The air-phase tranfer resistance in the lumen of a hollow fiber at low air flow[☆]

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Abstract

Removal of volatile organic compounds (VOCs) from water/wastewater by membrane air stripping (MAS) was studied using microporous polypropylene hollow fiber membrane modules. Chloroform was used as model organic compound. The mass transfer of organic compound from feed solution to air stream was studied with liquid cross-flow on the shell side and airflow in the lumen of hollow fibers. The Graetz numbers for airflows in the lumen of the fibers in this study ranged from 3.1×10^{-3} to 6.9×10^{-3}. For such low Graetz numbers, the widely used Lévéque's [Annales de Mines 12 (13/14) (1928) 201] model was found to be inapplicable. An alternative model is presented to predict the mass transfer resistance. This model was found to be useful, particularly when the airflow rate on the lumen side of the hollow fiber is very low.
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Keywords: Mass transfer resistance; Airflow; Hollow fiber; Membrane; Air stripping

1. Introduction

Separation of volatile organic compounds (VOCs) from liquid streams by MAS is being considered as an alternative that may help overcome some of the shortfalls of the conventional treatment methods. It is reported that MAS offers an order of magnitude higher overall volume specific mass transfer coefficient (K_La) than that of packed-tower air stripping (PTA) and needs much lower air-to-water ratio to achieve the same degree of removal due to its multi-pass nature [2]. The benefit of lower air-to-water ratio facilitates the use of a closed-loop system to avoid transferring VOCs from aqueous to air phase [3], either by destroying or trapping the VOCs. However, MAS provides additional mass transfer resistance due to the presence of a membrane. The resistance was reported as negligible when the membranes were dry and the pores were air-filled [4,5] and almost equal to the liquid-phase resistance when the membrane became partially wetted as a result of its prolonged contact with water [4]. It was also reported in the literature that the Lévéque

[1] correlation overestimated mass transfer coefficient at low liquid flow rates in a cylindrical tube, when the Graetz numbers ($d_i^2 v^w / LD$) were below 4 and experimental values deviated from that predicted by Lévéque's [1] correlation [6,7]. Such deviations have been observed for hollow fibers by a number of researchers [7–11]. Analogous deviations have also been reported in heat transfer [12,13]. Wickramasinghe et al. [7] stated that the theory and experiments do not agree at low flow rates, apparently because of slight polydispersity in hollow fiber diameter. However, the deviations at low flow rates were also reported for heat transfer, where the tube diameter of the heat exchanger might be free of this polydispersity. As the low air flow rate is one of the benefits MAS offers, proper prediction of its resistance is important for design purposes. The objective of this paper is to present a model developed to predict the local air-film mass transfer resistance for low air flows in the lumen of hollow fibers.

2. Theory

Mass transfer fundamentals for the transport of VOCs in MAS systems have been reviewed in detail by Mahmud et al. [4,14]. VOCs are transferred from water to air through intimate contact of the two phases at the mouth of

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Nomenclature

a	surface to volume ratio (m^2/m^3)
C_t	VOC concentration in the reservoir at time t (g/m^3)
C_0	VOC concentration in the reservoir at time 0 (g/m^3)
d_i	inner diameter of the hollow fiber/(cylindrical tube) (m)
d_o	outer diameter of the hollow fiber (m)
D	diffusion coefficient of compound (m^2/s)
D_c	continuum (ordinary) diffusion coefficient of compound in air phase (m^2/s)
D_{eff}	effective diffusion coefficient of compound in air (m^2/s)
D_w	diffusion coefficient of compound in water (m^2/s)
D_{Kn}	Knudsen diffusion coefficient of compound in air (m^2/s)
Gr	Graetz number
h	length of the hollow fiber module compartment ($0.5L$) (m)
H	dimensionless Henry's Law constant
k	rate constant (min^{-1})
k_a	local air-phase mass transfer coefficient (m/s)
k_L	local liquid-phase mass transfer coefficient (m/s)
k_m	membrane mass transfer coefficient (m/s)
K_L	overall liquid-phase-based mass transfer coefficient (m/s)
$K_L a$	overall volume specific mass transfer coefficient (h^{-1})
L	hollow fiber length (m)
Pe	Peclet number ($d_i v^w / D_w$)
Q_a	air flow rate (m^3/s)
Q_w	water flow rate (m^3/s)
r_{in}	inner radius of the membrane module (m)
r_{out}	outer radius of the center tube (m)
R	stripping factor ($Q_w / Q_a H$)
Re	Reynolds number ($d_o u^w / \nu$)
Sc	Schmidt number (ν / D_w)
Sh	Sherwood number ($k_L d_o / D_w$) $= 1.62 Gr^{0.33}$
$\overline{Sh}$	average Sherwood number
t	time (s, min)
u^w	aqueous solution velocity on the shell side of the hollow fiber (m/s)
v^a	air velocity in the lumen of the hollow fiber (m/s)
v^w	aqueous solution velocity in the lumen of the hollow fiber (m/s)
V_w	reservoir volume (m^3)
x	fraction of the pore filled with air
$1-x$	fraction of the pore filled with water

