Mass Transfer in Multiphase Mechanically Agitated Systems

Anna Kiełbus-Rąpała and Joanna Karcz West Pomeranian University of Technology, Szczecin Poland

1. Introduction

In this chapter, the results of the experimental studies concerning the volumetric mass transfer coefficient $k_{L}a$ obtained for mechanically agitated gas – liquid and gas – solid – liquid systems are discussed.

Mechanically agitated gas – liquid and gas –solid – liquid systems are widely used in many processes, for example oxidation, fermentation or wastewater treatment. In such cases, oxygen mass transfer between gas and liquid phases in the presence of solid particles can be described and analyzed by means of the volumetric mass transfer coefficient $k_{L}a$. In the gas – liquid and gas – solid – liquid systems, the $k_{L}a$ coefficient value is affected by many factors such as geometrical parameters of the vessel, type of the impeller, operating parameters of the process (impeller speed, aeration rate), properties of the continuous phase (density, viscosity, surface tension, etc.) and also by the type, size and loading of solid particles.

To improve the efficiency of the processes conducted in gas – liquid and gas – solid – liquid three – phase systems two or more impellers on the common shaft are often used (Kiełbus – Rąpała & Karcz, 2009). Multiple – impeller stirred vessels due to the advantages such as increased gas hold – up, higher residence time of gas bubbles, superior liquid flow characteristics and lower power consumption per impeller are becoming more important comparing with single – impeller systems (Gogate et al., 2000). As the number of energy dissipation points increased with an increase in the impellers number on the same shaft, there is likely to be an enhancement in the gas hold – up due to gas redistribution, which results into higher values of volumetric gas – liquid mass transfer coefficient (Gogate et al., 2000).

Correct design of the vessel equipped with several agitators, therefore, the choice of the adequate configuration of the impellers for a given process depends on many parameters. That, which of the parameters will be the most important depends on the kind of process, which will be realized in the system. For the less oxygen demanding processes the designer attention is focused on the mixing intensity much more than on the volumetric mass transfer coefficient. When the most important thing is to achieve high mass transfer efficiency of the process, the agitated vessel should be such designed that the configuration of the agitators used ensure to get high both mixing intensity and the mass transfer coefficient values (Kiełbus-Rąpała & Karcz, 2010).

1.1 Survey of the results for gas-liquid system

Good mass transfer performance requires large interface area between gas and liquid and a high mass transfer coefficient. The obtaining the good dispersion of the gas bubbles in the liquid agitated depends on the respectively high agitator speed, therefore the characteristics of the gas-liquid flow are intensively studied (Paul et al., 2004); Harnby et al., 1997).

In literature, there are a large number of correlations for the prediction of volumetric mass transfer coefficients $k_L a$ in mechanically stirred gas-liquid systems (Linek et al., 1982, 1987; Nocentini et al., 1993; Gogate & Pandit, 1999; Vasconcelos et al., 2000; Markopoulos et al., 2007). Markopoulos et al. (2007) compared the results which were obtained by other authors for agitated Newtonian and non-Newtonian aerated liquids. The comparative analysis carried out by Markopoulos et al. (2007) shows that significant disagreement is observed taking into account the form of the correlations for $k_L a$. Therefore, no single equation exists representing all of the mass transfer data given in literature. The differences can mainly be ascribed to the differences in the geometry of the system, the range of operational conditions, capability to gas bubbles coalescence and the measurement method used. For a given geometry of the agitated vessel and liquid properties, volumetric mass transfer coefficient $k_L a$ for gas-liquid system is often described in literature by means of the following dependences

or

$$k_{\rm L}a = f(P/V_{\rm L}, w_{\rm og}) \tag{1}$$

$$k_{\rm L}a = f(n, w_{\rm og}) \tag{2}$$

where: $P/V_{\rm L}$ - specific power consumption, $w_{\rm og}$ - superficial gas velocity, n - agitator speed. Most often, empirical correlations have the form of the following equations

$$k_{\rm L}a = C_1 (P_{\rm G-L}/V_{\rm L})^{\rm a1} (w_{\rm og})^{\rm a2}$$
(1a)

or

$$k_{\rm L}a = C_2(n)^{\rm a3}(w_{\rm og})^{\rm a4} \tag{2a}$$

Detailed values of the exponents a1 and a2 in Eq. (1a) obtained by different authors are given in paper by Markopoulos et al. (2007).

Recently, Pinelli (2007) has studied the role of small bubbles in gas-liquid mass transfer in agitated vessels and analyzed two-fraction model for non-coalescent or moderately viscous liquids. Martin et al. (2008) analyzed the effect of the micromixing and macromixing on the k_{La} values. This study shows that each mixing scale has a particular effect on the mass transfer rate.

It is worth to notice that values of the k_La coefficient can be depended on the measurement method used (Kiełbus-Rąpała & Karcz, 2009). Although a number of techniques were developed to measure the k_La values, the unsteady-state gassing-out dynamic methods (Van't Riet, 1979; Linek et al., 1982; Linek et al., 1987; Linek et al., 1996; Ozkan et al., 2000; Lu et al, 2002, Garcia-Ochoa & Gomez, 2009) are preferably used as they are fast, experimentally simple and applicable in various systems. These methods are used in many variants. For example, Machon et al. (1988) used a variation of the dynamic method in which the gas from liquid was firstly removed by vacuum and the system was then aerated for some time. When aeration was stopped and the bubbles escaped from the liquid, the steady state concentration of dissolved oxygen was measured.

Apart from experimental studies, new methods are recently used to prediction the values of the volumetric mass transfer coefficient. Lemoine & Morsi (2005) applied artificial neural network to analyze mass transfer process in a gas-liquid system. Using CFD technique, Moilanen et al. (2008) modeled mass transfer in an aerated vessel of working volume 0.2 m³ which was equipped with Rushton, Phasejet or Combijet impeller.

1.2 Survey of the results for gas-solid-liquid system

The critical impeller speed for the solid suspension in liquid can be defined as the impeller speed at which no particles resting on the bottom of the vessel longer then 1- 3s (Zwietering, 1958). In multiple –impeller system introduction of gas into solid – liquid system implicate an increase in the impeller speed required for particles suspension just like in a single – impeller system. For solid suspension multiple impeller systems would prove to be disadvantageous if the distance between impellers is greater than the diameter of the impeller as there is an increase in the critical impeller speed for solid suspension (Gogate et al., 2000).

