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SCIENTIFIC PAPER

UDC 66.023+621.929.4:66.021.3+531.73

MASS TRANSFER IN A MULTIPHASE VIBRATION COLUMN. I – THE VOLUMETRIC MASS TRANSFER COEFFICIENT

The results of the analysis of the mass transfer characteristics of a reciprocating plate column (RPC) of the Karr type in the case of two-phase (gas-liquid) and three-phase systems (gas-liquid-solid) are presented. The volumetric mass transfer coefficient in the liquid phase ($k_{L,a}$) was analyzed and the obtained results indicated that $k_{L,a}$ depends on the vibration intensity, the superficial gas velocity and in the case of three-phase system, on the content of the solid phase present in the RPC. An empirical correlation was derived indicating that $k_{L,a}$ does not depend on the column diameter and that for its calculation only the average power consumption in the column and the superficial gas velocity must be known.

Many investigations of the characteristics of different two- and three-phase column type reactors have been performed in the past with goal of decreasing the energy consumption and increasing the mass transfer rates in such systems, i.e. intensifying the volumetric mass transfer coefficient, gas hold-up and interfacial area. The reciprocating plate column (RPC) represents such a column type reactor, and in the case of a gas-liquid system it includes some advantages of a bubble column (BC). However, contrast to the BC, the mixing of the phases in a RPC is realized, not only by gas flow through the column, but more by vibration of an axially mounted set of perforated plates. So, in such a gas-liquid column a large specific interfacial area could be created [1,2] and, according to some experimental investigations, a uniform distribution of the bubbles. Also, a larger gas hold-up and smaller backmixing in a RPC, compared to a bubble column with or without the liquid circulation [3-5] or to the stirred tank reactor (ST), could easily be established. A RPC represents a new generation bioreactor according to its good hydrodynamic characteristics and thus the very effective oxygen mass transfer rate from the gas to the liquid phase [6].

The volumetric mass transfer coefficient in a RPC depends on the vibration intensity, the superficial velocities of the gas and liquid phase, the column geometry and on the physical characteristics of the liquid phase. An increase of the mass transfer could also be obtained by introducing solid particles in the column thus realizing a three-phase system RPC.

THE INFLUENCE OF OPERATING CONDITIONS

The volumetric mass transfer coefficient ($k_{L,a}$) could be easily increased at constant gas flow rate by

increasing the vibration intensity (defined as the product of the amplitude and frequency of vibration) [7-11]. Such an effect is caused by the bubble breaking when the mixing intensity determines the mass transfer rate between the gas and liquid phase. There is only a minor increase in the case of low vibration intensity depending on the aeration intensity, i.e. on the gas flow rate [5, 12, 13], but it could be very important if the vibration intensity is greater than 3 cm/s [11] or 5 cm/s [9,10].

By increasing the gas flow rate, at some constant vibration intensity, the volumetric mass transfer coefficient also increases, achieving some final value on the basis of the energy consumption used for making a fine bubble dispersion. The gas hold-up is increased in such cases and bubble breaking is intensified during bubble movement through the holes of the vibration plates.

Different results could be found in the literature regarding the influence of the liquid flow rate on the mass transfer coefficient. Some of them indicated that such an influence does not exist [10] or that it is negligible [13], while others have shown that $k_{L,a}$ increases by increasing the liquid flow rate [7,11]. It seems that the influence of the liquid flow rate on $k_{L,a}$ depends on the geometry of the vibration mixer and liquid flow velocity, as a consequence of the turbulent motion of the liquid phase created by the liquid flow through the holes of the perforated plates.

The arrangement of the gas and liquid flows (counter or co-current flow) seems to influence the mass transfer rate. A larger value of the mass transfer coefficient could be obtained in the case of co-current compared to the counter-current flow of the gas and liquid phases [10].

INFLUENCE OF THE PHYSICAL CHARACTERISTICS OF THE LIQUID PHASE

Such an influence is very complex, because the liquid characteristics may change both the mass transfer

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Paper received: May 30, 2001
Paper accepted: August 20, 2001

in the liquid phase (k_L) and the interfacial area (a). The addition of non-electrolytes to the liquid phase, such as for example an alcohol, only in a small amount, might cause an increase of the volumetric mass transfer coefficient ($k_L a$) [14]. The results of some other investigations have shown that the increase of the volumetric mass transfer coefficient ($k_L a$) could be correlated to a change (increase) of the dimensionless parameter Z , which includes the physical characteristics of the liquid phase [15]:

$$Z = (\rho_L \cdot \sigma^3) / (g \cdot \mu^4) \quad (1)$$

The $k_L a$ decreases with increasing liquid viscosity in the case of different liquids with similar density and surface tension. Thus, in the case of sucrose solution, the volumetric mass transfer coefficient decreases with increasing sucrose concentration as a result of the viscosity increase, while at the same time the average bubble diameter and interfacial area are negligibly changed [5].