Greek letters

δ	pore length (m)
ε	fiber porosity (dimensionless)
ν	kinematic viscosity of air/water (m^2/s)
τ	pore tortuosity (dimensionless)
ω	coefficient of variation for the fiber radius measurements

the membrane's air-filled pores. The driving force for the mass transfer is the difference in concentration between the two phases. Mass transfer in membrane air stripping involves three sequential steps. Firstly, a VOC molecule diffuses from the bulk aqueous solution across the liquid boundary layer to the membrane surface. Secondly, it diffuses through the air- and or liquid-filled pores. This diffusion step does not exist in PTA. Finally, it diffuses through the air boundary layer outside the membrane into the stripping air. The overall mass transfer resistance is hence the combined effect of these three separate mass transfer resistances. As mass transfer resistances are considered to be proportional to the inverse of the corresponding mass transfer coefficients, the overall liquid-phase-based mass transfer resistance ($1/K_L$)

can be expressed as follows:

$$\frac{1}{K_L} = \frac{1}{k_L} + \frac{1}{k_m H} + \frac{1}{k_a H} \tag{1}$$

where K_L is the overall liquid-phase-based mass transfer coefficient (m/s), k_a the local air-phase mass transfer coefficient (m/s), k_L the local liquid-phase mass transfer coefficient (m/s), k_m the membrane mass transfer coefficient (m/s), and H is the dimensionless Henry's Law constant.

For liquid cross-flow on the shell side and air flow in the lumen of the hollow fibers, the individual mass transfer coefficients can be predicted using mass transfer correlations based on dimensionless numbers developed by a number of researchers [1,4,13,15–18].

The local liquid-phase mass transfer coefficient, k_L, is predicted based on the following correlation developed by Kreith and Black [15] for cross-flow in closely packed tube bank heat exchangers:

$$Sh = 0.39Re^{0.59}Sc^{0.33} \tag{2}$$

where Re is the Reynolds number ($d_o u^w / \nu$), Sh the Sherwood number ($k_L d_o / D_w$), Sc the Schmidt number (ν / D_w), d_o the outer diameter of the hollow fiber (m), u^w the aqueous solution velocity on the shell side of the fibers (m/s), ν the kinematic viscosity of water (m²/s), and D_w is the diffusion coefficient of compound in water (m²/s).

The membrane mass transfer coefficient, k_m, is predicted using the equation developed by Mahmud et al. [4] as the pores appeared to be partially air-filled and partially water-filled [4,19].

$$\frac{1}{k_m H} = x\frac{\delta\tau}{D_{eff}\varepsilon H} + (1-x)\frac{\delta\tau}{D_w\varepsilon} \tag{3}$$

where δ is the pore length (m), D_{eff} the effective diffusion coefficient of compound in air (m²/s), τ the pore tortuosity (dimensionless), ε the fiber porosity (dimensionless), x the fraction of the pore filled with air, and $1-x$ is the fraction of the pore filled with water.