The data on the multiple – impeller systems working in the three – phase gas – solid – liquid systems are limited (Kiełbus-Rapała & Karcz, 2009). Various aspects of the three - phase system stirring in the vessel equipped with more than one impeller on the common shaft were the aim of the studies in papers (Dutta & Pangarkar, 1995; Dohi et al., 1999, 2004; Roman & Tudose, 1997; Majirowa et al, 2002; Jahoda et al., 2000; Jin & Lant, 2004). The impellers used were usually Rushton turbines or pitched blade turbine. Dutta & Pangarkar (1995) and Dohi et al. (1999) analyzed experimentally the critical impeller speed needed to gas dispersion simultaneously with solid suspension in the systems, with four and three impellers, respectively. Critical impeller speed in multiple - impeller system can be modified easily by choosing optimal impeller combination. Power consumption in multiple - impeller systems was studied by Dohi et al., 1999, 2004; Majirowa et al., 2002; Roman & Tudose, 1997. Investigations in such systems concerned also gas hold - up (Dutta & Pangarkar, 1995; Dohi et al., 1999, 2004; Majirowa et al., 2002; Jahoda et al., 2000) and mixing time (Dutta & Pangarkar, 1995; Dohi et al., 1999; Jahoda et al., 2000). Hydrodynamics problems in the system stirred by means of triple impellers were analysed by Jin & Lant (2004). Authors compared hydrodynamic conditions in the stirred vessel with the air - lift and bubble column.

Although k_{La} coefficients were widely studied in the two – phase gas – liquid multiple – impeller systems (Arjunwadkar et. al, 1998; Fujasowa et al., 2007; Puthli et al., 2005; Moucha et al., 2003; Linek et al., 1996; Wu, 1995; Yoshida et al., 1996; Yawalkar et al., 2002) and many papers (Alper et al., 1980; Brehm et al., 1985; Lu et al., 1993; Kralj & Sinic, 1984; Chapman et al., 1983; Ruthiya et al., 2003) regard k_{La} measurements in the three – phase system stirred in the vessel equipped with single – impeller, the information in literature on this parameter in multi impeller gas – solid – liquid system is substantial.

On the basis of the literature data, the effect of solid particles on gas – liquid oxygen mass transfer rate in single – impeller systems at low solid concentrations analyzed Kiełbus – Rąpała & Karcz (2009) and Karcz & Kiełbus – Rąpała (2006). This effect was investigated by many authors (Alper et al., 1980; Brehm et al., 1985; Oguz et al., 1987; Lu et al., 1993; Kralj &

NT.	D.(T	Size of	Solids	$k_L a$ coefficient
No	Kef.	Type of solids	particles	concentration	Value
1	Chandrasekaran	activated carbon	<100	0-0.2 mass %	increases 1.5 times
1.	& Sharma, 1977	0–2 mass %	<100 µm	0.2–2 mass %	Constant
2.	Joosten et al., 1977	Polypropylene, sugar, glass beads, 0–40% vol.	53-105, 250 μm 74-105 μm 53 μm, 88 μm	0–20% vol. 20–40% vol.	Constant rapidly decreases
3.	Alper et al., 1980	activated carbon, quartz sand 0–2 mass %	< 5 μm < 5 μm	0–0.2 mass % 0.2–2 mass %	increases 1.5 times constant
4.	Lee et al., 1982	glass beads 0–50 mass %	56 µm	0–30 mass % 30–50 mass %	decreases rapidly decreases slowly
5.	Chapmann et al., 1983	glass ballotini 3 and 20 mass %		0–3 mass % 3–20 mass %	slightly decreases significantly decreases
6.	Kralj & Sincic, 1984	activated carbon 0.25 and 0.5 mass %		the whole range	Constant
7	Brehm et al.,	Al ₂ O ₃	50 um 300 um	0–5% vol.	increases 1.5 times
7.	1985	0–10% vol.	50 µm, 500 µm	5–10% vol.	Decreases
8.	Greaves & Loh, 1985	ion exchange resin 0–40 mass % glass ballotini 0–50 mass %	780±70μm 665 μm, 1.300 μm	0–15 mass % 15–50 mass %	constant decreases rapidly
9.	Bartos & Satterfield, 1986	glass beads 0-30% vol.	60 μm	the whole range	decreases with solid concentration increasing
10.	Mills et al., 1987	glass beads 0-40% vol.	66 µm	the whole range	decreases with solid concentration increasing
11.	Lu & Liang, 1990; cit. in Lu et al., 1993	kaolin 0-4 mass %	5.7 µm	the whole range	decreases with solid concentration increasing
12.	Lu et al., 1993	kaolin powder 0-4.5% vol. yeast 0-8% vol.	5.5 μm 3.7 μm	the whole range	increases to maximum (c.a. 2%) and then decreases
13.	Özbek & Gayik, 2001	Schotchbrite™ pads pieces (biomass): 5–25% vol.	0.1625 cm ³ , 0.65 cm ³ , 1.4625 cm ³	the whole range	decreases with solid concentration increasing
14	Kiełbus & Karcz,	Sea sand	335 um	0.5 mass %	Increases
	2006	0.5–5 mass %	ccc pin	2.5-5mass %	Decreases
15.	Littlejohns & Daugulis, 2007	nylon 6.6, glass beads; Silicone rubber, styrene-butadiene copolymer	2.59 mm 6 mm 2.5 mm 3.59 mm	167g/l	Increases up to 268% Reducing by up to 63%

Table 1. The effect of presence and concentration of solid particles on $k_{\rm L}a$ values determined by different authors

Sinic, 1984; Chapman et al., 1983, Ozkan et al., 2000; Kordac & Linek, 2010). The results of their studies were not always similar. The effect of presence and concentration of solid particles on volumetric mass transfer coefficient $k_{L}a$ determined by different authors is compared in Table 1.