THE INFLUENCE OF COLUMN GEOMETRY

The volumetric mass transfer coefficient increases by increasing the number of perforated plates (i.e. decrease of the distance between the plates), at a constant value of aeration and mixing intensity [13]. A larger number of perforated plates means greater energy dissipation, thus suppressing bubble coalescence and an increase of the interfacial gas-liquid area [8]. An increase of the hole diameter and plate free area decreases the volumetric mass transfer coefficient [8,10].

THE INFLUENCE OF THE PRESENCE OF SOLID PHASE IN THE COLUMN

By adding Rashig rings made from glass in each space between two plates (total amount 2.5% by volume) the volumetric mass transfer coefficient is increased by 30%. Such an effect is caused by an increase of the bubble breaking process resulting in a fine gas-liquid dispersion [10]. The influence of some other types of solid particles has not yet been investigated.

CORRELATION FOR THE CALCULATION OF THE VOLUMETRIC MASS TRANSFER COEFFICIENT

The volumetric mass transfer coefficient was correlated by the vibration intensity, i.e. by the power consumption in a RPC and the superficial gas velocity (Table 1 give a summary of the data presented in the scientific literature). In the case of a RPC, with a larger hole diameter of the perforated plates and a large free sectional area of the plate, and when the liquid phase in the column is arranged as a charge, relatively ineffective usage of the external power was detected [7]. Some other investigators also pointed out a larger influence of

the superficial gas velocity on the volumetric mass transfer coefficient compared to the influence of externally used power for mixing the gas-liquid dispersion [5,12,16]. The values of the exponent shown in Table 1, in different correlation equations, are in the range of 0.25–0.74 in the case of the superficial gas velocity and 0.44–1.55 for power consumption. The volumetric mass transfer coefficient mostly depends on the superficial gas velocity when small vibration intensities are applied. On the other hand, at a higher vibration intensity, $k_L a$ mostly depends on the power consumption if a smaller hole diameter of the perforated plate is used or mixing and aeration have the practically the same influence [8,12]. The correlation for the volumetric mass transfer coefficient in a RPC with small hole diameters and a smaller free sectional area of the perforated plates [8] has an almost identical form as the correlation derived for a vessel with a turbine impeller [16,17].

COMPARISON WITH DIFFERENT REACTOR TYPES

A higher mass transfer rate could be obtained in a two-phase RPC than in a classical bubble column or bubble column with stationary plates [18]. Comparison with a bubble column or stirred tank reactor showed that a RPC had the highest aeration capacity ($k_L a$) at the same specific power of mixing and at a lower superficial gas velocity [5,16].

In this paper the influence of different operating conditions on the mass transfer of oxygen in a RPC column of 2.54 and 9.2 cm in diameter was investigated. The goal of the investigation was to identify the relationship between the $k_L a$ and vibration intensity, superficial gas velocity and content of the solid phase in a column. The main focus in the present investigation was to derive a correlation, which enable $k_L a$ calculation independently of the column geometry and of the characteristics of the liquid phase. A derived correlation is always useful for making comparisons of the mass transfer characteristics between RPC columns (two- and three-phase system) and RPC and other reactor types operating under the same conditions.

EXPERIMENTAL

The experimental set-up unit (Figure 1) is made of perforated plates mounted on one axis as a vibration set. The plates are spaced at a specific and equal distance at the axis. The experiments were performed in a column of 2.54 and 9.2 cm diameter, while other important geometrical characteristics of the unit, as well as the range of operating parameters (conditions) are given in Table 2. Polypropylene spheres (density of 930 kg/m³) were used as the solid phase. The diameter of the sphere was 8.3 ± 0.1 mm and the total content of solid phase 3.82 and 6.61% (by volume). This solid

