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Short communication

Mass transfer coefficient for two-phase countercurrent flow in a packed column with a novel internal

Yanhui Yuan, Minghan Han*, Lunwei Wang, Dezheng Wang, Yong Jin

Department of Chemical Engineering, Tsinghua University, Beijing 100084, China

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Abstract

The gas–liquid mass transfer coefficient k_La for gas–liquid countercurrent flow in a packed column with a novel internal was measured using the absorption of oxygen. A correlation formula for predicting k_La that shows a satisfactory agreement with experimental results is presented. The mass transfer coefficient is directly proportional to the liquid hold-up, which depends on the structural parameters of the internal, the superficial liquid flow velocity and the porosity of the packing bed. A suitable internal should be made of many springs of small diameter.

Keywords: Gas–liquid-solid reactors; Countercurrent flow; Internal; Mass transfer

1. Introduction

For operations using gas–liquid countercurrent flow in a packed column, such as reactive distillation, it is important to avoid excessive pressure drop and "flooding". To achieve this, various catalyst loading methods have been developed to increase the average voidage of the packed bed, such as catalyst-containing bales, catalytic random packings, and structured catalyst supports (KATAPAK®, MULTIPAK®) [1]. Although some of these catalyst loading methods have been applied in industry, they have their drawbacks. For example, it is difficult to manufacture the catalyst-containing structures properly and to change and recover deactivated catalysts. Recently, a novel internal for a packed column was invented by Han et al. [2]. Cold model experiments [3–6] showed that a packed column with the novel internal has many advantages such as low pressure drop, simple structure, low operating cost, convenience of installation and removal of catalyst, and a large catalyst loading fraction.

The mass transfer coefficient is one of the most important parameters for gas–liquid countercurrent flow in a packed column. It affects the chemical reaction and the design of the column. Numerous attempts have been made to describe the mass transfer characteristics of packed columns operating as countercurrent gas–liquid contactors. In a reactive distillation column packed with catalytic bales, mass transfer was studied using an ammonia-air–water system for the gas phase and a carbon dioxide-sodium-bicarbonate-water system for the liquid phase [7]. The absorption of ammonia and desorption of carbon dioxide were used to determine gas and liquid phase mass transfer in MULTIPAK®, respectively [8]. The absorption of oxygen method was employed to study mass transfer in a TLP (three-levels-of-porosity) reactor [9] and in a cross-flow packed bed [10].

However, the flow behavior of a fluid in a column with the new internal is different from that in the above conventional columns. It depends not only on the properties and flow behavior of the fluid, but also on the geometric parameters of the internal [3,4]. To determine the mass transfer coefficients, the mass transfer model must be correlated with the new flow behavior of the fluid and the geometric parameters of the internal. In the present work, the mass transfer was measured by means of absorption of oxygen in an air–water system, and a model was proposed to estimate the mass transfer coefficient for design and scale-up purposes.

2. Experiments

2.1. Experiment setup

The experimental apparatus and internals are the same as that described in Han et al. [3,6]. The column is a Plexiglas tube of 140 mm i.d. packed with porcelain balls of

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* Corresponding author. Tel.: +86-10-6278-1469; fax: +86-10-6277-2051.
E-mail address: hanmh@flotu.org (M. Han).

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Nomenclature

a	effective gas–liquid mass transfer specific surface (m^2/m^3)
a_b	specific surface of packing bed (m^2/m^3)
a_t	specific surface of total column (m^2/m^3)
a_0, a_1	parameters in mass transfer model in Eq. (3)
C	oxygen concentration in water (mol/l)
C^*	oxygen concentration in air-saturated water (mol/l)
d_p	diameter of the porcelain balls (mm)
$\hat{d}_p$	the equivalent diameter of catalyst bales in Eq. (2) (m)
$\bar{d}_I$	the equivalent diameter distance in Eq. (3) (m)
D_c	inside diameter of the column (m)
D_I	outside diameter of the internal (m)
D_L	diffusion coefficient in liquid phase (m^2/s)
F_I	volume fraction of the internal, dimensionless
h_i	distance between the ith and the $i+1$th sampling location (m)
h_{L0}	liquid hold-up (m^3 liquid/m^3 column)
i	number of the sampling location, dimensionless
k_L	mass transfer coefficient (m/s)
k_La	volumetric mass transfer coefficient per volume of column (s^{-1})
l_B	height of the stage between two baffles (mm)
n	the total number of sampling-points along the column
u_G	superficial gas velocity in the column (m/s)
u_L	superficial liquid velocity in the column (m/s)

Greek letters

ε_p	void fraction in packing beds
μ	viscosity (kg/m s)
ρ	density of fluid (kg/m^3)

Subscripts

b	packing bed
G	gas phase
L	liquid phase

three average diameters, which are 2.5, 3.5 and 5 mm, and the corresponding bulk void fractions are 0.35, 0.38, and 0.4, respectively. Nitrogen, air and water at normal temperature and atmospheric pressure were used to measure the liquid mass transfer rate of the packed column with the internal. The superficial liquid velocities (u_L) were varied from 0.0018 to 0.0072 m/s and the superficial gas velocities (u_G) were varied from 0.14 to 0.57 m/s. Dissolved oxygen concentrations in water were measured using a dissolved oxygen meter (HANNA H19143).

The experimental procedure is similar as that in Han et al. [3]. The main steps of the liquid-phase mass transfer experiments are: (1) pre-wetting all packing with larger superficial velocities of gas and liquid; (2) starting-up of the experiment system to achieve a steady-state in a controlled hydrodynamic condition; (3) sampling the liquid using the sampling-points along the column, and measuring the oxygen concentrations in the samples.