Volumetric mass transfer coefficient value increased (Alper et al., 1980; Brehm et al., 1985; Lu et al., 1993), was constant (Kralj & Sinic, 1984) or slightly decreased (Chapman et al., 1983) with adding the particles in the system. Such differences can suggest that very important are properties of the particles, containing particle size (Ozbek & Gayik, 2001), density, hydrophobicity, oxygen diffusivity (Zhang et al., 2006; Littlejohns et al., 2007) and concentration. Various mechanisms were proposed for describing an enhancement of k_{La} caused by the particles. Shuttling effect, where absorptive particles enter the liquid boundary layer, absorbing dissolved gas and then desorbing this gas when back in the bulk phase. This effect is used especially for small particles with a size equal or smaller than the gas - liquid boundary layer (Ruthiya et al., 2003). For inert particles, without absorptive properties, the possible mechanisms which explain an enhancement of k_{1a} include boundary layer mixing and gas - liquid interface changing. Boundary layer mixing, involving an increase in $k_{\rm L}$ due to turbulence at gas – liquid interface (Ruthiya et al., 2003; Kluytmants et al., 2003; Zhang et al., 2006), which causes a larger refreshment rate of liquid boundary layer by mixing with the bulk fluid. Changes in the gas – liquid interfacial area can be result of the particles presence at the gas - liquid interface, which can collide and interact with the gas liquid interface or may induce turbulence near or at it, leading to a smaller effective diffusion layer or causing coalescence inhibition and an increase in a interfacial area (Ruthiya et al., 2003). However, a simple mechanism describing all cases does not exist. Comparing the data shown in Table 1 for a high concentration of the particles in the agitated three phase system it is worth to notice that values of the k_{La} coefficient rather decrease when the solid concentration increases (for example data given by Joosten et al., 1977; Lee et al., 1982; Brehm et al., 1985).

The results of the volumetric mass transfer coefficient $k_L a$ for the agitated three phase gassolid-liquid system are usually correlated by different authors in the form of the following dependence

$$k_{\rm L}a = f(P/V_{\rm L}, w_{\rm og}, \ldots) \tag{3}$$

Some correlations presented in literature are shown in Table 2.

Galaction et al. (2004) conducted the $k_{L}a$ coefficient measurements in the three – phase system containing various type of microorganisms. Authors studied the effect of concentration and morphology of biomass, specific power input and superficial air velocity on volumetric gas – liquid mass transfer coefficient value in the stirred vessel equipped with two turbine stirrers. They proposed the following mathematical correlation

$$k_{\rm L}a = aX^{\alpha} (P_{\rm G-L-S}/V_{\rm L})^{\beta} w_{\rm og}^{\gamma}$$
⁽⁴⁾

describing the influence of considered factors on $k_{L}a$. The values of the parameters a, α , β and γ in Eq. (4) are given in Table 3. On the basis of the measurements authors have found that, in the bioreactor, the value of $k_{L}a$ depends not only on operating and geometrical parameters but also on the type and morphology of the microorganisms in the system.

Author	Equation	System	Impeller	D [m]	Operating parameters	Comments
Kralj & Sincic 1984	$k_L a = 0.109 \left(\frac{P_{G-L-S}}{V_L}\right)^{0.213} w_{og}^{0.34}$	Aqueous solution of NaOH - air - active carbon	Rushton turbine; d/D = 0.33	0.1	n = 12.5-50 1/s; $w_{\text{og}} = 10^{-3}-10^{-2}$ m/s; X = 0.25, 0.5 mass%	required the presence of the antifoams (0.03-0.1% mass.)
Brehm et al. 1985	$k_{\rm L}a = 0.5612 \left(\frac{P_{G-L-S}}{V_L}\right)^{0.65} \left(\frac{\eta_m}{\eta}\right)^{0.47} w_{og}^{0.4}$	water or aqueous solution of polyetylen glycol – air – different solid particles	Turbine with 4 flat blades; d/D = 0.5	0.145	n = 8.33-13.33 1/s; Vg = 0.139- 2.78 x10-4 m ³ /s; $X \le 10$ % obj.	solids: Al ₂ O ₃ , Fe ₂ O ₃ kiselguhr, glass beads $(d_p = 50-300 \mu m)$; sand $(d_p = 300 \mu m$ and powder); 185 exp. points; $\Delta \pm 11.5\%$
Brehm & Oguz 1988	$k_L a = 3.07 \cdot 10^{-3} \frac{\left(\frac{P_{G-L-S}}{V_L}\right)^{0.75} w_{og}^{0.5a} D_L^{0.5}}{\eta_m^{0.34} \sigma_L^3}$	water (or various organic liquids) – air – various solid particles	Turbine with 4 flat blades; d/D = 0.5	0.145	$\begin{split} \eta_{\rm m} = &1.3-40.7 \\ x10^3 \ {\rm Pas}; \\ P_{\rm G-L,S}/V_{\rm L} = &0.75- \\ &6.3 \ {\rm kW/m^3}; \\ &w_{\rm og} = &0.84-4.2 \\ {\rm m/s}; \\ &\sigma_{\rm L} = &24.3-71.8 \\ x10^{-3} \ {\rm N/m}; \\ &D_{\rm L} = &0.68-2.41 \\ {\rm m^2/s}; \end{split}$	Particles: CaCO ₃ , Fe ₂ O ₃ , BaSO ₄ , n [1/s] = const = 10.83; $\alpha = (\sigma_{\text{water}}/\sigma_{\text{L}})^{1/2}$ $\Delta \pm 11\%$

Table 2. The correlations of the volumetric mass transfer coefficient $k_L a$ for the agitated gassolid-liquid systems and the applicability of the equations

Type of biomass	Α	α	β	γ	±Δ, %
Propionibacterium shermanii	6.586	-0.282	-0.0286	0.429	7.8
Saccharomyces cerevisiae	52.44	-0.702	-0.0762	0.514	6.8
Penicillium chrysogenum (aggregates)	0.193	-0.269	0.0288	0.257	8.4
Penicillium chrysogenum (free structure)	33.59	-1.012	-0.0463	0.94	7.6

Table 3. The values of the coefficient and exponents in Eq. (4)