Table 1. The correlation of $k_L a$ in a two – phase gas – liquid RPC

Liquid flow	D_c cm	n_p	l_c m	d_o mm	ϵ	Operating cond. (u_g, u_l - cm/s; P_g/V_l - W/m ³)	Method	Correlation	References
Charge	2.54	65	2	7	0.41	$u_g < 2.5$ cm/s ili $u_g < 5.0$ cm/s	Chemical Physical Dynamic (water-O ₂)	$k_L a = 0,467(P_g^*)^{0,25} u_g^{0,6}$	[5, 13]
↑↓	2.54	33	2	8	0.51				
↑↑	5.08	84	3.96	15	0.53	$0.72 < u_g < 4.98$ $u_l > 2$	Physical (water-O ₂)	$k_L a = 0,00401(Af)^{0,18} u_g^{0,42} u_l^{0,918}$	[11]
Charge	5.08	54	2	14	0.57	$0.49 < u_g < 0.99$ $23 \leq \frac{P_g}{V_l} \leq 3620$ $0.99 < u_l < 3.95$	Physical (water-O ₂)	$k_L a = 4,86 \left(\frac{P_g}{V_l}\right)^{0,364} u_g^{1,55}$	[7]
↑↓								$k_L a = 3,90 \left(\frac{P_g}{V_l}\right)^{0,246} u_g^{0,623} u_l^{0,825}$	
↑↑	9.3	10	0.56	3- 65	0.09- 0.306	$0.6 < u_g < 4.6$ $0.41 < u_l < 3.68$	Physical (water-O ₂)	$Af < 5$ cm/s (segregated regime): $k_L a = 0,0033(Af)^{0,1} u_g^{0,25} d_o^{-0,2} \epsilon^{-0,5}$ $Af \geq 5$ cm/s (homogeneous regime): $k_L a = 0,0033(Af)^{0,18} u_g^{0,2} d_o^{-0,12} \epsilon^{-0,52}$	[10]
Charge	10.16	18	1.26	6.35	0.28	$0.68 < u_g < 1.86$ $290 \leq \frac{P_g}{V_l} \leq 7200$	Physical (water-O ₂)	$k_L a = 0,00723 \left(\frac{P_g}{V_l}\right)^{0,59} u_g^{0,44}$	[8]
Charge	20.6	18	3.85	6.35	0.33	$0,05 \leq u_g \leq 0,40$ $35 \leq \left(\frac{P_g}{V_l}\right) \leq 6600$	Physical (water-O ₂)	$k_L a = 0,281 \left(\frac{P_g}{V_l}\right)^{0,632} u_g^{1,138}$ Small vibration intensity: $k_L a = 1,677 \left(\frac{P_g}{V_l}\right)^{0,296} u_g^{1,142}$ Large vibration intensity: $k_L a = 0,0196 \left(\frac{P_g}{V_l}\right)^{1,017} u_g^{1,195}$	[12]
							Physical (50%glycerine-O ₂)	$k_L a = 0,0039 \left(\frac{P_g}{V_l}\right)^{0,735} u_g^{0,668}$ Small vibration intensity: $k_L a = 0,775 \left(\frac{P_g}{V_l}\right)^{0,159} u_g^{1,043}$ Large vibration intensity: $k_L a = 0,336 \cdot 10^{-4} \left(\frac{P_g}{V_l}\right)^{1,308} u_g^{1,624}$	
↑↑	22.8	6 12	0.55	6.35- 19	0.5- 0.25	$u_g = 0-1.2$	Physical (water-O ₂)	Small vibration intensity: $k_L a \propto \left(\frac{P_g}{V_l}\right)^a u_g^b$ $a=0.028-0.084; b=0.86-1.05$ Large vibration intensity: $k_L a \propto \left(\frac{P_g}{V_l}\right)^a u_g^b$ $a=0.73-0.92; b=0.50-0.61$	[16]

content enabled the maximal gas hold-up in the column according to some other investigations [19].

