DETERMINATION OF LIQUID-SIDE AND GAS-SIDE VOLUMETRIC MASS TRANSFER COEFFICIENTS IN A BUBBLE COLUMN

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Experiments were carried out on bubble stripping of aqueous solutions of five organic solutes and oxygen dissolved at initial concentrations ranging from 0.008 to 0.04 kg·m^{-3}. Little interaction was found to exist among the solutes, as far as gas holdup and overall volumetric mass transfer coefficients at low concentrations are concerned. The liquid-side and gas-side volumetric mass transfer coefficients were obtained from measured overall coefficients. Correlations for the liquid-side volumetric mass transfer coefficients were obtained in terms of liquid-phase diffusivity and gas flow rate for two types of gas spargers: single-nozzle and porous-tube spargers. Correlations for the gas-side volumetric mass transfer coefficients were also obtained for these spargers.

Introduction

Much attention has been paid, especially from the environmental standpoint, to the volatilization of organic solutes from water bodies to the atmosphere. Surface aeration has been studied by Smith *et al.*,[26] Mackay and Yeun,[14] Roberts and Dändliker,[22] Okouchi *et al.*,[17,18] and Rathbun and Tai,[19,20] and bubble aeration by Mehta and Sharma,[16] Akita and Yoshida,[1] Hikita *et al.*,[7] Matter-Müller *et al.*,[15] Shah *et al.*,[25] Koide *et al.*,[10] Roberts *et al.*,[23] Sada *et al.*,[24] and Cho and Wakao.[2] While much work on aeration has accumulated, the authors know of no investigation that has been made on separation of the individual liquid-side and gas-side volumetric mass transfer coefficients from measured overall coefficients. The objective of the present work is to determine these individual coefficients in a bubble stripping column.

1. Theoretical

The theoretical treatment is based on the following assumptions: the liquid is completely mixed in a batch bubble stripping column; the volatile organic solute transfers from the liquid phase to the gas bubbles which rise in a plug flow mode in the column. The mass balance then gives

$$u_G\frac{dC_G}{dx}-K_La\left(C_L-\frac{C_G}{H}\right)=0 \tag{1}$$

$$C_G=0 \quad \text{at} \quad x=0 \tag{1a}$$

$$V_L\frac{dC_L}{dt}=-K_LaV\left(C_L-\frac{(C_G)_{av}}{H}\right) \tag{2}$$

$$C_L=C_{L0} \quad \text{at} \quad t=0 \tag{2a}$$

where $(C_G)_{av}$ is the average gas-phase concentration of the solute in the column.

$$(C_G)_{av}=\frac{1}{X}\int_0^X C_G dx \tag{3}$$

The Henry's law constant is defined by

$$H=\left(\frac{C_G}{C_L}\right)_{\text{equilibrium}} \tag{4}$$

The solution to the above equations then describes the system as

$$-\ln\frac{C_L}{C_{L0}}=\frac{F_GHt}{V(1-\varepsilon_G)}\left[1-\exp\left(-\frac{K_LaV}{HF_G}\right)\right] \tag{5}$$

In the case of $K_LaV/(HF_G)>5$, Eq. (5) reduces to

$$-\ln\frac{C_L}{C_{L0}}=\frac{F_GHt}{V(1-\varepsilon_G)} \tag{6}$$

This equation is the same as that from which Mackay *et al.*[13] experimentally measured Henry's law constants for various organic solutes.

When $K_LaV/(HF_G)<0.1$, Eq. (5) becomes

$$-\ln\frac{C_L}{C_{L0}}=\frac{K_Lat}{1-\varepsilon_G} \tag{7}$$

This equation can be applied when the exit gas concentration (at $x=X$) is considerably lower than the equilibrium concentration, i.e., $(C_G)_X \ll HC_L$.