D_{eff} is estimated using the relationship $D_{eff} = ((1/D_c) + (1/D_{Kn}))^{-1}$, given by Pollard and Present [20]. Here, D_c is the continuum (ordinary) diffusion coefficient of the compound in the air phase (m²/s) and D_{Kn} the Knudsen diffusion coefficient of the compound in air (m²/s).

The air-phase mass transfer coefficient, k_a is estimated by the following equation, derived from Lévéque's [1] correlation for laminar flow in a cylindrical tube, incorporating Henry's law constant, as the boundary layer is gaseous:

$$\frac{1}{k_a H} = \frac{0.617}{H}\left(\frac{Ld_i}{v^a D_c{}^2}\right)^{0.33} \tag{4}$$

where v^a is the air velocity inside the hollow fiber (m/s), d_i the inner diameter of the hollow fiber (m), and L is the length of fiber (m).

As Lévéque's [1] correlation overestimated the mass transfer coefficient at low liquid flows, Wickramasinghe et al. [7] proposed the following relationship based on

their experiments with liquid flow in the lumen to take into account the polydispersity in hollow fiber diameter:

$$\bar{k}_L = 1.5 \times 10^{-4}\frac{v^w}{L} \tag{5}$$

where $\bar{k}_L$ is the average local liquid-phase mass transfer coefficient (m/s) and v^w is the liquid velocity inside the hollow fiber (m/s).

Eq. (5) does not incorporate any physicochemical properties of the compounds involved and will thus be the same for an identical set of operating conditions irrespective of the types of compounds. But the local film resistance on the lumen side of the fiber depends on the film thickness as well as on the diffusion coefficient of the solute and should depend on the physicochemical properties of the compound. Hence, Wickramasinghe et al. [7] proposed the following correlation, which has been developed based on average values and takes the physicochemical properties of the compounds into account:

$$\overline{Sh} = Sh\left[1 - \left(\frac{18Sh}{Gr} + 7\right)\omega^2 + \cdots\right] \tag{6}$$

where Sh is the Sherwood number ($1.62Gr^{0.33}$), $\overline{Sh}$ the average Sherwood number, Gr the Graetz number ($Pe \cdot d_i / L$), Pe the Peclet number ($d_i v^w / D_w$), and ω is the coefficient of variation for the fiber radius measurements.

3. Experimental

3.1. Materials

Commercial chloroform (99.8%, BDH, Inc., Toronto, Ont., Canada) was used to prepare the feed solutions and the standards for the gas chromatographs. Its relevant physicochemical properties are given in Table 1.

3.2. Experimental set-up and method

The detailed description of the set-up and the method of testing are given elsewhere [4]. The membrane air-stripping

Table 1
Physicochemical properties of chloroform at 23 °C

D_c ($\times 10^5$ m²/s)	0.923[a]
D_{Kn} ($\times 10^4$ m²/s)	2.29[b]
D_w ($\times 10^9$ m²/s)	0.893[c]
H	0.151[d]

[a] D_c: continuum diffusion coefficient of the component in air phase, calculated using the correlation given by Fuller et al. [21].

[b] D_{Kn}: Knudsen diffusion coefficient, calculated using the correlation given by Cussler [22].

[c] Diffusion coefficient of chloroform in water, calculated using the correlation given by Wilke and Chang [23], multiplied with a factor of 0.9 to match the observed deviation by Smith et al. [24] and Roberts and Dändliker [25].

[d] Experimentally determined by Mahmud et al. [26].

experimental set-up included a reservoir (vol. $= 6.675 \times 10^{-3} \, m^3$), a hollow fiber membrane module, an aqueous solution feed circulation line and an air-stripping line. The shell side of membranes was kept in contact with the aqueous phase for 48 h prior to the start of the tests to reach a steady wet state. The membrane module was a Liqui-Cel® extra-flow 63 mm × 203 mm laboratory-scale membrane contactor (Separation Products Division, Hoechst Celanese Corporation, Charlotte, NC, USA, www.Membrana.com), made of polypropylene microporous hollow fibers.