The chemical method was used for $k_L a$ determination. The chemical method is based on the consumption of sodium sulphite aqueous solution when oxygen from the air oxidizes sodium sulfite in the

presence of CuSO₄ as a catalyst (concentration 10⁻⁴ mol/dm³). The volumetric mass transfer coefficient was calculated on the basis of the experimentally determined rate of oxygen absorption and oxygen solubility in aqueous solution:

$$k_L a = (Ro_2)/Co_2^* \quad (2)$$

Table 2. Experimental set-up (characteristics) and operating conditions

	RPC1	RPC2
Column diameter (internal), cm	2.54	9.20
Column height (total), cm	200	105
Liquid height in a column, cm	183.6	85.6
Working volume, cm ³	831.5	5687
Nozzle diameter, cm	0.385	0.3 (two nozzles)
Axis diameter, cm	0.32	0.6
Length of the tie-rod, cm	12.4	12.4
Number of plates	65	15
Plate diameter, cm	2.5	9.0
Distance between plates, cm	2.54	5.0
Plate thickness, cm	0.1	0.15
Hole diameter (plate perforation; internal/external), cm	0.8/0.6	0.8/0.8
Plate free section area, %	0.51	0.454
Material:		
Column	Glass	Plexyglass
Plate	S.S.	Aluminium
Spacer between plates	Teflon	Teflon
Solid content, %	3.82 and 6.61	3.82 and 6.61
Solid sphere arrangement	5 spheres in each second or third space between the plates	52 or 90 spheres in each space between two plates
Amplitude, cm	2.35	
Vibration frequency, s ⁻¹	From 2 to 6	
Superficial gas velocity, cm/s	0.5; 1.0 and 1.5	

RESULTS AND DISCUSSION

The dependence of the volumetric mass transfer coefficient on the vibration intensity in a two- and three-phase RPC is shown in Figure 2. For the purpose of comparison data are shown in the same figure, which correspond, to the case of a RPC of smaller diameter [13]. It is obvious that increase of the $k_L a$ value followed by increases of the vibration intensity and the superficial gas velocity did not depend on the column diameter. Compared to the case of a two-phase RPC, the volumetric mass transfer coefficient has a greater value as a result, probably, of the much better end effective mixing and the influence of the solid phase on the bubble dispersion. Increases of the solid phase content from 3.8 to 6.6% do not markedly increase the volumetric mass transfer coefficient, although increase of the solid phase present in the RPC increases the gas hold-up in the column [19].

The volumetric mass transfer coefficient is more expressed in a column with a larger diameter (Figures 3-5), and does not depend on the system type (two or

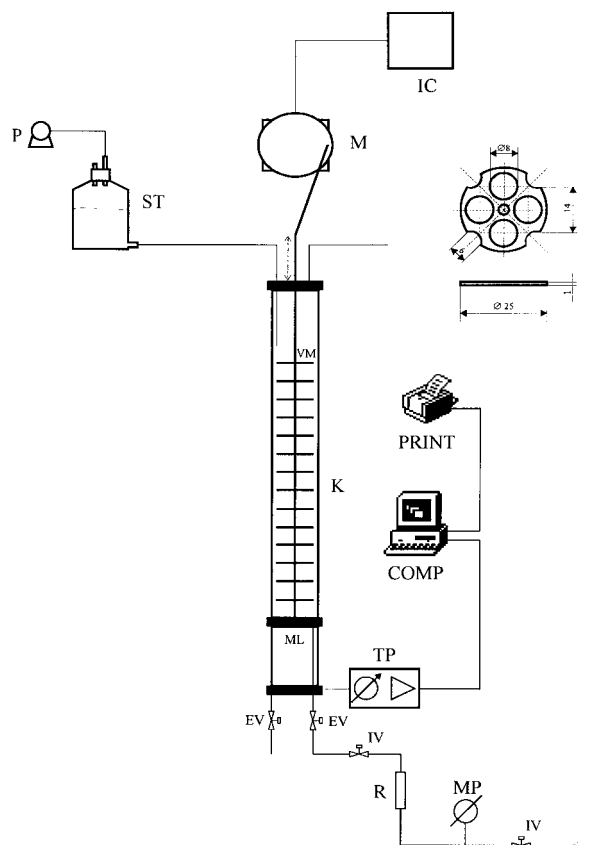


Figure 1. Experimental set-up scheme

three phase RPC), as a result, most probably of better mixing in the column and finally bubble dispersion. A higher value of the mass transfer coefficient could be achieved in a two-phase or three-phase RPC compared to a simple bubble column, air-lift reactor or stirred tank working at the same superficial gas velocity (Figure 5). Such evidence is the result of additional bubble dispersion caused by mechanical mixing and larger increases of the interfacial area in the RPC than in the other compared systems. As can be seen from the Figure 6, the volumetric mass transfer coefficient (gas-liquid, two-phase system) depends on the RPC geometry. At the same specific power of mixing, the highest value of the volumetric mass transfer coefficient was detected in a column of 9.2 cm diameter.