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Also, it should be noted that, according to the two-film theory, the overall mass transfer coefficient, K_L, is related to the liquid-side (k_L) and gas-side mass transfer coefficients (k_G). It is expressed in terms of the volumetric coefficients as

$$\frac{1}{K_L a}=\frac{1}{k_L a}+\frac{1}{Hk_G a} \tag{8}$$

2. Experimental

Figure 1 shows a schematic diagram of the experimental apparatus. A polyacrylonitrile column (inside diameter = 11 cm, height = 40 cm) was used for batch stripping by nitrogen gas. The two types of gas spargers employed were a single nozzle of 5-mm diameter and three straight porous tubes of square cross section, 7.3-cm long with 2 cm × 2 cm cross-section, placed in parallel with centers 2.2 cm apart. Note that bubbles are formed uniformly on the total surfaces of the three porous tubes.

The spargers were each positioned in turn on the bottom of the column. Commercial cylinder nitrogen (purity 99.95%) was introduced to the gas spargers. The solutions were oxygen-saturated aqueous solutions of five organic solutes (benzene, carbon tetrachloride, chloroform, 1,2-dichloroethane and 1,1,2-trichloroethane), used both individually and in mixtures. In each case, the initial concentrations of the organic solutes and oxygen ranged from 0.008 to 0.04 kg·m^{-3}. The column held 0.003 m^3 of aqueous solution, and its temperature was maintained at 298 K during the runs.

A small amount of the liquid was sampled from the stripping column at measured time intervals; the concentrations of dissolved oxygen were determined by Winkler titrations,[27] and the concentrations of the organic solutes were measured by an FID-gas chromatograph (Shimadzu Co., Tokyo) equipped with a glass column of Chemipack C18 (Gaschro Co., Tokyo) for the analyses of benzene and chloroform, or a glass column of Gaschropack 54 (ibid) for all other chlorinated hydrocarbon solutes. **Table 1** shows the Henry's law constants at 298 K and the liquid-phase and gas-phase molecular diffusivities of the solutes at this temperature.

Preliminary experiments performed with and without a flow of nitrogen gas over the top of the column gave the same dissolved oxygen concentration-time curves. Therefore, it is considered that the top of the column is covered with the nitrogen gas which has risen in bubbles through the column so that no absorption of oxygen of the air takes place at the water surface.

The gas holdup in the stripping column was also measured from the difference in liquid height before and during the bubbling.

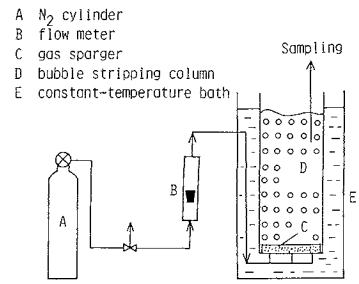


Fig. 1. Schematic diagram of experimental apparatus

Table 1. Henry's law constants and diffusivities of solutes at 298 K

Solute	Henry's law constant H [(kg·m^{-3}-gas)/(kg·m^{-3}-liquid)]	Diffusivities	
		Water-phase[e] D_L [m^2·s^{-1}]	Nitrogen gas-phase[f] D_G [m^2·s^{-1}]
CCl_4	1.244[a]	0.99×10^{-9}	0.86×10^{-5}
C_6H_6	0.224[b]	1.01×10^{-9}	0.93×10^{-5}
$CHCl_3$	0.150[a]	1.12×10^{-9}	0.95×10^{-5}
CH_2ClCH_2Cl	0.0502[c]	1.11×10^{-9}	0.96×10^{-5}
$CHCl_2CH_2Cl$	0.0374[c]	1.01×10^{-9}	0.85×10^{-5}
O_2	32[d]	2.42×10^{-9}	2.08×10^{-5}

[a] Gossett[6]
[b] Mackay *et al.*[13]
[c] Dilling[4]
[d] Matter-Müller *et al.*[15]
[e] Calculated by the Wilke–Chang correlation, with molecular volumes estimated as described in Table 3-11 of literature reference (21).
[f] Calculated by the Fuller *et al.* correlation, with atomic diffusion volumes estimated as described in Table 11-1 of literature reference (21).