2. Experimental

In this part of the chapter, our experimental results of the mass transfer coefficient k_1a obtained for the gas - liquid or gas - solid - liquid systems agitated in a baffled vessel equipped with single high-speed impeller or the system of the two impellers are presented. Experimental studies were conducted in cylindrical transparent vessel of the inner diameter D = 0.288 m. Geometrical parameters of the vessel used are shown in Fig. 1. The vessel, equipped with a flat bottom and four baffles of the width B = 0.1D, was filled with liquid up to the height H = D (Fig. 1a) and H = 2D (Fig. 1b). The working liquid volumes were $V_L =$ 0.02 m³ and $V_{\rm L}$ = 0.04 m³, respectively. In the vessel with single impeller the impeller was placed at a hight h = 0.33D. In the tall vessel two high-speed impellers were located on the common shaft. The distance of the impeller from the bottom of the vessel was $h_1 = 0.167H$ for the lower and $h_2 = 0.67H$ for the upper impeller, respectively. All impellers have diameter d = 0.33D. Gas was introduced into liquid through a ring shaped sparger, of the diameter $d_d = 0.7d$. The sparger with 44 symmetrically drilled holes of 2 mm in diameter was located at the distance $e = 0.5h_1$ from the bottom of the vessel. To avoid surface aeration of the liquid which could disturb the measurements, the vessel was covered at the top with a transparent lid.



Fig. 1. Geometrical parameters of the stirred vessel; D = 0.288m; d = 0.33D; a) H = D; b) H = 2D

Four types of high-speed agitators (Fig. 2) differing in generated fluid pumping mode were tested: Rushton and Smith turbines (radial flow impellers), A 315 (mixed flow) and HE 3 (axial flow impeller). As a single agitator were used: RT, CD 6 and A 315.

The measurements were carried out for gas-liquid and gas-solid liquid systems. Distilled water were used as a continuous phase, whilst air as gas phase. The second dispersed phase in a three-phase system was fraction of sea sand particles with mean diameter $d_p = 335 \ \mu m$

and density $\rho_{\rm p} = 2600 \text{ kg/m}^3$. The measurements were conducted under various aeration rates, impeller speeds, and solid particles concentration. The measurements were performed for three different solid concentrations, X = 0.5, 2.5 and 5 mass %. The experiments were conducted at five different values of gas flow rate from the following range: $V_{\rm g}$ (m³/s) $\in \langle 0; 5.56 \times 10^{-3} \rangle$ (superficial gas velocity $w_{\rm og}$ (m/s) $\in \langle 0; 8.53 \times 10^{-3} \rangle$). Power consumption was measured using the strain gauge method.



Fig. 2. Agitators used in the study; a) Rushton disc turbine (RT), b) Smith disc turbine (CD 6), c) A 315, d) HE 3

All measurements were carried out within the turbulent regime of the fluid flow in the agitated vessel. During the measurements conducted in the gas-solid-liquid system, impeller speeds higher than the critical impeller speeds n_{JSG} for the three-phase system were maintained. Critical agitator speeds in the gas-solid-liquid system were evaluated on basis of the Zwietering criterion (Zwietering, 1958).

To determine the volumetric gas-liquid mass transfer coefficients $k_L a$ the unsteady-state gassing-out dynamic method (Van't Riet, 1979; Ozkan et al., 2000; Lu et al., 2002) was used. The dynamic methods are used in many variants. In this study, the variant in which, at constant values of the impeller speed and gas flow rates, the kind of gas introduced into the vessel is changed was used. The gases were air and nitrogen. At the beginning of the

experiment, in order to remove oxygen, nitrogen was passed through the system. When the value of dissolved oxygen was lowered, air replaced nitrogen. After interchanging $N_2 \rightarrow air$, absorption of oxygen followed. The change of the oxygen concentration dissolved in liquid was measured by means of fast oxygen probe (galvanic type) coupled with an oxygen meter (CO-551, ELMETRON). The time delay of the measuring probe was ca. 3 s. This sensor was immersed in the liquid and placed at half distance between the baffles.

In calculation of k_La , the response time of the probe should be taken into account (Kiełbus-Rąpała & Karcz, 2009). Response time is defined as the time required by the probe used in the study to measure 63 % of the overall value of the oxygen concentration change (Van't Riet, 1979) and it is related to the oxygen diffusion through the membrane of the probe. According to this fact, the measured values are sufficiently reliable (error below 6 %) only for the response time of the probe shorter or equal to $(1/k_La)$. Therefore, in the experiments, evaluation of the oxygen probe response time was made by measuring the oxygen concentration changes after moving the probe from a liquid saturated by nitrogen to a saturated oxygen solution. The results indicate that using this probe, the k_La values of less than 0.1 s⁻¹ are reliable. All values obtained in the measurements were lower than 0.1 s⁻¹, so the error resulting from the response time τ could be neglected.

The $k_L a$ coefficient values were determined from the slope of the plot $\ln[(C^*-C_0)/(C^*-C)] = f(t)$. In this dependence, *t* denotes time whilst C^* , C_0 , and *C* describe saturation concentration of gas in liquid, and concentrations of the gas at time t = 0 and *t*, respectively. The calculation was performed using the least-squares method.

3. The results

3.1 The results for gas-liquid system

Experimental investigations of the mass transfer process in the two-phase gas-liquid systems enable to determine the changes of the volumetric mass transfer coefficient values with operating parameters (impeller speed *n* and superficial gas velocity w_{og}). The results for the vessel with single impeller are represented by the data obtained for CD 6 impeller (Fig. 3). In this fig. the dependencies of $k_{La} = f(n)$ for various values of w_{og} are compared. It follows from the results that for all agitator tested the k_{La} coefficient value significantly increased with the increase both impeller speed and superficial gas velocity.



Fig. 3. The dependence of $k_{L}a = f(n)$ for single Smith turbine (CD 6) working in a gas-liquid system; varied values of $w_{og} \times 10^3 \text{ m/s}$: (\diamond) 1.71; (\blacksquare) 3.41; (x) 5.12; (\bullet) 6.82; (Δ) 8.53

The data, concerning double impeller systems, obtained for various values of superficial gas velocity w_{og} , are presented in Fig. 4a for the vessel with A 315 – Rushton turbine system and in Fig. 4b for the vessel with Smith turbine–Rushton turbine system. It can be observed that similarly to the vessel with single impeller, volumetric mass transfer coefficient is strongly affected by both impeller speed and superficial gas velocity. The differences in the k_La values between particular values of superficial gas velocity w_{og} are greater in the case of Smith turbine – Rushton turbine configuration.