The volumetric mass transfer coefficient could be correlated with the power consumption for mixing and with the superficial gas velocity (Table 3). Generally, in both cases (two- and three-phase RPC) in RPCs of different diameter, it was observed that the exponent in such a correlation, for the dependency of the $k_L a$ on average power consumption, or on the superficial gas velocity, are very similar. As consequences of such evidence, the volumetric mass transfer coefficient could be correlated by both parameters with an equation of the same form, which is valid for columns of smaller and larger diameter, not depending on the investigated (two- or three-phase) system:

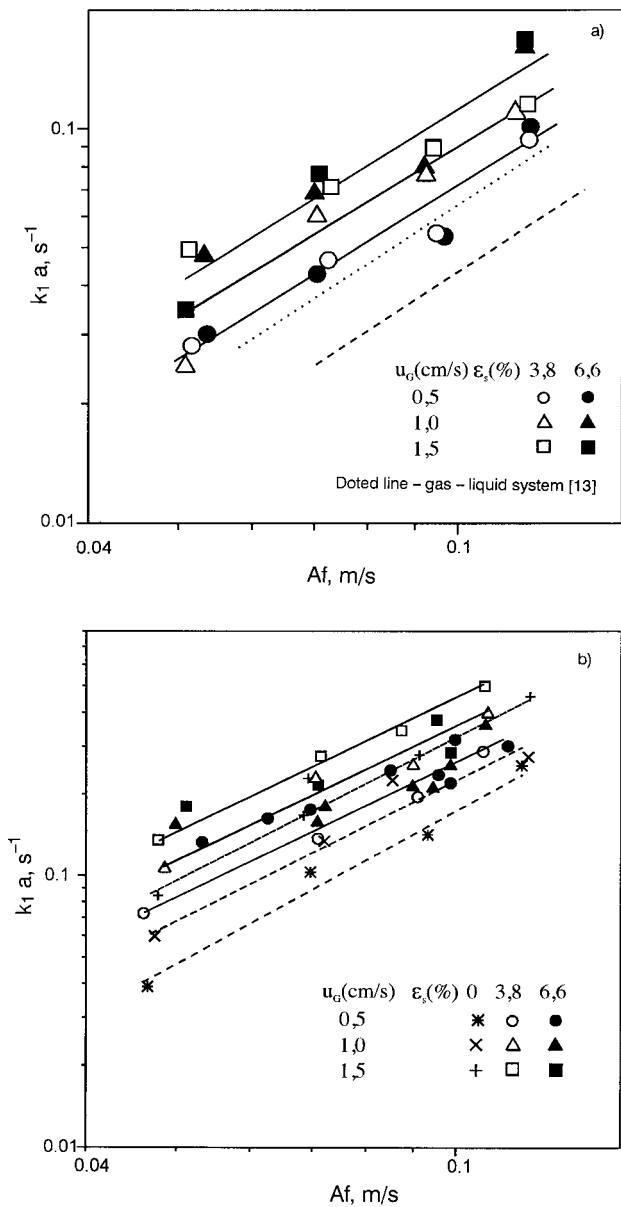


Figure 2. Dependence of $k_L a$ on vibration intensity: a) 2.54 and b) 9.2 cm column diameter

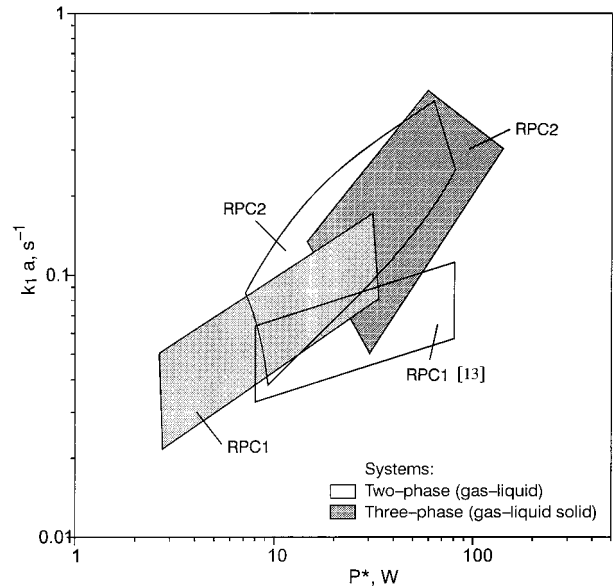


Figure 3. Comparison of the $k_L a$ values in RPCs of different geometry (RPC1 = 2.54 cm diameter; RPC2 = 9.2 cm)