The analyses were made on the assumptions that the liquid in the column was completely mixed and that gas bubbles rose in a plug flow mode. The former assumption was verified when red ink was dropped onto the top of the column. The plug gas flow was considered to be adequate when the porous-tube sparger was employed. In this case gas bubbles of almost uniform size of 3–4 mm were observed to rise straight upward in the column. In the case of single-nozzle sparger some circulation flow of small gas bubbles was observed at flow rate higher than about 0.03 m·s^{-1}. However, no information on bubble mixing in terms of the mixing coefficient has been available for the single-nozzle sparger. We then assume plug gas flow throughout this work.

3. Results

3.1 Gas holdup

The gas holdup measured with the aqueous solution of the mixed solutes is shown in **Fig. 2** in comparison with the correlations in the literature, in which $u_G = F_G$/area of the column cross-section. At

Table 2. Overall volumetric mass transfer coefficients for each component of a solute mixture in an aqueous solution subjected to nitrogen bubble stripping using a three-porous-tube sparger. Numbers in parentheses are the corresponding values for each component stripped under single-solute conditions.

u_G [m·s^{-1}]	K_La [10^{-3}s^{-1}]					
	$CHCl_2CH_2Cl$	CH_2ClCH_2Cl	$CHCl_3$	C_6H_6	CCl_4	O_2
0.0071	4.25 (4.31)	5.06 (5.11)	8.77 (8.91)	9.67 (9.49)	12.5 (12.8)	21.4 (21.0)
0.017	8.42 (8.32)	10.2 (9.99)	17.9 (17.5)	20.7 (21.0)	27.0 (26.9)	45.1 (46.0)
0.025	12.1 (11.9)	17.0 (16.8)	25.4 (24.9)	26.5 (26.2)	33.3 (33.5)	55.4 (55.1)

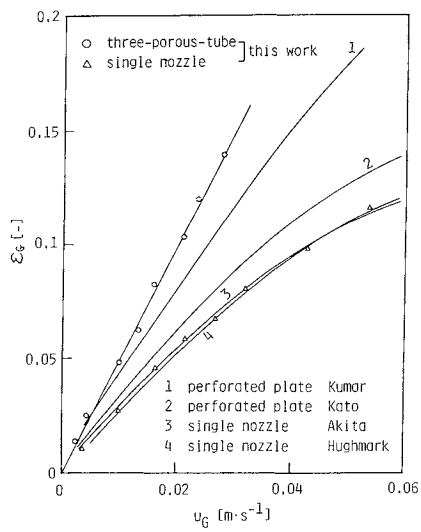


Fig. 2. Gas holdup $-u_G$ relationships in comparison with correlations in the literature

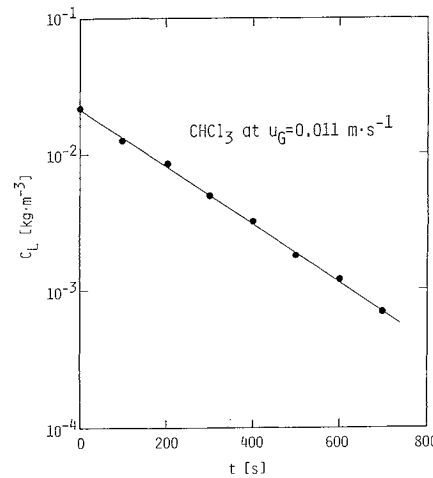


Fig. 3. C_L-t relationship for chloroform obtained from stripping with three-porous-tube sparger

a given u_G the gas holdup is low with the single-nozzle sparger and the highest with the three-porous-tube sparger. As shown, the data for the single-nozzle sparger are in good agreement with the correlations for single nozzle proposed by Hughmark,[8] and Akita and Yoshida.[1] The gas holdup data for perforated-plate sparger measured by Kato and Nishiwaki,[9] and Kumar *et al.*[11] are found to lie between those for the single-nozzle sparger and the three-porous-tube sparger. The gas holdup measured in a column of plain water was almost the same as that of the mixed solutes. It shows that the organic solutes at the low concentration levels employed had no appreciable effect on the gas holdup in the column.