The samples were collected from the reservoir for analysis every 10 min in the beginning of each run but the interval was increased at later stages. Stripping airflow rates were varied from 3.33×10^{-5} to $9.17 \times 10^{-5} \, m^3/s$, while the liquid flow rate was kept constant at 3.33×10^{-5} or $5.33 \times 10^{-5} \, m^3/s$. Initial chloroform concentrations of the feed solutions were between 650 and 790 g/m^3. Stock solutions were prepared in a big reservoir and then transferred to the system reservoir. The temperature of the solution as well as the air were kept at $23.0 \pm 0.2\,°C$. The pressure drops for the air side and solution side were 1.2–3.0 and 10.0–12.5 kPa, respectively. The total duration of a typical test was 160 min.

According to a review by Mahmud et al. [14], the change of organic concentration of the solution in a completely mixed reservoir of a batch MAS system with time, can be described by the standard experimental first-order relationship [10]:

$$\ln\left(\frac{C_0}{C_t}\right) = kt \qquad (7)$$

where k is the rate constant (min^{-1}) and t is the time (min).

Application of this equation with substitution of prevailing parameters provides the overall liquid-phase-based mass transfer coefficient, K_L, for the system when air and liquid solution streams are on the lumen and shell side, respectively [14]:

$$K_L = \frac{u^w}{aL}(1-R)^{-1}\ln\left\{\left[\frac{Q_w}{Q_w - V_w k}\right](1-R) + R\right\} \qquad (8)$$

where u^w is the aqueous solution velocity outside the hollow fiber (m/s), a the surface to volume ratio (m^2/m^3), Q_w the water flow rate (m^3/s), R the stripping factor ($Q_w/Q_a H$), Q_a the air flow rate (m^3/s), and V_w is the reservoir volume (m^3).

The values for the parameters in Eq. (8) are available numerically from the experiments. The water velocity outside the hollow fiber, u^w for the present study was estimated using the following equation [4].

$$u^w = \frac{(Q_w/2\pi h)(1/(r_{out} - r_{in}))\ln(r_{out}/r_{in})}{\text{void fraction}} \qquad (9)$$

where h is the length of the hollow fiber module compartment ($0.5L$) (m), r_{out} the outer radius of the center tube (m), and r_{in} is the inner radius of the membrane module (m).

3.3. Analytical equipments

Chloroform samples were analyzed by a total organic carbon (TOC) analyzer and counter checked by a gas chromatograph (Varian—Vista Series 6000, Varian Instrument Group, Walnut Creek Division, Walnut Creek, CA) to which a liquid purge and trap sample concentrator (Tekmar-LSC-2, Tekmar Company, Cincinnati, OH) was attached. The GC system had a flame ionization detector (FID), operated with a packed column (Carbopack B 60/80 Mesh, 1% SP-1000, 2.43 m × 3 mm SS, Supelco Canada Ltd., Oakville, Ont.) and an integrator (Waters 820 Chromatography Data Station, Water Chromatography Division, Millipore Corporation, Milford, MA).

4. Results and discussion

4.1. Effect of low air velocity on mass transport

In the MAS experiments, the liquid velocity was constant while the air velocity was changed; the liquid film resistance should hence be constant. Since it is reasonable to assume that the mass transfer resistance due to the membrane should also not change with the change of air velocity, the air-phase mass transfer resistance is the only variable with air velocity. The rate constant k, calculated according to Eq. (7), increased with the increase of air velocity as shown in Fig. 1. As a result, the observed overall mass transfer coefficients, K_L, calculated from the rate constants using Eq. (8) for MAS of chloroform, increased with an increase in air velocity as shown in Fig. 2, as expected by the reduction of the gas film resistance. However, the sensitivity to the variation in air velocity was much stronger than predicted by Eq. (4) based on Lévéque's [1] correlation (see Fig. 2, dashed line) and shows that the mass transport in this process is not strictly liquid-phase controlled. Within the range of experimental air velocities studied, Eq. (4) overestimated the local air-phase mass transfer coefficient. The Graetz numbers for this study ranged from 3.1×10^{-3} to 6.9×10^{-3}, much lower than 4, the lower limit for the applicability of the Lévéque's [1] model [6,7], which explains the deviations of the experimental values from those of Lévéque [1] predictions. Wickramasinghe et al. [7] proposed Eq. (5) as an alternative for the average local mass transfer coefficient on the lumen side.