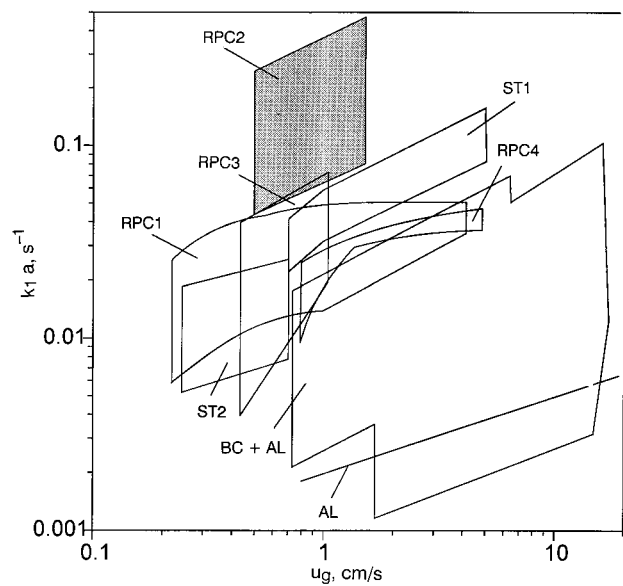


Figure 4. Comparison of $k_L a$ values for different reactor types and gas-liquid systems: RPC1=2.54 cm diameter [5]; RPC2 = 9.2 cm (this paper); RPC3 = 9.3 cm [10]; RPC4 = 5.08 cm [11]; BC (bubble column) and AL (air lift) [21, 22]; AL [23]; ST (stirred tank) [24]; ST [25]

Table 3. The empirical correlation for $k_L a$ determination

D_c (cm)	System	ϵ_s (%)	Correlation	r^2	Relative difference (%)
2.54	Three-phase	3.8	$k_L a = 1.025 (P_{av})^{0.94} u_G^{0.71}$	0.97	± 5.5
		6.6	$k_L a = 1.343 (P_{av})^{1.03} u_G^{0.81}$	0.94	± 13.0
		3.8 and 6.6	$k_L a = 0.548 (P_{av})^{0.71} u_G^{0.58}$	0.82	± 17.3
9.2	Two-phase	0	$k_L a = 1.198 (P_{av})^{0.74} u_G^{0.72}$	0.96	± 10.4
	Three-phase	3.8	$k_L a = 3.841 (P_{av})^{0.77} u_G^{0.97}$	0.98	± 5.4
		6.6	$k_L a = 0.633 (P_{av})^{0.88} u_G^{0.73}$	0.96	± 9.6
	Two-phase	0 - 6.6	$k_L a = 0.985 (P_{av})^{0.65} u_G^{0.64}$	0.94	± 10.7

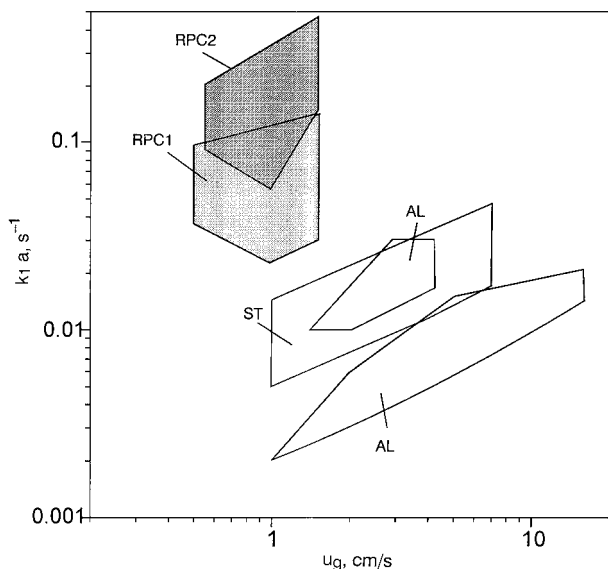


Figure 5. Comparison of k_La values obtained in different reactor types (three-phase system): RPC1 = 2.54 cm diameter (this paper); RPC2 = 9.2 cm (this paper); AL [22, 26]; ST [27].