Fig. 4. The dependence of $k_L a = f(n)$ for two configurations of the agitators: a) A 315 (lower)–RT (upper), b) CD 6 (lower) –RT (upper); gas–liquid system; varied values of $w_{og} \times 10^3$ m/s: (\diamond) 1.71; (\bullet) 2.56; (Δ) 3.41; (\bullet) 5.12; (\Box) 6.82

According to the literature data our results confirmed that in a gas-liquid system the volumetric mass transfer coefficient is strongly affected by the type of the agitator used for mixing of the system. The comparison of the $k_{L}a$ coefficient values obtained for three single agitators: RT, CD 6 and A 315 are presented in Fig. 5. In the vessel with radial flow both Rushton and Smith turbines the values of $k_{L}a$ coefficient were significantly higher compared to the values characterized the vessel equipped with mixed flow A 315 impeller. This tendency was observed in the whole range of the measurements.



Fig. 5. The effect of a single impeller type on the $k_{L}a$ coefficient value in a gas-liquid system; n = const = 11.67 1/s; (Δ) A 315; (\circ) CD 6; (\Box) RT

The effect of the impeller configuration on the volumetric mass transfer coefficient $k_L a$ value is presented in Figs. 6 and 7. The results concerning mass transfer processes conducting in the vessel equipped with two agitators on a common shaft showed that the type of both

lower and upper impeller strongly affects $k_L a$ coefficient values. In Fig. 6 the dependencies $k_L a = f(n)$ for different impeller configurations working in a gas-liquid system were compared. Fig. 6a presents the effect of the type of an upper agitator, whilst Fig. 6b the effect of the type of a lower agitator.



Fig. 6. The effect of the configuration of agitators on the $k_{L}a$ coefficient value; a) the effect of an upper agitator; empty points: RT-A 315 (upper), filled points: RT-HE 3 (upper); b) the effect of lower agitator; empty points: A 315 (lower)–RT; filled points: CD 6 (lower)–RT (upper); gas–liquid system; varied values of $w_{og} \times 10^3$ m/s: ($\diamond \bullet$) 1.71; ($\Delta \bullet$) 3.41; ($\Box \bullet$) 6.82

In the two-phase distilled water-air system significantly higher values of the volumetric mass transfer coefficient were obtained using the set of the impellers with A 315 as an upper one. Comparison of the $k_{L}a$ coefficient values for three chosen values of superficial gas velocity w_{og} and both configurations differ in an upper impeller is presented in Fig. 6a, 7a. The A 315 impeller (data represent by empty points in Fig. 6a), which generates radial-axial fluid flow in the vessel, let to ensure more advantageous conditions for conducting of mass transfer process, comparing with generating typical axial liquid circulation HE 3 impeller (data represent by filled points in Fig. 6a). At the constant values of agitator speeds, the differences in $k_{L}a$ coefficient values increase with the increase of quantity of gas phase introduced into the liquid volume. At the gas velocity $w_{og} = 6,28 \times 10^{-3} \text{ m/s}$, $k_{L}a$ values in the system agitated by RT-A 315 configuration were over 20 % higher, than those obtained analogously using RT-HE 3 set of agitators.



Fig. 7. Dependence $k_{L}a = f(w_{og})$ for different impeller configuration; a) various upper impeller: filled points: RT-HE 3, empty points: RT-A 315, n = 12.5 1/s (triangles), n = 15.83 1/s (circles); b) various lower impellers: (Δ) A 315-RT, (\Box) CD 6-RT; n = 13.33 1/s;

Comparison of the results obtained for double-impeller system differ in a lower impeller (Fig. 6b) shows, that at constant value of the impeller speed, the highest values of the k_{La} coefficient were obtained using Smith turbine–Rushton turbine configuration (data represent by filled points in Fig. 6b). At higher values of superficial gas velocity the differences between volumetric mass transfer coefficient values adequate to each configuration increased. The A 315 impeller, which differs in the generated profile of the fluid circulation from the turbine, is less effective as a lower impeller for the mass transfer processes in the agitated vessel comparing with CD 6 impeller (Fig. 7b).

The results of the studies of the volumetric mass transfer coefficient in the gas-liquid system were described mathematically, by means of the dependency, which connect k_La coefficient with unit power consumption (P_{G-L}/V_L , W/m^3) and superficial gas velocity (w_{og} , m/s):

$$k_L a = A \left(\frac{P_{G-L}}{V_L}\right)^B w_{og}^{C}$$
⁽⁵⁾

The values of the coefficient *A* and exponents *B*, *C* in this Eq. are collected in Table 4 for single impeller system and in Table 5 for all tested configurations of double impeller. In these tables mean relative errors $\pm \Delta$ for each set of agitators is also presented. For the vessel equipped with single impeller Eq. (5) is applicable within following range of the measurements: $P_{\text{G-L}}/V_{\text{L}} \in <160$; 5560 W/m³>; $w_{og} \in <1.71 \times 10^{-3}$; 8.53×10⁻³ m/s[>]. The range of an application of this equation for the double impeller systems is as follows: $Re \in <7.9 \times 10^4$; 16.3×10⁴>; $P_{\text{G-L}}/V_{\text{L}} \in <540$; 4500 W/m³>; $w_{og} \in <1.71 \times 10^{-3}$; 6.82×10⁻³ m/s[>].

No.	Impeller	Α	В	С	±Δ, %
1.	Rushton Turbine	0.029	0.44	0.534	3.7
2.	CD 6	0.028	0.374	0.622	3.3
3.	A 315	0.122	0.432	0.79	5.2

Table 4. The values of the coefficient *A* and exponents *B*, *C* in Eq. (5) for single impeller systems (Kiełbus-Rąpała et al., 2010)

No	Configuration	Δ	В	C	+ 1 %		
INU.	lower	ower upper		D	C	±Δ, ⁄0	
1.	Rushton Turbine	A 315	0.098	0.404	0.689	7	
2.	Rushton Turbine	HE 3	0.081	0.374	0.636	6.5	
3.	A 315	Rushton Turbine	0.027	0.466	0.575	3	
4.	CD 6	Rushton Turbine	0.092	0.334	0.6	2.6	

Table 5. The values of the coefficient *A* and exponents *B*, *C* in Eq. (5) for double impeller systems (Kiełbus-Rapała & Karcz, 2010)

3.2 The results for gas-solid-liquid system

On the basis of the experimental studies performed within turbulent regime of the liquid flow the volumetric mass transfer coefficient values for gas-solid-liquid systems agitated by single or double impellers were determined. The results obtained confirm that in the system with solid particles the $k_{L}a$ coefficient value is strongly affected by operating parameters, and also the presence and concentration of solid phase in the agitated systems.