Although Eq. (5) is given for the local liquid-phase mass transfer coefficient, $\bar{k}_L$, for liquid flow on the lumen side, it is assumed that the equation can be used for the local air-phase mass transfer coefficient, $\bar{k}_a$, when air flows on the lumen side. Thus, $\bar{k}_a$ and $1/\bar{k}_a H$ were calculated based on Eq. (5) for different air velocities. $1/k_L$ was calculated using the Kreith and Black [15] correlation (Eq. (2)) and $1/k_m H$ was obtained by Eq. (3) with $x = 0.75$ corresponding to a wet condition of the membrane. The overall mass transfer resistance $1/K_L$ was then obtained by adding individual values of $1/k_L$, $1/\bar{k}_a H$ and $1/k_m H$. The predicted
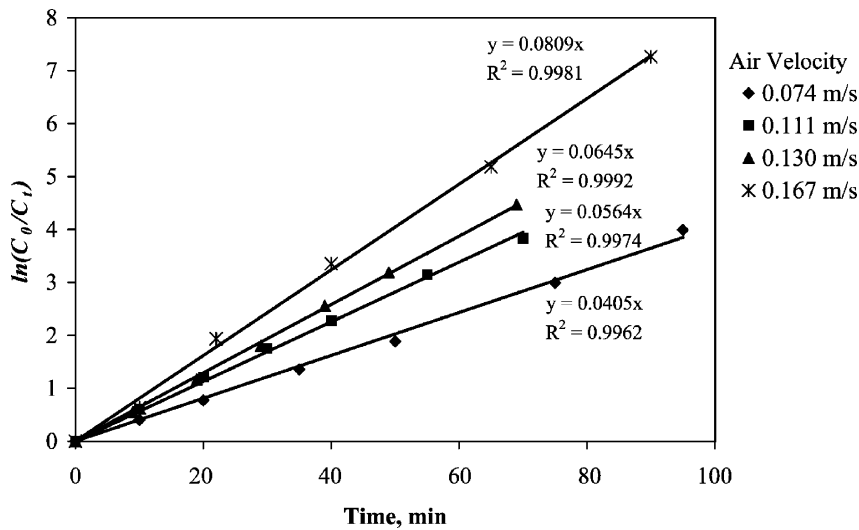
Fig. 1. Comparison of the rate constants observed at different air velocities for MAS of chloroform (solution velocity $= 5.95 \times 10^{-3}$ m/s).

overall mass transfer coefficients and the experimental data from this study are compared in Fig. 3. Although the agreement is fair at low air velocities, the model underestimates the overall mass transfer coefficient, K_L, at higher velocities. This is probably caused by the underestimation of $\bar{k}_a$ by Eq. (5). In an attempt to describe the data in a better way, Eq. (6) was also evaluated.

Eq. (5) ignores physicochemical parameters, which does not seem reasonable. Although, Wickramasinghe et al. [7] showed that the $\overline{Sh}$ calculated by Eq. (6) fit their data well, it did not fit our experimental data. The reason was apparently that their calculation was for liquid flow in the lumen, whereas in our study, air flow was in the lumen. The gas-phase diffusion coefficient of a compound is almost four orders of magnitude higher than that for the liquid-phase. Graetz numbers for this study were very small, the $\overline{Sh}$ be-

came negative and the correlation was thus found to be inapplicable for predicting the air-film resistance at low flows on the lumen of hollow fibers.

