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Gas–Liquid Mass Transfer Coefficient in Stirred Tank Reactors

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nowledge of gas–liquid mass transfer rate and therefore volumetric gas–liquid mass transfer coefficient $(k_L a)$ is very important in designing or in the scale-up of gas–liquid stirred tank reactors (STR). In the literature $k_L a$ is correlated either by using dimensionless groups (Smith and Warmoeskerken, 1985; Smith, 1991) or energy input criterion (Smith et al., 1977; van't Riet, 1979; Hickman, 1988; Whitton and Nienow, 1993) based on Kolmogoroff's theory. Correlations based on dimensionless groups are of the form $k_L a = f(Fr, Fl_G, D/T, \text{ etc.})$ while correlations based on energy input criterion are of the form $k_L a = f(P/V_L)^{\alpha}$ $(V_G)^{\beta}$.

In our earlier work (Yawalkar et al., 2002), it has been shown that gas hold-up (ε_G) in STR can be reliably predicted by a correlation based on a relative dispersion parameter, $N/N_{cd'}$ over wide range of geometric conditions and operating parameters. Gas hold-up data available in the literature (Greaves and Barigou, 1990; Smith, 1991; Rewatkar et al., 1993) for larger tanks (T = 0.57 m to 2.7 m) equipped with either radial flow impeller (standard six-blade disc turbines (DTs)) or axial flow impellers (pitched bladed turbines (PBTs)) of different sizes and different sparger designs show unanimity with the proposed correlation.

In the present work it has been shown that similar to gas hold-up (ε_G), volumetric gas–liquid mass transfer coefficient ($k_L a$) can be correlated reliably over a wide range of geometric configurations and operating parameters (impeller speed/power input and gas rate) by a correlation based on the relative dispersion term N/N_{cd} .

 N_{cd} represents the minimum impeller speed at which all the liquid in a tank is in contact with the sparged gas. Extensive studies are available on N_{cd} in the literature (Nienow et al., 1977; Chapman et al., 1983; Rewatkar and Joshi, 1993). For any economical gas–liquid mixing operation in STR, minimum impeller speed for complete dispersion of the gas (N_{cd}) must be known, because at N_{cd} complete dispersion of the gas in liquid phase is obtained (Nienow et al., 1977; Chapman et al., 1983). Also, the mixing process at any impeller speed below N_{cd} results in incomplete dispersion of the gas. In other words, at an impeller speed below N_{cd} , there is very little or no gas in the region below the impeller. Therefore, the lower part of the tank is wasted, resulting in poor performance of the STR.

If the gassed power number (N_{PG}) or the ratio of power numbers under gassed conditions and ungassed condition (N_{PG}/N_{Po}) versus the Volumetric gas–liquid mass transfer coefficient (k_La) data available in the literature for larger tanks (T = 0.39 m to 2.7 m) have been analyzed on the basis of relative dispersion parameter, N/N_{cd} . It was observed that at a given superficial gas velocity (V_G) , k_La values were approximately the same irrespective of geometric configuration (size of the tank, type and size of the impellers, type of the sparger, etc.) at a particular N/N_{cd} . A single correlation based on N/N_{cd} is presented which shows satisfactory agreement with the k_La data of different workers.

Les données sur le coefficient de transfert de matière gaz–liquide volumétrique (k_La) fournies par la littérature scientifique pour des réservoirs de grande taille (T = 0,39 m à 2,7 m), ont été analysées relativement au paramètre de dispersion relative, N/N_{cd} . On a observé qu'à une vitesse de gaz superficielle donnée (V_G), les valeurs de kLa sont approximativement les mêmes peu importe la configuration géométrique (taille du réservoir, type et taille des turbines, type d'aérateur, etc.) pour un N/N_{cd} donné. On présente une corrélation simple basée sur N/N_{cd} qui montre un accord satisfaisant avec les données de k_La de différents chercheurs.

Keywords: STR, $k_L a$, $N_{cd'}$ relative dispersion (N/N_{cd}), V_G .

flow number (Fl_G) curve is plotted, a minimum occurs in the gassed power number at N_{cd} (Nienow et al., 1977; Bakker and van den Akker, 1994) (Figure 1). At N_{cd} the growing gas cavity covers the whole of the rear face of the blade, which causes drop in power input (Chapman et al., 1983). Studying the large amount of data generated from the experimental work incorporating wide range of system configurations (flat/dished bottom tanks with T = 0.29 to 1.83 m, D/T = 0.125 to 0.5, C/T = 0.25 to 0.5, ring spargers, pipe spargers and point spargers) and operating

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Table 1. Experimental details of the works on k _L a in large STR in coalescing (air-water) systems, studied in the present work								
Researchers	Т, т	Type of the impeller	Type of the sparger	D, m	V _G , m/s	N, rev/s	<i>P/V_L</i> , W/m ³	Method employed or measurement f of k _L a
Smith et al. (1977)	0.61, 0.91 and 1.83	6DT	Pipe	0.20, 0.30, 0.67 and 0.91	4×10^{-3} to 4.6×10^{-2}	0.9 to 8.5	20 to 5000	Dynamic method
Calderbank (1958; van't Riet 1979)	0.50	6DT	Orifice	0.166	5×10^{-3} to 4×10^{-2}		300-3500	Transmission
Chandrasekharan and Calderbank (1981)	1.22	6DT	Pipe	0.40	3.5×10^{-3} to 1.8×10^{-2}	1.75 to 4.43	94 to 1596	Dynamic method
Smith and Warmoeskerken (1985)	0.44	6DT	Ring	0.175	3×10^{-3} to 1.3×10^{-2}	3 to 9		Dynamic method
Linek et al. (1987)	0.29	6DT	Pipe	0.095	2.12×10^{-3} and 4.24×10^{-3}	4.17 to 14.17	100-3500	Dynamic method
Hickman (1988)	0.60 and 2	6DT		0.2 and 0.66	2×10^{-3} to 1.7×10^{-2}		50-3500	Steady state method
Smith (1991)	0.6, 2.4 and 2.7	6DT		0.198, 0.33, 0.79, 0.89 and 1.2	5×10^{-3} to 1.50×10^{-2} *	0.36 to 5.1*		Dynamic method
Whitton and Nienow (1993)	0.61 and 2.67	6DT	Ring	0.20 and 0.88	4.7×10^{-3} to 3.32×10^{-2}	1.42 to 5.75	210-1350	Dynamic method
Bakker and van den Akker (1994)	0.44	A315 PBT	Ring	0.177	1 × 10 ⁻²	3.43 to 9.16	100-1500	Dynamic method
Zhu et al. (2001)	0.39	6DT	Ring	0.13	1×10^{-3} to 7.5 × 10 ⁻³		100-1500	Dynamic method

