M., Fajner D., Pasquali G., Magelli F.: *Ind. Eng. Chem. Res*. **1993**, *32*, 19.
5. Moucha T., Linek V., Sinkule J.: *Trans. Inst. Chem. Eng*. **1995**, *73*, 286.
6. Linek V., Sinkule J.: *Trans. Inst. Chem. Eng*. **1991**, *69*, 308.

7. Linek V., Moucha T., Sinkule J.: *Chem. Eng. Sci*. **1996**, *51*, 3203.
8. Linek V., Moucha T., Sinkule J.: *Chem. Eng. Sci*. **1996**, *51*, 3875.
9. Linek V., Benes P., Sinkule J., Moucha T.: *Chem. Eng. Sci*. **1993**, *48*, 1593.