Another attempt was made to modify Lévéque's [1] correlation (Eq. (4)) for a better prediction of the resistance at these low air flows. Rearranging Eq. (4) to

$$\frac{1}{k_a H} = \left[\frac{0.617}{H} \left(\frac{L d_i}{D_c^2} \right)^{0.33} \right] \left(\frac{1}{v^a} \right)^{0.33} \tag{10}$$

it became clear that quantities in the square bracket do not change when the air velocity v^a is changed. In the following equation, the exponent applicable to $1/v^a$ was changed from 0.33 to q

$$\frac{1}{k_a H} = \left[\frac{0.617}{H} \left(\frac{L d_i}{D_c^2} \right)^{0.33} \right] \left(\frac{1}{v^a} \right)^{q} \tag{11}$$
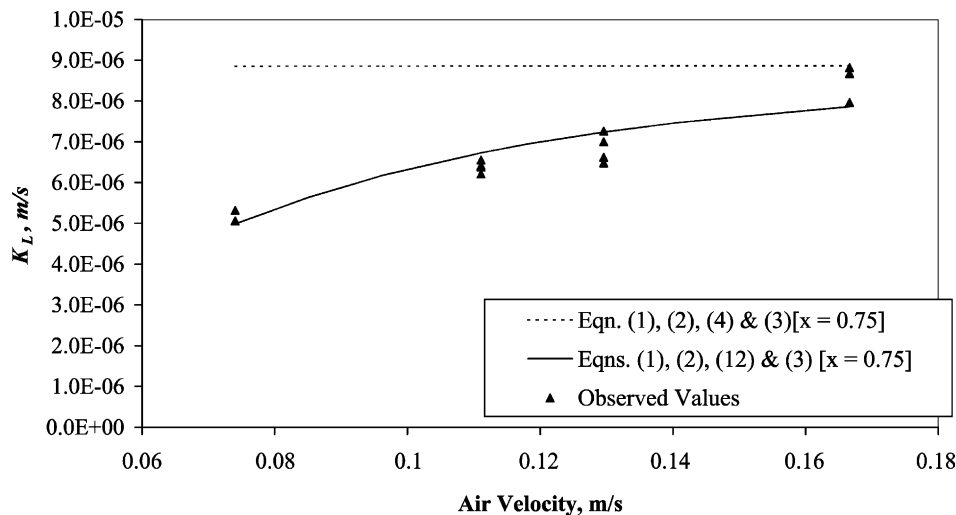

Fig. 2. Comparison of the Lévéque equation and the modified Lévéque equation with experimental data (solution velocity $= 5.95 \times 10^{-3}$ m/s).
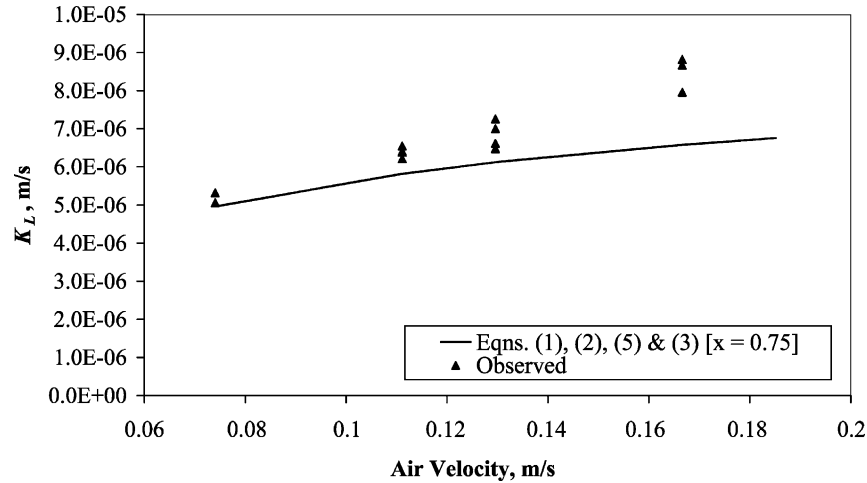
Fig. 3. Modeling of experimental data using the Wickramasinghe et al. [7] correlation (solution velocity $= 5.95 \times 10^{-3}$ m/s).