* This range is derived from the $k_L a$ values given in the parity plot of Smith (1991), correlation proposed by him and the range of tank diameter (T) and D/T ratio studied by Smith (1991).



Figure 1. Gassed to ungassed power number ratio (N_{PG}/N_{Po}) versus gassed flow number (Fl_G) for gas–liquid STR.

conditions (P/V_L up to 8200 W/m³ and $V_G = 0.2$ to 4.6×10^{-2} m/s), Nienow et al. (1977) have given a correlation for N_{cd} in an STR equipped with a standard six-blade disc turbine (DT). The correlation proposed by Nienow et al. (1977) for DT is:

$$N_{cd} = \frac{4(Q_G)^{0.5}(T)^{0.25}}{(D)^2}$$
(1)

Equation (1) is very reliable for predicting N_{cd} for disc turbines (Yawalkar et al., 2002). Chapman et al. (1983) studied N_{cd} for different impellers such as four-blade mixed flow impellers pumping up (MFUs) and pumping down (MFDs), six-blade MFUs and MFDs, axial flow impellers pumping up (AFUs) and pumping down (AFDs), disc turbines (DTs) and angular-bladed disc turbines (ADTs). Rewatkar and Joshi (1993) have extensively studied N_{cd} in an STR equipped with six-blade downflow pitched turbines. N_{cd} can be determined by simple methods such as visual observation, from the power input curve, and from mixing time studies (Rewatkar and Joshi,

Table 2. Correlations proposed by different workers to estimate volumetricgas-liquid mass transfer coefficient ($k_L a$) in STR and used in the present work.						
Researchers	Correlation Proposed					
Calderbank (1958) van't Riet ⁽ 1979)	$k_L a = 0.026 \ (P/V_L)^{0.4} (V_G)^{0.5}$					
Smith et al. (1977)	$k_L a = 0.01 \ (P/V_L)^{0.475} (V_G)^{0.4}$					
Smith and Warmoeskerken (1985)	BLC regime: $k_L a = 1.1 \times 10^{-7} (H_G)^{0.6} (Re_N)^{1.1} (N)$ ALC regime: $k_L a = 1.6 \times 10^{-7} (H_G)^{0.42} (Re_N)^{1.02} (N)$					
Linek et al. (1987)	$k_L a = 4.95 \times 10^{-3} \ (P/V_L)^{0.593} (V_G)^{0.4}$					
Hickman (1988)	For $T = 0.60 \text{ m}$ $k_l a = 0.043 (P/V_l)^{0.4} (V_G)^{0.57}$ For $T = 2 \text{ m}$ $k_l a = 0.027 (P/V_l)^{0.54} (V_G)^{0.68}$					
Smith (1991)	$ \begin{array}{l} k_L a = 1.25 \times 10^{-4} (D/T)^{2.8} \\ ({\rm Fr})^{0.6} (Re_N)^{0.7} \\ (H_G)^{0.45} (D/g)^{-0.5} \end{array} $					
Whitton and Nienow (1993)	$k_L a = 0.57 \ (P/m)^{0.4} (V_G)^{0.55}$					
Zhu et al. (2001)	$k_L a = 0.031 \ (P/V_L)^{0.4} (V_G)^{0.5}$					

1993). The last two are objective methods but show good agreement with the first, which is subjective (Rewatkar and Joshi, 1993).

Relative Gas Dispersion (N/N_{cd})

Turbulent fluctuations in the gas-liquid dispersion control the drag on the bubble and hence the bubble size (Calderbank et al., 1959; Hinze, 1955; Hughmark 1974; Miller, 1974). The bubbles break up when the hydrodynamic stresses (Reynolds

Table 3. Hughmark's (1980) correlation for impeller power input under aerated condition (P_G/P_O) for standard six bladed disc turbine is based on following experimental details.

Tank diameter, <i>T</i> (m) <i>D</i> /T	0.21 to 3.33 0.33 to 0.576
C/T	0.33 to 0.67
H/T	0.75 to 1.87
V _G (max.)	0.053 m/s
Blade width (w),m	0.2D
μ (mPa·s)	0.8 to 28
ρ (kg/m³)	870 to 1600
σ (g/s ²)	25 to 72
P_G/P_O	0.31 to 0.8

stresses) outweigh the surface tension force (Hinze, 1955; Calderbank et al., 1959; Walter and Blanch ,1986). Walter and Blanch (1986) gave the correlation of maximum stable bubble size on the assumption that bubble break-up is caused by the eddies of the same scale as the maximum stable bubble size. Parthasarathy and Ahmed (1991) have discussed the process of bubble break-up in a turbulent field using the criterion of power dissipation according to Kolmogoroff's theory. Accordingly, they concluded that bubbles are broken by eddies of the inertial subrange.