3.2 Overall volumetric mass transfer coefficients K_La

(a) Single solutes

Figure 3 illustrates the decrease in the liquid-phase concentration, C_L, of chloroform with time in bubble stripping at $u_G=0.011\,m\cdot s^{-1}$ with the three-porous-tube sparger. As expected from Eq. (5), the data are represented by a straight line. Therefore, using the Henry's law constant given in Table 1, the overall volumetric mass transfer coefficient is determined from the slope of the straight line in Fig. 3. **Table 2** shows the overall volumetric mass transfer coefficients thus obtained for each solute when tested in the bubble stripping column with the three-porous-tube sparger.

(b) Mixed solutes

Experiments were also carried out with an aqueous solution of all the solutes combined. The concentrations of the solutes were again measured with time. Following the same procedure given in the preceding section, the overall volumetric mass transfer coefficients were obtained. As compared in Table 2, the overall coefficients for each solute obtained under the mixed solutes conditions are almost the same as those for the single solutes conditions. This indicates that little interaction exists among the solutes at the present low concentration levels. This agrees with the work of Mackay and Yeun,[14] who measured the mass transfer coefficients for some organic solutes on the wavy surface of a surface aeration tank. Cho and Wakao[2] have also found that there is no interaction between benzene and toluene in the bubble aeration process.

3.3 Determination of liquid-side and gas-side volumetric mass transfer coefficients k_La and k_Ga

The liquid-phase and gas-phase diffusivities of the five organic solutes fall respectively in the narrow ranges of $D_L=(0.99\text{–}1.12)\times10^{-9}\,m^2\cdot s^{-1}$ and $D_G=(0.85\text{–}0.96)\times10^{-5}\,m^2\cdot s^{-1}$, i.e., $D_L=1.055\times10^{-9}$

$m^2 \cdot s^{-1}$ within an error of about $\pm 6\%$ and $D_G = 0.905 \times 10^{-5}\ m^2 \cdot s^{-1}$, again within an error of about $\pm 6\%$. Considering the experimental errors themselves, the 6% deviations in D_L and D_G are not very significant, so that the five organic solutes may be regarded as a single solute with $D_L = 1.055 \times 10^{-9}\ m^2 \cdot s^{-1}$ and $D_G = 0.905 \times 10^{-5}\ m^2 \cdot s^{-1}$. However, as listed in Table 1, the Henry's law constants of these five organic solutes vary in a wide range from 0.0374–1.244 or within a 33-fold difference.

The overall volumetric mass transfer coefficients for the five organic solutes at constant gas flow rates obtained with the solutions of the mixed solutes are then examined in a plot of $1/K_La$ versus $1/H$. As illustrated in **Fig. 4**, for stripping with the three-porous-tube sparger, for instance, a straight line is drawn at each gas flow rate. Equation (8) shows that the intercept of the straight line gives $1/k_La$ and the slope gives $1/k_Ga$. These k_La and k_Ga values are then plotted versus the gas flow rates in **Figs. 5** and **6** respectively. As depicted in each graph, the experimental data for each sparger are represented by a straight line.

3.4 Correlations of k_La

The experimental k_La data for the three-porous-tube sparger are plotted versus D_L in **Fig. 7**. Note that in the plot the organic solutes are again regarded as one single solute with $D_L = 1.055 \times 10^{-9}\ m^2 \cdot s^{-1}$. The k_La data with $D_L = 2.42 \times 10^{-9}\ m^2 \cdot s^{-1}$ are those obtained from stripping of the dissolved oxygen. Since the D_L's are rather in a limited range, no precise determination of the power of D_L in the plot of k_La vs. D_L can be made. However, the data are well represented by straight lines with a slope of 0.5, so that