The best value of q was found using the data in Fig. 2 as per the following approach: $1/k_a H$ values were calculated for each air velocity by subtracting the theoretical values of $1/k_L$ and $1/k_m H$ from the experimental $1/K_L$ values. The $1/k_a H$ values so obtained were then used in Eq. (11) to calculate the best-fit q values by nonlinear regression analysis. The coefficients were determined by a non-linear regression routine in STATGRAPHICS PLUS (Manugistics, Inc., Rockville, MD). The model provided the best value of q with a 95% confidence interval as 2.19 ± 0.016 with a standard error of 0.026. Eq. (11) is hence transformed into:

$$\frac{1}{k_a H} = \left[\frac{0.617}{H} \left(\frac{Ld_i}{D_c^2} \right)^{0.33} \right] \left(\frac{1}{v^a} \right)^{2.19} \qquad (12)$$

which is applicable for this system.

The overall mass transfer coefficients, K_L were calculated using Eqs. (1)–(4) with $x = 0.75$ and the results are shown in Fig. 2 as dashed line. They were also calculated by

Eqs. (1)–(3) and (12) with $x = 0.75$. Note that the Lévéque's [1] correlation, Eq. (4) was modified to Eq. (12) in the latter approach. Both approaches were compared with experimental K_L values in Fig. 2. The figure shows that the modified Eq. (12) fits the experimental data far better. To verify the proposed model, a new set of MAS tests with chloroform at a higher aqueous solution flow was carried out. The results are shown in Fig. 4. It was found whereas the Lévéque's [1] correlation failed, the modified Eq. (12) successfully predicted the K_L values. It is important to note that at higher air flows, both model predictions are similar as the air-film resistance seems to be negligible and can be ignored. This is shown in Fig. 4 by plotting the predicted K_L values without incorporating air-film resistance, i.e. K_L calculated using Eqs. (1)–(3) with $x = 0.75$ only. From this comparison, it becomes clear that the estimation of K_L values with or without incorporating Lévéque's [1] model has very little effect on the prediction. On the other hand, the modified model provides a far better prediction, especially at low airflows,
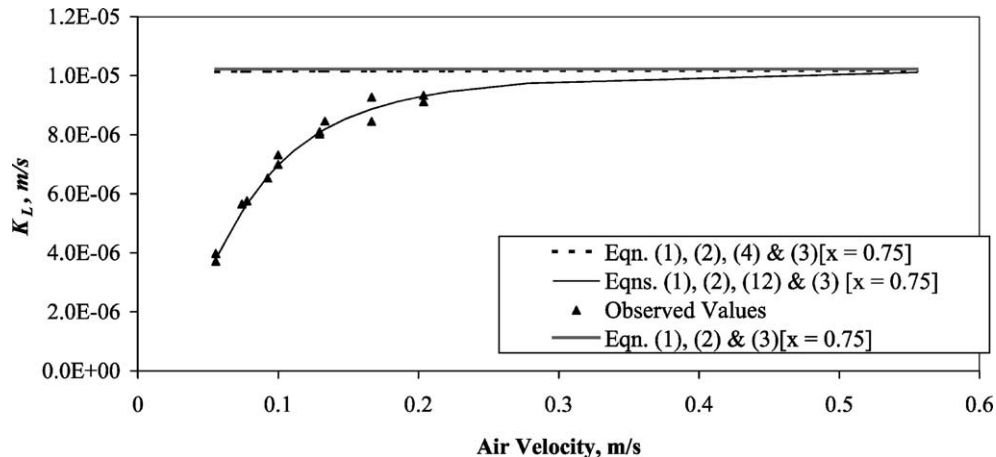

Fig. 4. Comparison of experimental data with predicted K_L values, using the Lévéque equation, the modified Lévéque equation, as well as with omission of the air-film resistance (solution velocity $= 9.52 \times 10^{-3}$ m/s).

which are the conditions where MAS has an advantage over PTA [2].

5. Conclusions

The Lévéque's [1] correlation overestimates the local air-phase mass transfer coefficient in the lumen of the fiber at low air velocities. The alternative correlations developed by Wickramasinghe et al. [7] for predicting the local liquid-phase mass transfer coefficient had some limitations when applied to the prediction of the local air-phase mass transfer coefficient. Experimental data were fitted by a modified approach.

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