The turbulence increases the drag on the bubbles (Hughmark, 1974) thereby reducing the bubble rise velocity. These bubbles with reduced rise velocity can be more easily entrained in the downward liquid flow generated by the impeller. Thus, the overall effect of increasing turbulence results in increasing gas hold-up (ε_G) and therefore gas–liquid interfacial surface area (*a*) in STR.

At N_{cd} complete dispersion of the sparged gas is achieved. Therefore, from the above discussion it can be concluded that at N_{cd} the bubble size generated in the vicinity of the impeller has a buoyancy lesser than the downward drag caused by the downward liquid flow and the bubbles are pulled down in the lower

Table 4. Correlations obtained for the $k_{L}a$ data of different workers based on relative dispersion parameter, N/N_{cd} .							
Researchers	Correlation based on the term, N/N _{cd}	R ²	Standard error	Number of data analyzed			
Calderbank (1958), van't Riet (1979)	$k_L a = 2.76 \ (N/N_{cd})^{1.14} (V_G)^{0.97}$	1	-	7			
Smith et al. (1977)	$k_L a = 2.42 \ (N/N_{cd})^{1.38} \ (V_G)^{0.96}$	0.98	0.07	41			
Chandrasekharan and Calderbank (1981)	$k_L a = 2.68 \ (N/N_{cd})^{1.15} \ (V_G)^{0.96}$	0.99	0.04	11			
Smith and Warmoeskerken (1985)	$k_L a = 12.63 (N/N_{cd})^{1.54} (V_G)^{1.27}$	0.99	0.02	15			
Linek et al. (1987)	$k_L a = 5.16 \ (N/N_{cd})^{1.69} \ (V_G)^{1.09}$	1	-	7			
Hickman (1988)	$k_L a = 4.28 \ (N/N_{cd})^{1.35} \ (V_G)^{1.04}$	0.97	0.12	21			
Smith (1991)	$k_L a = 6.48 \ (N/N_{cd})^{1.44} \ (V_G)^{1.12}$	0.99	0.02	32			
Whitton and Nienow (1993)	$k_L a = 3.50 \ (N/N_{cd})^{1.17} \ (V_G)$	0.99	0.02	13			
Bakker and van den Akker (1994)	$k_L a = 0.030 \ (N/N_{cd})^{1.42}$	0.93	0.10	16			
Zhu et al. (2001)	$k_L a = 3.31 (N/N_{cd})^{1.14} (V_G)^{0.97}$	1	-	15			

Researchers	Type of the impeller	<i>T,</i> m	D/T	$V_G imes 10^2$, m/s	N _{cd} , rev/s	N/N _{cd}	<i>P/V_L,</i> W/m ³	$k_L a \times 10^2$, 1/s (observed)	$k_L a \times 10^2,$ 1/s (Smith, 1991)	$k_L a \times 10^2$, 1/s (Equation 4)
Smith and	6DT	0.44	0.40	0.65	3.36	0.89		1.72 (ALC)	2.03	1.86
Warmoeskerken				0.46	2.81	1.77		3.11 (BLC)	3.63	3.57
(1985)				1.31	4.75	1.68		11.0 (ALC)	11.50	9.42
				0.98	4.11	2.18		11.8 (BLC)	12	10.37
Smith (1991)	6DT	0.60	0.33	1	2.08	0.51		1.48		1.27
						1.51		7.11		6.15
		2.4	0.50	0.75	0.63	0.86		2.13		2.03
						2.04		7.42		7.14
		2.7	0.33	1.25	1.72	0.75		3.10		2.76
						1.44		8.01		7.19
			0.50	1.50	1.89	1		5.84		5.05
				1.13	0.71	1.40		6.89		6.17
Smith et al. (1977)	6DT	0.91	0.33	2	4.79	1.04	1238	6.1	9.03	7.12
				3	5.86	0.59	405	4.25	4.89	4.71
		0.61	0.33	1.70	6.04	0.82	551	3.92	4.43	4.30
						1.40	2501	8.05	11.66	9.37
Whitton and Nienow	ı 6DT	0.61	0.33	0.7	3.94	1	339	2.41	2.50	2.35
(1993)		2.67	0.33	1.19	1.70	0.98	523	3.84	4.30	3.88
				2.5	2.46	0.67	434	5.37	5.91	4.74
Hickman (1987)	6DT	2	0.33	1	1.93	1.54	1606	6.34	6.79	6.36
· · · ·				1.70	2.52	0.59	195	2.91	3.16	2.66
						1.03	935	6.79	7.9	5.94
Calderbank (1958),	6DT	0.50	0.33	1.50	6.67	1.55	134	2.25	2.37	1.95
van't Riet (1979)				4.0	10.89	1.10	3507	13.61	22.32	15.40
Linek et al. (1987)	6DT	0.29	0.33	0.212	3.79	1.58	328	1.31	1.22	1.39
	-			0.424	5.36	0.77	98	0.84	0.97	0.98
Chandrasekharan an	d 6DT	1 22	0 33	0.35	1 65	1.05		1 26	1 1 5	1 27
Calderbank (1981)				1.80	3.76	0.89		4.49	6.25	5.08
				1.10	2.94	1.32		4.84	6.21	5.53
Bakker and	A315	0.44	0.40	1	6.1	0.56	92	1.50	3.05	1.44
van den Akker (1994)					1.28	787	4.75	10.12	4.84
	PBT	0.44	0.40	1	4.99	0.76	153	1.75	3.70	2.20
						1.37	822	4.87	8.34	5.30

part of the tank. Thus, at N_{cd} the turbulence intensity is such that complete dispersion of the gas in all the liquid is achieved.