The results of the measurements of volumetric mass transfer coefficients k_La in two phase gas-liquid and three phase gas-solid-liquid system agitated by single CD 6 impeller are compared in Fig. 8 (Karcz & Kiełbus-Rąpała, 2006). In this Fig. empty points are ascribed to the two-phase system whilst filled points correspond to the three-phase system with the concentration of the solid particles 0.5 (Fig. 8a) and 2.5 mass% (Fig. 8b), respectively.



Fig. 8. Dependence $k_{L}a = f(n)$ for a single impeller CD 6; filled points: X = 0 mass %; empty points: a) X = 0.5 mass %, b) X = 2.5 mass %; various $w_{og} \times 10^3$ m/s: ($\diamond \bullet$) 1.71; ($\Box \bullet$) 3.41; (+×) 5.12; ($\circ \bullet$) 6.82; ($\Delta \bullet$) 8.53 (Karcz & Kiełbus–Rąpała, 2006)

Analyzing the data in this Fig it could be concluded that in gas-solid-liquid system both superficial gas velocity and impeller speed influenced $k_{\rm L}a$ coefficient value. Similarly to the two-phase system $k_{\rm L}a$ coefficient value depends on the impeller speeds linearly and $k_{\rm L}a$ increases with the increase of *n*. Also the increase in aeration rate of three-phase system (described by superficial gas velocity $w_{\rm og}$) causes the increase of the volumetric mass transfer coefficient.

It follows from the results that introducing solids to the system affects volumetric mass transfer coefficient k_{La} . In the system with 0.5% of solid particles inside, the values of the k_{La} are significantly higher than in the two-phase system (Fig. 8a). In coalescing system, it can be explained that solids in a small quantity during their movement in the system hit the gas bubbles, which are breaking into the smaller ones and in consequence, the interfacial area increases. However, adding more particles – 2.5 mass %, causes the decrease of k_{La} values. For lower values of superficial velocity w_{og} , the volumetric mass transfer coefficient values are even lower than those obtained in the gas – liquid system (Fig. 8b).

Results of the k_La coefficient measurements for two double-impeller configurations working in gas-liquid, and gas-solid-liquid systems are compared in next Figs (Kiełbus-Rapała & Karcz, 2009). The data for A 315 (lower)–RT (upper) impeller configuration are presented in Fig. 9, whilst those agitated by CD 6 (lower)–RT (upper) can be found in Fig. 10. In these figures, similarly to the data in Fig. 8 filled points are related to the two-phase system whilst empty points correspond to the three-phase system with the concentration of solid particles 0.5 mass % (Figs 9a, 10a) and 2.5 mass % (Figs 9b, 10b), respectively.

The results for the system agitated by two impellers on a common shaft show the same dependency of the volumetric mass transfer coefficient as was observed in the vessel equipped with single impeller. In the system with 0.5 mass % of solid particles, the values of k_{La} are significantly higher compared to the data for two-phase system for both

configurations of the impellers (Figs. 9a, 10a). It is worth to notice that the differences between the k_{La} values for the system with or without particles are greater for the A 315–RT configuration (Fig. 9). In the three-phase system stirred by CD 6–RT impellers, volumetric mass transfer coefficients for the lowest values of superficial gas velocity were even lower than those for gas-liquid system. These observations are also confirmed by the literature results concerning single-impeller (Alper et al., 1980; Özbek & Gayik, 2001) or double-impeller (Galaction et al., 2004) systems. For low concentration of solids, the increase of k_{La} values can be explained by the interaction of particles with gas bubbles. The particles present in the liquid cause an increase in the turbulence on the film surrounding the gas bubbles, promoting thus the bubbles surface renewal and breaking, or leading to a smaller effective diffusion layer (Galaction et al., 2004; Alper et al., 1980; Kluytmans et al., 2003). Solids in low concentration and of small particle size can, during their movement in the system, collide and interact with the gas-liquid interface and, in consequence, the gas bubbles break into smaller ones causing an increase in interfacial area (Kiełbus-Rapała & Karcz, 2009).



Fig. 9. Dependence $k_L a = f(n)$ for configuration A 315 (lower)–Rushton turbine (upper); filled points: X = 0 mass %; empty points: a) X = 0.5 mass %, b) X = 2.5 mass %; various $w_{og} \times 10^3$ m/s: ($\diamond \bullet$) 1.71; ($\Delta \bullet$) 3.41; ($\Box \bullet$) 6.82 (Kiełbus–Rąpała & Karcz, 2009)



Fig. 10. Dependence $k_{L}a = f(n)$ for the stirrers design CD 6 (lower)–Rushton turbine (upper); filled points: X = 0 mass %; empty points: a) X = 0.5 mass %, b) X = 2.5 mass %; various $w_{\text{og}} \times 10^3 \text{ m/s:}$ ($\diamond \bullet$) 1.71; ($\Delta \blacktriangle$) 3.41; ($\circ \bullet$) 5.12; ($\Box \blacksquare$) 6.82 (Kiełbus–Rąpała & Karcz, 2009)

The investigations of mass transfer process in the system agitated by double-impellers confirm that the volumetric mass transfer coefficient increases, compared to the system without solids, only below a certain level of particles concentration.

Introducing more particles, X = 2.5 mass % into the system causes a decrease of $k_{L}a$ in A 315–RT (Fig. 9b) as well as in CD 6–RT (Fig. 10b) configuration. This tendency (reduction of $k_{L}a$ with increasing solid concentrations) was observed also in literature (Özbek & Gayik, 2001). It was proposed that increasing solid concentration causes an increase of slurry viscosity, resulting in an increase of coalescence processes and, in consequence, decrease of $k_{L}a$ coefficients.