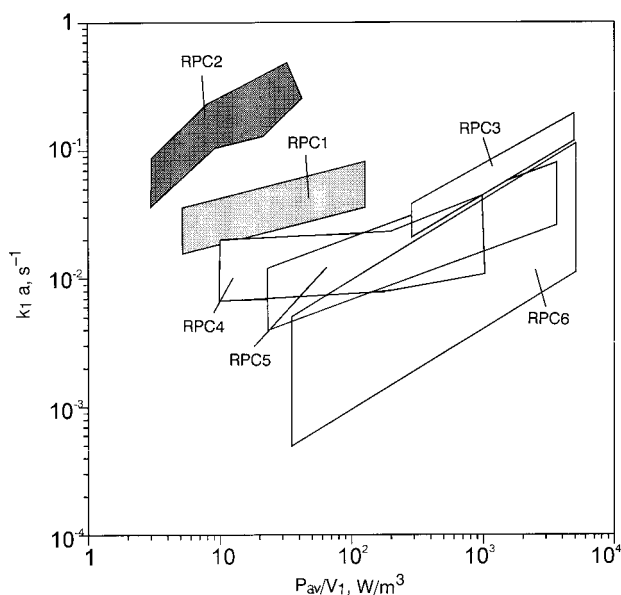


Figure 6. Comparison of k_La values in a RPC of the Karr type for different geometries (gas-liquid system): RPC1 = 2.54 cm diameter (this paper); RPC2 = 9.2 cm (this paper); RPC 3 [8]; RPC4 [16]; RPC5 [7]; RPC6 [12].

$$k_La = 0.719(P_{av})^{0.74}U_G^{0.63} \quad (3)$$

The values of the average power consumption in equation (3) were calculated on the basis of the empirically derived correlation [20], for each column geometry and the used liquids. There was fairly good agreement was proven between the calculated (equation 3) and experimentally determined values of the volumetric mass transfer coefficient (Figure 6; $r^2 = 0.93$; 45 data; relative percentage deviation $\pm 15.9\%$). The values of the exponents in equation (3) are very

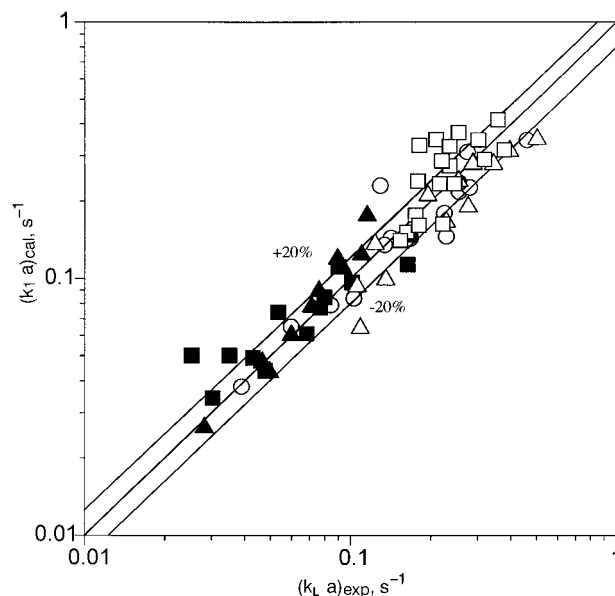


Figure 7. Comparison of the calculated (equation 3) and experimentally measured values of k_La ; legend: (black symbols – 2.54 cm diameter; open symbols – 9.2 cm); u_G cm/s: 0.5–1.5 cm/s; solid phase content – es, %; 0 – circle; 3.8 – triangle; 6.6 – square).

close, indicating that the same influence of the power consumption and superficial gas velocity could be expected.

CONCLUSION

The volumetric mass transfer coefficient in a three-phase RPC depends on the vibration intensity, the superficial gas velocity and content of the solid phase in the column. By increasing the vibration intensity and superficial gas velocity, the volumetric mass transfer coefficient increases. Such an influence does not depend on the investigated system (two- or three-phase RPC) and column diameter. Greater k_La values were detected in a three-phase RPC compared to values identified in a two-phase RPC under the same other operating conditions (vibration intensity and superficial gas velocity). Increases of the solid phase content in the case of a three-phase RPC did not influence the increase of k_La to a larger extent. However, a larger k_La value was detected in a column of 9.2 cm compared to the column of 2.54 cm diameter under other operating conditions being the same. Comparison of the k_La values obtained in a RPC to some other multiphase columns or bioreactors (bubble column, stirred tank reactor, air-lift) indicated the advantages of such a type of vibration mixing. Thus, in the case of a RPC with 9.2 cm diameter, the highest value of k_La was obtained at the same specific power consumed for mixing. A simple correlation was derived for the calculation of the k_La value in a three-phase RPC, which could be used for the purpose of the scale-up of such devices.