At any impeller speed N the ratio u'/u'_{cd} gives the turbulence intensity at impeller speed N relative to that at the impeller speed N_{cd} for a given gas flow rate and geometric configuration (Jadhav and Pangarkar, 1991; Kushalkar and Pangarkar, 1994). As drag on the gas bubble and therefore bubble size and hence a depends upon turbulence intensity, volumetric gas–liquid mass transfer coefficient (k_La) also depends upon turbulence intensity. Therefore $k_La/k_La_{cd} = f(u/u_{cd})$. Here, k_La_{cd} is the volumetric gas–liquid mass transfer coefficient at minimum impeller speed for complete dispersion of the gas, N_{cd} . Deshpande (1988) indicated that turbulence intensity u, is approximately proportional to impeller speed *N*. Thus, $k_L a/k_L a_{cd} = f(N/N_{cd})$. The ratio N/N_{cd} represents relative dispersion of the gas and, therefore, relative volumetric gas–liquid mass transfer coefficient, $k_L a/k_L a_{cd'}$ in STR for a given system configuration and gas input rate.

As discussed in our earlier work (Yawalkar et al., 2002), N/N_{cd} represents relative amount of gas retained in the liquid at impeller speed N with respect to that at impeller speed N_{cd} . Therefore, it seems logical to define volumetric gas–liquid mass transfer coefficient (k_La) in terms of a relative dispersion parameter N/N_{cd} . Jadhav and Pangarkar (1991) have shown that particle–liquid mass transfer coefficient (k_{SL}) data of different workers show unanimity when compared on the basis of

relative particle suspension parameter, N/N_s . N_s is the minimum impeller speed for complete suspension of the particles.

In the present work, $k_L a$ data available in the literature for large STRs (T = 0.39 m to 2.7 m) equipped with standard disc turbines for which N_{cd} values can be obtained from Equation (1) were studied. Also, $k_L a$ data for a lightnin A315 hydrofoil impeller and a pitched bladed turbine (PBT) for which N_{cd} values can be obtained from the power input versus flow number plots, provided by respective authors, were analyzed on the basis of N/N_{cd} .

Data Available on $k_L a$ in the Literature for Larger Stirred Tank Reactors and Used in the Present Study

Table 1 summarizes the experimental details of the studies on $k_L a$ in larger STRs whose data have been used in the present work. Table 2 gives the correlations proposed by these workers for predicting $k_L a$ in an air-water system. $k_L a$ data of different workers were generated by applying the respective correlation strictly over respective geometric configuration and operating parameters used in that specific study.

Most of the workers correlated their $k_L a$ data either in terms of dimensionless groups or power input criterion. While Chandrasekharan and Calderbank (1981) and Bakker and van den Akker (1994) have given their $k_L a$ data for different geometric and operating conditions in tabular and graphical forms, respectively.

Studies Based on Dimensionless Groups

Plotting the Froude number (*Fr*) versus gas flow number (Fl_G) Smith and Warmoeskerken (1985) and Smith (1991) defined flow regimes in gas–liquid STRs, such as vortex clinging cavities regime (high *Fr* and low Fl_G), large cavities regime (high *Fr* and high Fl_G), flooded region (low *Fr* and high Fl_G) and gas recirculation regime (very high *Fr* and low Fl_G).

Smith and Warmoeskerken (1985) considered their gas hold-up data and $k_L a$ data separately in two groups: (i) clinging cavity regime or before large cavity (BLC) regime; and (ii) higher gas loading regime or after large cavity (ALC) regime in STR. Smith (1991) in his systematic study on gas dispersion behavior, studied gas hold-up behavior and volumetric gas–liquid mass transfer rates in STRs equipped with DT. Smith (1991) observed that his $k_L a$ data for T = 0.60 m, 2.4 m and 2.7 m can be correlated satisfactorily by a correlation based on dimensionless groups.

The N/N_{cd} versus $k_L a$ data for both these studies were directly derived by applying the correlations over specific range of geometric configurations and operating parameters for a respective study.

Studies Based on Power Input Criterion

Smith et al. (1977) studied $k_L a$ in large STRs, T = 0.60, 0.91 and 1.83 m equipped with different impellers with majority of the data based on DT of different sizes. Their correlation for air-water system is of the form $k_L a = f (P/V_L)^{\alpha} (V_G)^{\beta}$ with maximum deviation of ±20%. Since reliable correlation for N_{cd} for such large-scale STRs equipped with DT is available (Equation 1), $k_L a$ data of Smith et al. (1977) for DTs could be converted to the N/N_{cd} form.

van't Riet (1979) carefully studied the available data on $k_L a$ and indicated that correlation of data from various reports on mass transfer for disc turbines is possible with power input per unit volume and gas superficial velocity. In his correlation treatment for coalescing media (air–water) van't Riet (1979) used large tank data from the work of Calderbank (1958) and Smith et al. (1977). In this work $k_L a$ data of Calderbank (1958) were obtained using van't Riet's correlation.

Linek et al. (1987) critically reviewed inconsistencies in various dynamic methods for measuring volumetric gas–liquid mass transfer coefficient and suggested a correct dynamic method. However, they showed that for coalescing systems (air–water) various incorrect techniques give practically the same k_La values (Linek et al., 1987). Linek et al. (1987) correlated their k_Ia data on T = 0.29 m using power input criterion.

Hickman (1988) used a steady-state technique to determine $k_L a$ by measuring rate of decomposition of hydrogen peroxide to oxygen in the liquid phase. He correlated $k_L a$ data on STRs of T = 0.60 m and 2 m using the power input criterion.

Whitton and Nienow (1993) studied gas hold-up behavior and gas–liquid mass transfer rates in STRs of diameter 0.61 m and 2.67 m with disc turbine of D = T/3. They correlated gas hold-up and k_La data through power input per unit mass and superficial gas velocity.

Recently, Zhu et al. (2001) studied the mass transfer performance of several commonly used impellers for an air-water system with T = 0.39 m. They concluded that data for DTs as well as other impellers such as A315, PBT and a concave blade turbine can be reasonably correlated by an equation of the form $k_L a = f(P/V_L)^{\alpha} (V_C)^{\beta}$ with a maximum deviation of ±30%. Data of Zhu et al. (2001) for DT were analyzed in the present work on the basis of N/N_{cd} .