$$k_La \propto D_L^{0.5} \tag{9}$$

Note that Akita and Yoshida[1] also found that k_La was proportional to $D_L^{0.5}$. The data for the single nozzle are also found to follow Eq. (9). Consequently we obtain for the single-nozzle sparger (nozzle size: 5 mm):

$$k_La = 6.5 \times 10^3 D_L^{0.5} u_G^{0.81} \tag{10}$$

and for the three-porous-tube sparger (bubble size: 3–4 mm):

$$k_La = 2.3 \times 10^4 D_L^{0.5} u_G^{0.81} \tag{11}$$

in which k_La is in s^{-1}, D_L in $m^2 \cdot s^{-1}$ and u_G in $m \cdot s^{-1}$.

3.5 Correlations of k_Ga

The gas-side volumetric mass transfer coefficients averaged for the five organic solutes, which may be considered those for the single solute with $D_G = 0.905 \times 10^{-5}\ m^2 \cdot s^{-1}$, have been obtained. The coefficients have not been available for oxygen, since in the case of oxygen stripping no gas-side resistance exists. Mehta and Sharma[16] carried out bubble stripping experiments in various solute-gas systems and found that $k_Ga \propto D_G^{0.5}$. If this relationship is assumed, we obtain for the single-nozzle sparger (nozzle size: 5 mm):

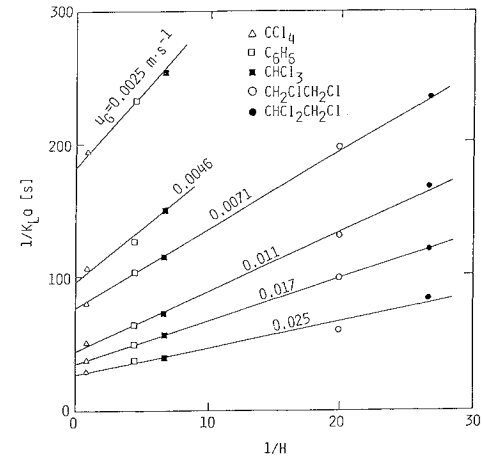


Fig. 4. $1/K_La - 1/H$ relationships for stripping with three-porous-tube sparger