The effect of solids on $k_L a$ could be probably associated with the differences in flow patterns generated in the agitated vessel as seen by the significant differences in the $k_L a$ values obtained using different impeller designs. Özbek and Gayik (2001) stated that the concentration at which $k_L a$ began to decline depended on solids type and size of the particles.

The results obtained for the solid concentration X = 2.5 mass % (Kiełbus-Rapała & Karcz, 2009) differ from those of a single-impeller system (Fig. 8) so the effect of particles with this concentration for the double – impeller system was not the same. Although in both cases, the values of $k_{L}a$ were lower compared to the system with X = 0.5 mass %, $k_{L}a$ coefficients were significantly lower compared to a gas-liquid system at all w_{og} values (Figs 9, 10), whilst in a single-impeller system (Fig. 8b), the volumetric mass transfer coefficient values were lower only for lower values of superficial gas velocity w_{og} (Karcz & Kiełbus-Rapała, 2006).

The effect of the solid phase concentration on the volumetric mass transfer coefficient value for both impeller configurations differ in lower impeller is presented in Fig. 11 (Kiełbus-Rąpała & Karcz, 2009). The values of k_La coefficient, obtained at a constant value of superficial gas velocity for the system with different solid concentration agitated by A 315-RT configuration (Fig. 11a) and CD 6-RT (Fig. 11b) are compared. In this Fig. triangles described data for gas-liquid system, squares – X = 0.5 mass%, circles – X = 2.5 mass%.



Fig. 11. Effect of the solid concentration on $k_{L}a$ value for double impeller configuration; a) A 315 (lower)–Rushton turbine (upper), b) CD 6 (lower)–Rushton turbine (upper); w_{og} = const = 3.41×10^{-3} m/s, X = 0 mass % (triangles); X = 0.5 mass % (squares); X = 2.5 mass % (circles) (Kiełbus–Rąpała & Karcz, 2009)

In the three-phase system agitated by both impeller configurations differ in an upper impeller type significantly lower $k_L a$ coefficient value were obtained in the system with solid

concentration X = 2.5 mass %. The data for RT-A 315 and RT-HE 3 agitator design are compared in Fig. 12 in which squares concerned system with X = 0.5 mass% whilst triangles : X = 2.5 mass%.



Fig. 12. The dependence of $k_L a = f(n)$ for two configurations of the agitator working in gassolid-liquid system; empty points: RT (lower)–A 315 (upper), filled points: RT (lower)– HE 3 (upper); various values of X: (\square) 0.5 mass %; ($\Delta \blacktriangle$) 2.5 mass %

The effect of the single agitator type on the volumetric mass transfer coefficient $k_{L}a$ is presented in Fig. 13. Comparing the results obtained for each impeller it can be stated that, in general, the highest values of the $k_{L}a$ coefficient are characterized for Rushton turbine. However, at higher values of the superficial gas velocity w_{og} the better results of the volumetric mass transfer coefficient correspond to CD 6 impeller. The A 315 impeller, which differs in the generated profile of the liquid circulation from the turbines, is less effective for the mass transfer processes in the agitated vessel (Karcz & Kiełbus-Rąpała, 2006).



Fig. 13. Comparison of the $k_L a = f(w_{og})$ for different single impellers working in the threephase system with solid concentration X = 2.5 mass %; (\Box) RT, n = 13.33 1/s; (\circ) CD 6, n = 13.33 1/s; (Δ) A 315, n = 12.50 1/s

The effect of upper agitator type on the volumetric mass transfer coefficient value in the gas-solid-liquid system is presented in Fig. 14.



Fig. 14. Comparison of values of $k_{L}a$ coefficient for two impeller configurations, working in gas-solid-liquid systems; $X \neq \text{const}$; n = 15 1/s; various values of superficial gas velocity $w_{\text{og}} \times 10^{-3} \text{ m/s}$

In the three-phase system that, which agitator ensure better conditions to conduct the process of gas ingredient transfer between gas and liquid phase, depends significantly on the quantity of gas introduced into the vessel. Comparison of values of k_1a coefficient for two impeller configurations, working in gas-solid-liquid systems with two different solid concentrations is presented in Fig. 14. At lower value of superficial gas velocity ($w_{og} = 1.71 \times 10^{-3} \text{ m/s}$) both in the system with lower solid particles concentration X = 0.5 mass % and with greater one: X = 2.5 mass %, higher of about 20 % values of volumetric mass transfer coefficient were obtained in the system agitated by means of the configuration with HE 3 impeller. With the increase of gas velocity w_{og} the differences in the values of k_La coefficient achieved for two tested impellers configuration significantly decreased. Moreover, in the system with lower solid concentration at the velocity $w_{og} = 3.41 \times 10^{-3} \text{ m/s}$, in the whole range of impeller speeds, k_La values for both sets of impellers were equal.

Completely different results were obtained when much higher quantity of gas phase were introduced in the vessel. For high value of w_{og} for both system including 0.5 and 2.5 mass % of solid particles, more favourable was configuration Rushton turbine – A 315. Using this set of agitators about 20 % higher values of the volumetric mass transfer coefficient were obtained, comparing with the data characterized the vessel with HE 3 impeller as an upper one (Fig. 12).

The data obtained for three-phase systems were also described mathematically. On the strength of 150 experimental points Equation (2) was formulated:

$$k_{\rm L}a = A \left(\frac{P_{\rm G-L-S}}{V_{\rm L}}\right)^b w_{og}^{\ c} \left(\frac{1}{1 + m_1 X^2 + m_2 X}\right)$$
(6)

The values of the coefficient *A*, m_1 , m_2 , and exponents *B*, *C* in this Eq. are collected in Table 6 for single impeller system and in Table 7 for the configurations of double impeller differ in a lower one. In these tables mean relative error $\pm \Delta$ is also presented. For the vessel equipped with single impeller Eq. (6) is applicable within following range of the measurements: $P_{\text{G-L-S}}/V_{\text{L}} \in <118$; 5700 W/m³>; $w_{og} \in <1.71 \times 10^{-3}$; 8.53×10⁻³ m/s>; $X \in <0.5$; 5 mass %>. The range of an application of this equation for the double impeller systems is as follows: $P_{\text{G-L-S}}/V_{\text{L}} \in <540$; 5960 W/m³>; $w_{og} \in <1.71 \times 10^{-3}$; 6.82×10⁻³ m/s>; $X \in <0.5$; 5 mass %>.