The correlation presented by Hughmark (1980) for power input in gas–liquid dispersion by a six-bladed disc turbine over a wide range of system configurations and operating parameters (Table 3) is:

$$\frac{P_G}{P_O} = 0.1 \left(\frac{Q_G}{NV_L}\right)^{-0.25} \left(\frac{N^2 D^4}{g D_i V_L^{2/3}}\right)^{-0.2}$$
(2)

Equation (2) is based on 391 data with standard deviation of 0.117. This correlation is considered to be reliable (Hughmark, 1980; van't Riet and Tramper, 1991; Yawalkar et al., 2002). Therefore, $k_L a$ versus impeller speed (*N*) data for different superficial gas velocities and power input for all the above discussed literature on DTs, based on power input criterion obtained by using Equation (2). Thereafter, $k_L a$ versus *N*/*N*_{cd} data were obtained by using Equation (1).

Data Obtained from Tabular and Graphical Results

Chandrasekharan and Calderbank (1981) measured $k_L a$ by dynamic method in an STR of T = 1.22 m. Analyzing $k_L a$ values obtained by Figueiredo and Calderbank (1979) for T = 0.91 m and their own data, Chandrasekharan and Calderbank (1981) concluded that for a given specific power input and gas superficial velocity there is an optimum size of tank with respect to $k_L a$ for a particular geometry. In other words, for a given P/V_L and V_G , as tank diameter increases, $k_L a$ also increases up to certain tank diameter, but at a higher tank diameter above the critical size , $k_L a$ decreases with tank diameter. However, Linek et al. (1987) have shown that the $k_L a$ measurement method used by Figueiredo and Calderbank (1979) is erroneous and, therefore, their $k_L a$ values for T = 0.91 are not correct.

Chandrasekharan and Calderbank (1981) presented their results in tabular form (Table 2 of their paper), which have been



Figure 2. Volumetric gas–liquid mass transfer coefficient $(k_l a)$ data fit by Equation (4). o; $k_l a$ data of Smith et al. (1977): •; $k_l a$ data of Calderbank (1958) van't Riet (1979): *; $k_l a$ data of Chandrasekharan and Calderbank (1981): **I**; $k_l a$ data of Smith and Warmoeskerken (1985): +; $k_l a$ data of Linek et al. (1987): ×; $k_l a$ data of Hickman (1988): ·; $k_l a$ data of Smith (1991): Δ ; $k_l a$ data of Whitton and Nienow (1993): **(a)**; $k_l a$ data of Bakker and van den Akker (1994): **(a)**; $k_l a$ data of Zhu et al. (2001).

converted in the form of k_{la} versus N/N_{cd} using Equation (1) in the present work.

Bakker and van den Akker (1994) studied gas dispersion characteristics and gas-liquid mass transfer in STR equipped with axial flow impellers, such as PBT, A315 impeller and Leeuwrik impeller. They have presented an excellent discussion on prevailing gas flow patterns and impeller hydrodynamics for these impellers for different operating and geometric conditions. Bakker and van den Akker (1994) reported that an A315 impeller is the most suitable axial flow impeller with respect to gas dispersion capacity and stability of the system. They observed that for a given power input and gas superficial velocity PBT yields lower gas hold-up and $k_i a$ than A315. Bakker and van den Akker (1994) have given plots of N_{PG} versus Fl_c for A315 impeller and PBT with large ring sparger (Figures 9 and 10 of their paper). From these plots minimum impeller speed for complete gas dispersion (N_{cd}) for both these impellers and respective impeller speed (N) for different power inputs were obtained. N_{cd} values for PBT and A315 impellers were 4.99 rev/s and 6.1 rev/s, respectively.

Using the graphs of P/V_L versus $k_L a$ (Figure 17 of their paper) for the A315 impeller and large-ring sparger, $k_L a$ versus N/N_{cd} data for the A315 impeller were obtained. Data of Bakker and van den Akker (1994) show that for a given P/V_L and V_G for A315 impeller, a small-ring sparger (SRS) gives slightly higher $k_L a$ values (10% to 15%) than a large-ring sparger (LRS). They also observed a similar 10% to 15% deviation of the gas hold-up data for an A315 impeller with both these spargers. Contrary



Figure 3. Correlating line plot for Equation (6). **Legends:** ×; Data of Smith et al. (1977): •; Data of Chandrasekharan and Calderbank (1981): ▲; Data of Hickman (1988): o; Data of Smith (1991): _; Data of Whitton and Nienow (1993).

to the results of the A315 impeller, Bakker and van den Akker (1994) observed an insignificant effect of sparger types on gas hold-up for PBT. However, considering the close relationship between gas hold-up and k_La , it can be concluded that the effect of sparger design on k_La for PBT in the work of Bakker and van den Akker (1994) is negligible or very small. They did not report any effect of sparger design on k_La for PBT. Bakker and van den Akker (1994) provided a k_La versus P/V_L plot for PBT with SRS and not for LRS. Thus, k_La values obtained from their plot of P/V_L versus k_La for PBT with SRS can be approximately considered to be the same as those for PBT with LRS for respective P/V and gas flow rate. Therefore, using the graph of P/V_L versus k_La (Figure 19 of their paper) for PBT and SRS, data of k_La versus N/N_{cd} for PBT have been obtained in the present work.