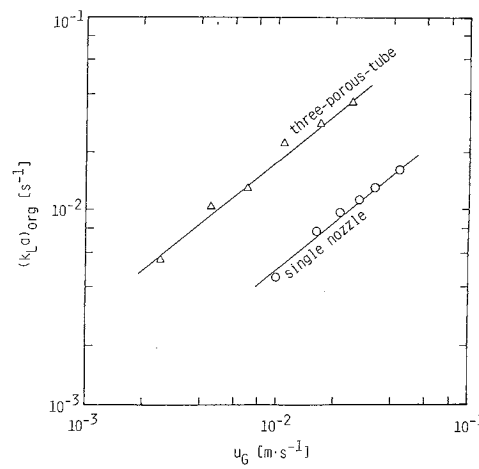


Fig. 5. Relationships between k_La averaged for the five organic solutes and u_G

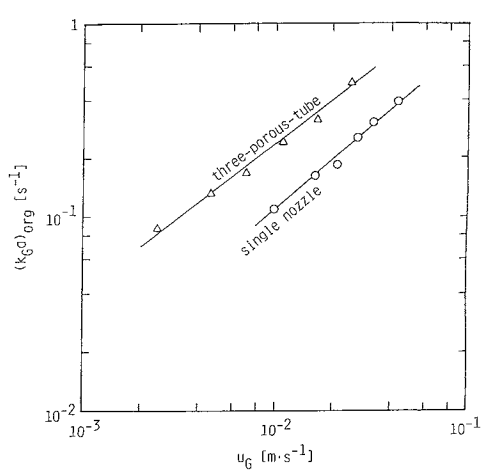


Fig. 6. Relationships between k_Ga averaged for the five organic solutes and u_G

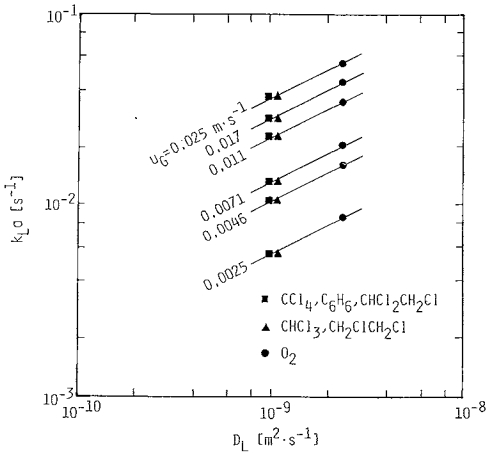


Fig. 7. k_La-D_L relationships at various nitrogen gas flow rates with three-porous-tube sparger

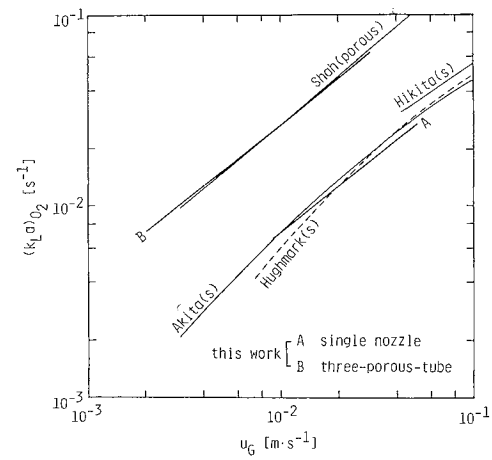


Fig. 8. Comparison of various correlations for $(k_La)_{O_2}$. s: single nozzle; porous: porous plate

$$k_Ga=2.0\times10^3 D_G^{0.5}u_G^{0.88} \tag{12}$$

and for the three-porous-tube sparger (bubble size: 3–4 mm):

$$k_Ga=2.6\times10^3 D_G^{0.5}u_G^{0.76} \tag{13}$$

where k_Ga is in s^{-1}, D_G in m^2·s^{-1} and u_G in m·s^{-1}.

4. Discussion

Prior to obtaining Eqs. (10)–(13) it was assumed that k_La was a function of D_L and u_G, and that k_Ga was a function of D_G and u_G. The concentrations of the solutes employed in the present work are so low (less than 0.04 kg·m^{-3} or 40 ppm) that, as mentioned already, the ε_G–u_G relationship was found to depend on the type of gas sparger but not on whether the liquid is aqueous organic solutions or plain water. The surface tensions and liquid viscosities at these low concentration levels are also considered not to be different, respectively, from those for the plain water-gas system. The specific bubble surface area, a, is considered to depend upon the type of gas sparger (or bubble size) and ε_G (or u_G). Therefore, as far as the present low-level aqueous solutions are concerned, it would be appropriate to correlate k_La in terms of D_L and u_G, and similarly k_Ga in D_G and u_G for each gas sparger.

Smith *et al.*[26] showed that the ratio of the liquid-side mass transfer coefficients for surface stripping of various organic solutes to that of dissolved oxygen at the same flow rate of nitrogen gas flowing over the top of a tank is expressed as

$$\frac{(k_L)_A}{(k_L)_B}=\left[\frac{(D_L)_A}{(D_L)_B}\right]^{\alpha} \tag{14}$$

where $\alpha=0.61$. Roberts and Dändliker[22] also found α to be 0.62 from experiments on stripping from the surface of a tank of mixed solutions of various organic solutes and dissolved oxygen into a stream of nitrogen gas over the tank. However, Matter-Müller *et al.*[15] obtained $\alpha=1$ from experiments on stripping of some organic solutes and absorption of oxygen in a bubble aeration tank. It should be noted that in these three papers k_L or k_La data were obtained under the assumption of $K_L=k_L$ or $K_La=k_La$. Our present measurements show, as expressed by Eq. (9), that $\alpha=0.5$.