No.	Impeller	А	b	С	m_1	<i>m</i> ₂	±Δ, %	Exp. point
1.	Rushton turbine (RT)	0.031	0.43	0.515	-186,67	11.921	6.8	100
2.	Smith turbine (CD 6)	0.038	0.563	0.67	-388.62	23.469	6.6	98
3.	A 315	0.062	0.522	0.774	209.86	-11.038	10.3	108

Table 6. The values of the coefficient A, m_1 , m_2 and exponents b, c in Eq. (6) for single impeller systems (Kiełbus-Rąpała et al., 2010)

No.	Configuration	of impellers	А	b	С	m_1	m_2	±Δ, %
	lower	upper						
1.	CD 6	RT	0.164	0.318	0.665	0.361	8.81	3
2.	A 315	RT	0.031	0.423	0.510	-0.526	-8.31	4

Table 7. The values of the coefficient A, m_1 , m_2 and exponents b, c in Eq. (6) for double impeller systems (Kiełbus-Rąpała & Karcz, 2009)

The results of the $k_L a$ coefficient measurements for the vessel equipped with double impeller configurations differ in an upper impeller were described by Eq. 7.

$$k_L a = A \left(\frac{P_{G-L-S}}{V_L}\right)^B w_{og}{}^C \frac{1}{1+mX}$$
⁽⁷⁾

The values of the coefficient *A*, *m*, and exponents *B*, *C* in this Eq. for both impeller designs are collected in Table 8. The range of application of Eq. 2 is as follows: $Re \in \langle 9.7; 16.8 \times 10^4 \rangle$; $P_{G-L-S}/V_L \in \langle 1100; 4950 \text{ W/m}^3 \rangle$; $w_{og} \in \langle 1.71 \times 10^{-3}; 6.82 \times 10^{-3} \text{ m/s} \rangle$; $X \in \langle 0; 0.025 \rangle$.

No.	Configuration of in	mpellers	A	В	С	т	±Δ, %
	lower	upper					
1.	Rushton Turbine	A 315	0.103	0.409	0.695	4.10	4.8
2.	Rushton Turbine	HE 3	0.031	0.423	0.510	6.17	6.4

Table 8. The values of the coefficient *A* and exponents *B*, *C* in Eq. (7) (Kiełbus-Rąpała & Karcz 2010)

4. Conclusions

In the multi – phase systems the k_La coefficient value is affected by many factors, such as geometrical parameters of the vessel, type of the impeller, operating parameters in which process is conducted (impeller speed, aeration rate), properties of liquid phase (density, viscosity, surface tension etc.) and additionally by the type, size and loading (%) of the solid particles.

The results of the experimental analyze of the multiphase systems agitated by single impeller and different configuration of two impellers on the common shaft show that within the range of the performed measurements:

- 1. Single radial flow turbines enable to obtain better results compared to mixed flow A 315 impeller.
- 2. Geometry of lower as well as upper impeller has strong influence on the volumetric mass transfer coefficient values. From the configurations used in the study for gasliquid system higher values of $k_{\rm L}a$ characterized Smith turbine (lower)–Rushton turbine and Rushton turbine–A 315 (upper) configurations.
- 3. In the vessel equipped with both single and double impellers the presence of the solids in the gas-liquid system significantly affects the volumetric mass transfer coefficient $k_{L}a$. Within the range of the low values of the superficial gas velocity w_{og} , high agitator speeds *n* and low mean concentration X of the solids in the liquid, the value of the coefficient $k_{L}a$ increases even about 20 % (for single impeller) comparing to the data obtained for gas-liquid system. However, this trend decreases with the increase of both w_{og} and X values. For example, the increase of the $k_{L}a$ coefficient is equal to only 10 % for the superficial gas velocity $w_{og} = 5.12 \times 10^{-3} \text{ m/s}$. Moreover, within the highest range of the agitator speeds *n* value of the $k_{L}a$ is even lower than that obtained for gas-liquid system agitated by means of a single impeller.

In the case of using to agitation two impellers on the common shaft k_La coefficient values were lower compared to a gas-liquid system at all superficial gas velocity values.

- 4. The volumetric mass transfer coefficient increases, compared to the system without solids, only below a certain level of particles concentration. Introducing more particles, X = 2.5 mass % into the system causes a decrease of $k_L a$ in the system agitated by both single and double impeller systems.
- 5. In the gas-solid-liquid system the choice of the configuration (upper impeller) strongly depends on the gas phase participation in the liquid volume:

-The highest values of volumetric mass transfer coefficient in the system with small value of gas phase init were obtained in the vessel with HE 3 upper stirrer;

-In the three-phase system, at large values of superficial gas velocity better conditions to mass transfer process performance enable RT–A 315 configuration.

Symbols

а	length of the blade	m
В	width of the baffle	m
b	width of the blade	m
С	concentration	g/dm ³
D	inner diameter of the agitated vessel	m
d	impeller diameter	m

$d_{\rm d}$	diameter of the gas sparger	m
$d_{\rm p}$	particles diameter	m
e	distance between gas sparger and bottom of the vessel	m
Н	liquid height in the agitated vessel	m
h_1	off – bottom clearance of lower agitator	m
h_2	off – bottom clearance of upper agitator	m
i	number of the agitators	
I	number of baffles	
$k_{\rm L}a$	volumetric mass transfer coefficient	S ⁻¹
п	agitator speed	S ⁻¹
$n_{\rm ISG}$	critical impeller speed for gas – solid – liquid system	S ⁻¹
P	power consumption	W
R	curvature radius of the blade	m
t	time	s
$V_{\rm L}$	liquid volume	m ³
$V_{\rm g}^{\bullet}$	gas flow rate	m^3s^{-1}
w_{og}	superficial gas velocity (= $4 V_g / \pi D^2$).	m s-1
X	mass fraction of the particles	% w/w
Ζ	number of blades	
_		

Greek Letters

β	pitch of the impeller blade	deg
$ ho_{ m p}$	density of solid particles	kgm-3

Subscripts

G-L refers to gas – liquid system

G-S-L refers to gas - solid - liquid system

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