Results and Discussion

In our previous work it was observed that the correlation of the form $\varepsilon_G = f(N/N_{cct'} \text{ vvm}, T, D/T)$ shows good fit to the gas hold-up data for larger tanks available in the literature over a wide range of system and operating conditions. Dependence of ε_G on T and D/T is rather small as compared to dependence on the terms N/N_{ccl} and vvm. Here, vvm is volume of gas sparged per unit volume of liquid per minute. Therefore, it was thought desirable to correlate the $k_L a$ data obtained from different studies discussed earlier in the form of $k_L a = f(N/N_{cct'} \text{ vvm}, T, D/T)$. Dependence of $k_L a$ on D/T at a given $N/N_{cct'}$ vvm and T was found to be negligible. The expression obtained after the regression is:

$$k_1 a = 0.0558 (N/N_{cd})^{1.464} (\text{vvm}) (T)^{1.05}$$
 (3)

Equation (3) is based on a total of 178 data points and has R^2 value of 0.96 with standard deviation of 0.13. The exponent of *T* in Equation (3) is very close to 1. Therefore, Equation (3) can be written in the form:

$$k_L a = 3.35 (N/N_{cd})^{1.464} (V_G)$$
⁽⁴⁾

Even though application of Equation (4) instead of Equation (3) to estimate $k_L a$ increases the deviation of predicted $k_L a$ from actual $k_L a$ values, the increases in deviation is very small (2% to 4%). $k_L a$ data along with the correlating line given by Equation (4) are shown in Figure 2. It can be seen that most of the data except few data points of Smith et al. (1977) lie within ±22% of the correlating line. Hughmark's (1980) correlation was used to determine impeller speed *N* for a given power input for some data sets. Standard deviation of this correlation given by Hughmark (1980) is 0.117. Therefore, it is likely that part of the deviation of the data points around the correlation given by Equation (4) arises from the use of Hughmark's correlation.

Table 4 indicates that $k_i a$ data of different workers can be correlated satisfactorily by the relative dispersion parameter, N/N_{cd} . To study the usefulness of the proposed correlation in the present work, Equation (4) is compared with the dimensionless correlation proposed by Smith (1991) for the data studied in this work. Smith's (1991) dimensionless correlation is based on a wide range of system configurations using DT. Table 5 shows some data based on the relative dispersion parameter N/N_{cd} along with the $k_{l}a$ values estimated by the correlation of Smith (1991) and Equation (4). It can be concluded that the correlation of Smith (1991) shows reasonable agreement (within 33%) with most of the data for disc turbines. However, the A315 impeller and PBT data correlation of Smith (1991) predicts $k_i a$ values almost two times higher than that observed by Bakker and van den Akker (1994). The correlation proposed in Equation (4) in the present work, on the other hand, shows satisfactory agreement (within 22%) with the data for DT, A315 impeller and PBT. Even though disc turbines are high shear-low pumping impellers while A315 and PBT impellers are low shear-high pumping, the k_1a data of all these impellers show good agreement with each other when correlated on the basis of N/N_{cd} .

It should be noted that Equation (4) is independent of the scale of the reactor, type, size and position of the impeller and sparger. It indicates that at a particular superficial gas velocity and $N/N_{cd'}$ k_La is same irrespective of system configuration. It can be argued that in Equation (4) all the geometric details of the system are defined by the term $N_{cd'}$ which is a characteristic of particular system configuration (*T*, *D*/*T*, type of the impeller, etc.). Jadhav and Pangarkar (1991) and Kushalkar and Pangarkar (1994) have shown that in solid–liquid/solid–liquid–gas STR the term N_s (minimum impeller speed for complete dispersion of the solid particles) defines geometric details of the system and therefore a correlation based on N/N_s , for predicting $k_{SI'}$ is geometric configuration independent.

The R.H.S. of Equation (4) has unit m/s while the L.H.S. has unit 1/s. This implies that, to make Equation (4) dimensionally consistent, the L.H.S. of Equation (4) should be multiplied by a term that has a unit of length. Considering the close dependence of gas-liquid mass transfer coefficient on the gas bubble size (d_B) which has the unit of length, the k_La data was regressed in the form of k_Lad_B/V_G versus N/N_{cd} . The correlation of Calderbank (1958) was used to determine values of the Sauter mean bubble diameter (d_R) in a tank for different system and operating conditions studied in this work. The correlation given by Calderbank (1958) for pure solutions is:

$$d_B = 4.15 \frac{\sigma^{0.6}}{(P/V_L)^{0.4} \rho_L^{0.2}} \varepsilon_G^{0.5} + 0.0009$$
⁽⁵⁾

The gas hold-up data required in this expression were obtained from the gas hold-up correlations given by respective authors and, if not, then derived from the gas hold-up correlation proposed in our previous work (Yawalkar et al., 2002). The expression obtained is:

$$k_1 a d_8 = 0.0186 (N/N_{cd})^{0.82} (V_C)$$
 (6)

Equation (5) has an R^2 value 0.88 with standard deviation of 0.14. Figure 3 shows the large tank data for the works of Smith et al. (1977), Chandrasekharan and Calderbank (1981), Hickman (1988), Smith (1991) and Whitton and Nienow (1993) in the form of Equation (6). Taking into account the deviation introduced due to the application of Hughmark's (1980) and Calderbank's (1958) correlations to generate impeller speed (N) and bubble diameter (d_B) data, respectively, at a given power input per unit volume (P/V_l), the agreement of the data points around a correlating line is satisfactory (30%).

Significance of the Terms vvm and *T* in Equation (4)

Equation (4), which arises out of Equation (3), is distinctly different in its dependence on gas flow rate as compared to the correlation for gas hold-up presented in our earlier paper (Yawalkar et al., 2002). Yawalkar et al. (2002) found that the use of vvm in the correlation for ε_G makes the correlation independent of scale. In the present correlation for $k_L a$ the terms on the R.H.S. of Equation (3), (vvm), *T* ultimately reduces to V_G and Equation (4) is obtained. This can be viewed as an inadequacy of the concept of vvm. However, in the following an explanation is given which indicates that the direct dependence of $k_L a$ on V_G arises out of complex dependence of the terms k_L and *a* on vvm and *T* and not because of any inadequacy of the vvm concept.