SYMBOLS USED

A	– specific interfacial area, m ² /m ³
A	– vibration amplitude, m
A _c	– cross-sectional area, m ²
A _f	– vibration intensity, m/s
C _L *	– equilibrium oxygen concentration, mol/dm ³
d _o	– hole diameter (plate), m
d _{av}	– average bubble diameter, m
D _c	– column diameter, m
f	– frequency, Hz
k _L	– mass transfer coefficient (in the liquid phase), m/s
k _{LA}	– volumetric mass transfer coefficient, 1/s
L _c	– column height, m
N _p	– number of plates
P _{av}	– average power consumption, W
P*	– total power consumption, W
r ²	– linear correlation coefficient
R _{O2}	– the rate of the oxygen mass transfer, mol/m ³ .s
U _G	– superficial gas velocity, m/s
U _L	– superficial liquid velocity, m/s
V _G	– volume of gas dispersion in the column, m ³

GREEK LETTERS

ε	– free area of the plate
ε _G	– gas hold-up
ε _s	– content of the solid phase in the column
μ	– dynamic viscosity, Pa.s
ρ	– density, kg/m ³
σ	– surface tension, N/m

INDEX

G	– gas
L	– liquid
max	– maximal value
av	– average value

ABBREVIATION

AL	– air lift reactor
BC	– bubble column
RPC	– reciprocating plate column
ST	– stirred tank reactor

IZVOD

PRENOS MASE U KOLONI SA VIBRACIONOM MEŠALICOM

I – Zapreminski koeficijent prenosa mase

(Naučni rad)

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U radu su prikazani rezultati ispitivanja maseno-prenosnih karakteristika kolone sa vibracionom (tipa Karr) mešalicom (dvo i trofazni sistemi). Za određivanje zapreminskog prenosa mase kiseonika upotrebljena je hemijska metoda oksidacije natrijum sulfita u prisustvu CuSO₄ kao katalizatora.

Izmerene vrednosti zapreminskog prenosa mase kiseonika pokazuju da ova vrednost zavisi od intenziteta vibracije, prividne brzine gasa i udela čvrste faze u koloni. Izvedena je korelaciona zavisnost koja povezuje zapreminski koeficijent prenosa mase kiseonika sa srednjom vrednošću snage utrošene pri mešanju sadržaja kolone (vibracije povratno periodičnog seta sa perforiranim pločicama, vibracione mešalice i prividne brzine gasa, a da ne zavisi od prečnika kolone i sistema koji je ispitivan (dvo- ili trofazni sistem):

$$k_{LA} = 0.719 (P_{av})^{0.74} U_G^{0.63}$$

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Ključne reči: vibraciona kolona • Kolona tipa Karr • Prenosenje mase • Zapreminski koeficijent prenošenja mase u tečnosti • Key words: reciprocating plate column • Karr type column • Mass transfer • Liquid phase volumetric mass transfer coefficient •

Doted line – gas – liquid system [13]	AL
$k_1 a, s^{-1}$	RPC2
$k_1 a, s^{-1}$	RPC1
$A_f, m/s$	ST
$A_f, m/s$	AL
Systems:	AL
Two-phase (gas-liquid)	RPC2
Three-phase (gas-liquid solid)	RPC1
$k_1 a, s^{-1}$	RPC3
$k_1 a, s^{-1}$	RPC4
P^*, W	RPC5
$k_1 a, s^{-1}$	RPC6
$u_g, cm/s$	
	RPC1
$u_g, cm/s$	
	RPC1
$k_1 a, s^{-1}$	
	RPC2
$P_{sr}/V_1, W/m^3$	
	RPC2
$(k_1 a)_{cal}, s^{-1}$	a, m^{-1}
$(k_1 a)_{exp}, s^{-1}$	a, m^{-1}
$(k_1 a)_{cal}, s^{-1}$	a, m^{-1}
RPC2	a, m^{-1}
RPC3	a, m^{-1}
RPC1	a, m^{-1}
ST1	a_{cal}, m^{-1}
RPC4	a_{cal}, m^{-1}
ST2	a_{exp}, m^{-1}
BC + AL	a_{exp}, m^{-1}