Using the diffusivity for dissolved oxygen, $(D_L)_{O_2}=2.42\times10^{-9}$ m^2·s^{-1}, correlations of $(k_La)_{O_2}$ are obtained from Eqs. (10) and (11). As shown in **Fig. 8**, the solid line A for the single-nozzle sparger agrees reasonably well with single-nozzle correlations of Hughmark,[8] Akita and Yoshida,[1] and Hikita *et al.*[7] The line B for the three-porous-tube sparger is also in good agreement with the porous-plate correlation proposed by Shah *et al.*[25]

In our previous work[2] on stripping of benzene and toluene in a batch bubble aeration, the $\log C_L$–t relationship deviated from linearity when time became long. This was also true in the present work. Although it is not shown in Fig. 3, the decrease in $\log C_L$ with time became small when C_L decreased further than about 5×10^{-4} kg·m^{-3}. As discussed in the earlier work, the decrease in slope of $\log C_L$–t curve at longer time or at lower C_L is considered to result partly from desorption of the organic solutes once adsorbed on the tank walls and partly from contamination at the air–water interface.

In fact, Davies *et al.*,[3] Lochiel[12] and Fujinawa *et al.*[5] have pointed out that the mass transfer coefficient decays considerably when the air–water interface is even slightly contaminated. In the previous work an air compressor was used to pump room air to the bottom of the aeration tank so that the air–water interface would have been easily contaminated with oily materials and impurities coming from the compressor as well as those accompanying the air. In the present work nitrogen gas was introduced directly

from a cylinder to the spargers. Therefore, interface contamination must have progressed considerably more slowly in the present work compared to the earlier experiments. However, it was not observed that the linear $\log C_L$–t relationship in the present work held for a longer period of time than that in the previous work. Desorption of the organic solutes from the tank walls to the aqueous solutions is, therefore, considered to be more responsible than interface contamination for the deviation from linearity between $\log C_L$ and t at low solute-concentration levels.

Conclusions

Experiments on stripping of five organic solutes and dissolved oxygen (at initial concentrations 0.008 to $0.04\,kg\cdot m^{-3}$) were conducted in a batch bubble stripping column using cylinder nitrogen.

Little interaction was found to exist among the solutes as far as gas holdup and overall volumetric mass transfer coefficient at low concentration levels are concerned. The liquid-side volumetric mass transfer coefficient was expressed to be proportional to the liquid-phase diffusivity of the solute to the power of 0.5. Correlations of the liquid-side and gas-side volumetric mass transfer coefficients for single-nozzle and porous-tube spargers were obtained.

Nomenclature

C_G	=	gas concentration	$[kg\cdot m^{-3}]$
C_L	=	liquid concentration	$[kg\cdot m^{-3}]$
C_{L0}	=	initial liquid concentration	$[kg\cdot m^{-3}]$
D_G	=	gas diffusivity	$[m^2\cdot s^{-1}]$
D_L	=	liquid diffusivity	$[m^2\cdot s^{-1}]$
F_G	=	gas volumetric flow rate	$[m^3\cdot s^{-1}]$
H	=	Henry's law constant	$[(kg\cdot m^{-3}\text{-gas})/(kg\cdot m^{-3}\text{-liquid})]$
K_La	=	overall volumetric mass transfer coefficient	$[s^{-1}]$
k_Ga	=	gas-side volumetric mass transfer coefficient	$[s^{-1}]$
k_La	=	liquid-side volumetric mass transfer coefficient	$[s^{-1}]$
t	=	time	[s]
u_G	=	superficial gas velocity $= F_G$/column cross-sectional area	$[m\cdot s^{-1}]$
V	=	volume of a mixture of liquid and gas bubbles	$[m^3]$
V_L	=	volume of liquid	$[m^3]$
X	=	height of liquid level	[m]
x	=	axial distance variable	[m]
ε_G	=	gas holdup	[—]

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