Studying large amount of data of different workers Hughmark (1980) has given a correlation for gas–liquid interfacial area per unit volume term, *a*, in STR. The correlation given by Hughmark (1980) is:

$$a = 1.38 \left(\frac{g\rho_L}{\sigma}\right)^{0.50} \left(\frac{Q_G}{NV_L}\right)^{0.33} \left(\frac{N^2 D^4}{gwV_L^{0.66}}\right)^{0.592} \left(\frac{d_B N^2 D^4}{\sigma V_L^{0.66}}\right)^{0.187}$$
(7)

This correlation was applied to the system configuration and operating parameters of different workers (given in Table 1) to estimate values of *a* in their respective works. Then the values of *a* obtained were regressed as $a = f(N/N_{cd'}, vvm, T)$. The expression obtained is:

$$a = 7.47 \ (N/N_{cd})^{1.23} (vvm)^{0.95} (T)^{0.30} \tag{8}$$

The R^2 value of Equation (8) is 0.99 with standard deviation of 0.05. Using this dependence of *a* on $N/N_{cd'}$ vvm and *T* and that of $k_L a$ given in Equation (3) (i.e. dividing Equation 3 by Equation 8) it can be noted that $k_L \propto (N/N_{cd})^{0.234}$ (vvm)^{0.05} $(T)^{0.75}$. Thus, k_L and *a* have different functional relationships with $N/N_{cd'}$ vvm and *T* but the combined dependence is such that ultimately an equation Equation(4) is obtained which directly involves V_G . Therefore, it can be concluded that the functional dependence Equation (4) proposes, $k_L a \propto (vvm)$. (7) (which eventually is equal to V_G) arises out of the combined effect of dependence of the terms k_L and a on vvm and tank diameter, T.

Conclusions

In this study, data available in the literature on volumetric gas–liquid mass transfer coefficient ($k_L a$) for larger STRs (T = 0.39 m to 2.7 m) have been analyzed in the form of $k_L a$ versus relative dispersion parameter, N/N_{cd} . An attempt has been made to provide a single correlation based on N/N_{cd} for $k_L a$ data obtained by different researchers for a coalescing (air–water) system. It was observed that, at a given superficial gas velocity (V_G), $k_L a$ values were approximately the same irrespective of geometric configuration (size of the reactor, type and size of the impeller, sparger type, etc) for a particular N/N_{cd} . The correlation obtained was:

 $k_1 a = 3.35 (N/N_{cd})^{1.464} (V_C)$

Almost all the $k_l a$ data points were found to lie within 22% of the correlating line.

Thus, results of the previous work on gas hold-up (Yawalkar et al., 2002) and the present work on $k_L a$ indicate that the relative dispersion parameter is useful in reliably correlating/ estimating hydrodynamic and mass transfer properties of gas–liquid STR.

Nomenclature

- *a* gas–liquid interfacial area per unit volume, (m²/m³)
- C clearance of the impeller from the bottom of STR, (m)
- D impeller diameter, (m)
- D_i width of the impeller blade, (m)
- Fl_G gassed flow number, Q_G/ND^3
- Fr Froude number, N^2D/g
- g gravitational acceleration, (=9.81 m/s²)
- *H* height of clear liquid in STR, (m)
- $k_L a$ volumetric gas–liquid mass transfer coefficient, (1/s)
- k_{SL} solid–liquid mass transfer coefficient, (m/s)
- m mass of liquid in STR, (kg)
- N impeller speed, (rev/s)
- N_{cd} minimum impeller speed for complete dispersion of the sparged gas, (rev/s)
- *N_s* minimum impeller speed for complete suspension of all the solid particles in STR, (rev/s)
- N_{PG} power number of the impeller in presence of gas, $P_G/\rho N^3 D^5$
- N_{p_0} power number of the impeller in absence of gas, $P/\rho N^3 D^5$
- N_R impeller speed at which recirculation of the sparged gas within the liquid starts, (rev/s)
- P_O power input in absence of gas, (W)
- P_G power input in presence of gas, (W)
- Q_G volumetric gas flow rate, (m³/s)
- Re_N Reynolds number based on impeller speed, $ND^2\rho/\mu$
- T inside diameter of STR, (m)
- *u'* turbulence intensity, (m/s)
- V_G superficial gas velocity, (m/s)
- V_L volume of the liquid in the reactor, (m³)
- *w* impeller blade width, (m)

Greek Symbols

- ϵ_G fractional gas hold up
- μ viscosity, (Pa·s)

- ρ density, (kg/m³)
- σ surface tension, (N/m)

Subscripts

- cd complete dispersion
- G gas
- L liquid
- S complete suspension
- SL solid-liquid

References

- Bakker, A. and H.E.A. Van Den Akker, "Gas-Liquid Contacting with Axial Flow Impellers", Trans. Inst. Chem. Eng. **72**, 573-582 (1994).
- Calderbank, P.H., "Physical Rate Processes in Industrial Fermentation. Part I: The Interfacial Area in Gas–Liquid Contacting with Mechanical Agitation", Trans. Inst. Chem. Eng. **36**, 443–463 (1958).
- Calderbank, P.H., "Physical Rate Process in Industrial Fermentation. Part II: Mass Transfer Coefficients in Gas–Liquid Contacting with and without Mechanical Agitation", Trans. Inst. Chem. Eng. **38**, 173–185 (1959).
- Chandrasekharan, K. and P.H. Calderbank, "Further Observations on the Scale-up of Aerated Mixing Vessels", Chem. Eng. Sci. **36**, 819–823 (1981).
- Chapman, C.M, A.W. Nienow, M. Cooke and J.C. Middleton, "Particle–Gas–Liquid Mixing in Stirred Vessels. Part II: Gas–Liquid Mixing", Chem. Eng. Res. Des. **61**, 82–95 (1983).
- Deshpande, G.B., "Fluid Mechanics in Mechanically Agitated Contactors", M.Chem.Eng. Thesis, University of Bombay, Bombay, India (1988).
- Dutta, N.N. and V.G. Pangarkar, "Particle–Liquid Mass Transfer in Multi-impeller Agitated Contactors", Chem. Eng. Commun. **129**, 109–121 (1994).
- Figueiredo, M. and P.H. Calderbank, "The Scale-up of Aerated Mixing Vessels for Specified Oxygen Dissolution Rates", Chem. Eng. Sci. 34, 1333–1338 (1979).
- Greaves, M. and M. Barigou, "Estimation of Gas Hold up and Impeller Power in a Stirred Vessel Reactor", in "Fluid Mixing III", Inst. Chem. Eng., Int. Chem. Eng. Symp. Series No: 108, (1990), pp. 235–255.
- Hickman, A.D., "Gas–Liquid Oxygen Transfer and Scale-up. A Novel Experimental Technique with Results for Mass Transfer in Aerated Agitated Vessels", in "Proceedings of 6th European Conference on Mixing", Pavia, Italy (1988), pp. 369–374.
- Hinze, J.O., "Fundamentals of the Hydrodynamic Mechanism Splitting in Dispersion Process", AIChE J. 1, 289–295 (1955).
- Hughmark, G.A., "Hydrodynamics and Mass Transfer for Suspended Solid Particles in a Turbulent Liquid", AIChE J. **20**, 202–204 (1974).
- Hughmark, G.A., "Power Requirements and Interfacial Area in Gas-Liquid Turbine Agitated Systems", Ind. Eng. Chem. Process Des. Dev. **19**, 641–646 (1980).
- Jadhav, S.V. and V.G. Pangarkar, "Particle–Liquid Mass Transfer in Mechanically Agitated Contactors", Ind. Eng. Chem. Res. **30**, 2496–2503 (1991).
- Kushalkar, K.B. and V.G. Pangarkar, "Particle–Liquid Mass Transfer in Mechanically Agitated Three Phase Reactors", Ind. Eng. Chem. Res. 33, 1817–1820 (1994).
- Linek, V., V. Vacek and P. Benes, "A Critical Review and Experimental Verification of the Correct use of the Dynamic Method for the Determination of Oxygen Transfer in Aerated Agitated Vessels to Water, Electrolyte Solutions and Viscous Liquids", Chem. Eng. J. 34, 11–34 (1987).

- Miller, D.N., "Scale-up of Agitated Vessels. Gas–Liquid Mass Transfer", AIChE J. 20, 445–453 (1974).
- Nienow, A.W., D.J. Wisdom and J.C. Middleton, "Effect of Scale and Geometry on Flooding, Recirculation and Power in Gassed Stirred Vessels", in "Proceedings of 2nd European Conference on Mixing", Cambridge, UK (1977), F1, pp. 1–16.
- Parthasarathy, R. and N. Ahmed, "Gas Holdup in Stirred Vessels: Bubble Size and Power Input Effects", in "Proceedings 7th European Congress on Mixing", Brugge Belgium (1991), pp. 295–302.
- Rewatkar, V.B. and J.B. Joshi, "Effect of Sparger Design on Gas Dispersion in Mechanically Agitated Gas/Lquid Reactors", Can. J. Chem. Eng. **71**, 278–291 (1993).
- Rewatkar, V.B., A.J. Deshpande, A.B. Pandit and J.B. Joshi, "Gas Hold-up Behavior of Mechanically Agitated Gas–Liquid Reactors using Pitched Blade Downflow Turbines", Can. J. Chem. Eng. 71, 226–237 (1993).
- Smith, J.M., "Simple Performance Correlations for Agitated Vessels", in "Proceedings of 7th European Congress on Mixing", Brugge Belgium (1991), pp. 233–241.
- Smith, J.M., K. Van't Riet and J.C. Middleton, "Scale-up of Agitated Gas–Liquid Reactors for Mass Transfer", in "Proceedings of 2nd European Conference on Mixing", Cambridge, UK (1977), F4, pp. 51–66.
- Smith, J.M. and M.M.C.G. Warmoeskerken, "The Dispersion of Gases in Liquids with Turbines", in "Proceedings of 5th European Conference on Mixing", Wurzburg, Germany (1985), pp. 115–126 (1985).

- Van't Riet, K., "Review of Measuring Methods and Results in Nonviscous Gas-Liquid Mass Transfer in Stirred Vessels", Ind. Eng. Chem. Process Des. Dev. **18**, 357–364 (1979).
- Van't Riet, K. and J. Tramper, "Basic Bioreactor Design", Marcel Dekker Inc., New York, NY (1991).
- Walter, J.F. and H.W. Blanch, "Bubble Break-up in Gas–Liquid Bioreactors: Break-up in Turbulent Flows", Chem. Eng. J. **32**, B7–B17 (1986).
- Whitton, M.J. and A.W. Nienow, "Scale up Correlations for Gas Hold up and Mass Transfer Coefficients in Stirred Tank Reactors", in "Proceedings of 3rd International Conference on Bioreactor and Bioprocess Fluid Dynamics", Cambridge, UK (1993), pp 135–149.
- Yawalkar, A.A., V.G. Pangarkar and A.A.C.M. Beenackers, "Gas Hold-up in Stirred Tank Reactors", Can. J. Chem. Eng. **80**, 158–166 (2002).
- Zhu, Y., P.C. Bandopadhayay and J. Wu, "Measurement of Gas–Liquid Mass Transfer in an Agitated Vessel – A Comparison between Different Impellers", J. Chem. Eng. Japan **34**, 579–584 (2001).

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