2.2. Measurement of liquid phase mass transfer

To enhance the reliability of experiment, the liquid was sampled at five different points along the column that divided the column into four is equal parts. The average mass transfer coefficient k_La in column can be calculated by Eq. (1) [11].

$$k_La = \frac{1}{n-1}\sum_{0}^{n-2}\frac{u_L}{h_i}\left[\ln\left(\frac{C^*-C_i}{C^*-C_{i+1}}\right)\right] \qquad (1)$$

where n is the total number of sampling-points along the column and $n = 5$ in our experiment system.

3. Result and discussion

3.1. Effects of the superficial gas/liquid velocity on mass transfer

The relationships between mass transfer and the superficial gas/liquid velocity are shown in Fig. 1. With an increase in the superficial liquid velocity, the mass transfer coefficient increases, but the superficial gas velocity has no significant effect on the mass transfer coefficient. The reason is, for the absorption of oxygen by water [12], the main resistance to mass transfer is in the liquid phase. Moreover, the mass transfer coefficient is nearly directly proportional to the superficial liquid velocity in the experimental range. This

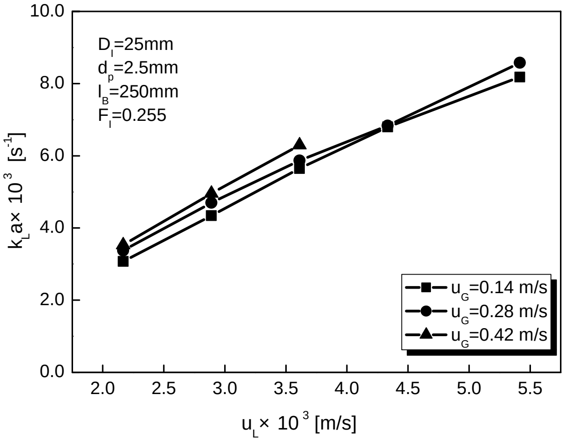

Fig. 1. Effects of the superficial gas/liquid velocity on mass transfer.

is different from the Sherwood and Semmelbauer correlations [13] of the mass transfer coefficient $k_{\mathrm{L}}a$ in a column packed with Raschig rings and Berl saddles, in which $k_{\mathrm{L}}a$ is directly proportional to $u_{\mathrm{L}}^{0.6\text{–}0.75}$. This may be due to the different operating range of the superficial liquid velocity. In this work, the operating range of the superficial liquid velocity is much narrower because the void fraction of the packed column in this work ($\varepsilon_{\mathrm{p}} = 0.35$–$0.4$) is much less than that of Raschig rings and Berl saddles ($\varepsilon_{\mathrm{p}} = 0.7$–$0.9$).

3.2. Influence of the internal on mass transfer

3.2.1. Volume fraction of the internal

The volume fraction of the internal is the ratio of the volume of internal to the total volume of the column. Fig. 2a and b show the influence of the volume fraction on mass transfer coefficient per column volume and per packing bed volume, respectively. It can be seen that at a low liquid velocity, the volume fraction has a small effect on the mass transfer coefficients per packing bed volume. The reason is that at a low liquid velocity the space between the packing particles is not totally filled with liquid and capillary effects draw the liquid into the catalyst bed, and so the liquid hold-up is only slightly changed with the variation of the volume fraction [4], as is the effective mass transfer area. However, at a high liquid velocity, the liquid load is above the load point, liquid fills both the catalyst bed and the internal, and the liquid hold-up decreases with an increase in the volume fraction of the internal [4]. Thus, the phase interface area between gas and liquid is decreased that causes the mass transfer coefficient to decrease.

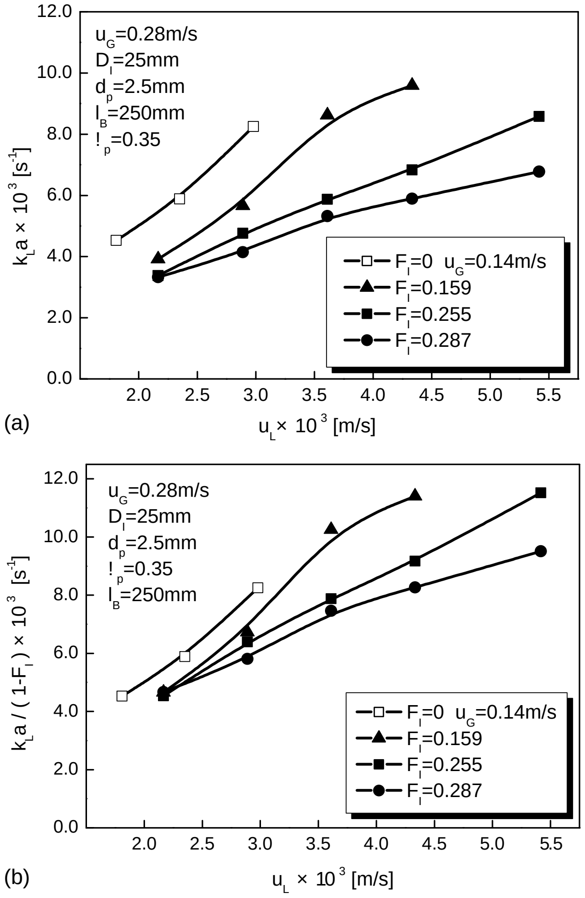

Fig. 2. (a) Influence of the volume fraction on the mass transfer coefficient per column volume. (b) Influence of the volume fraction on the mass transfer coefficient per packing bed volume.

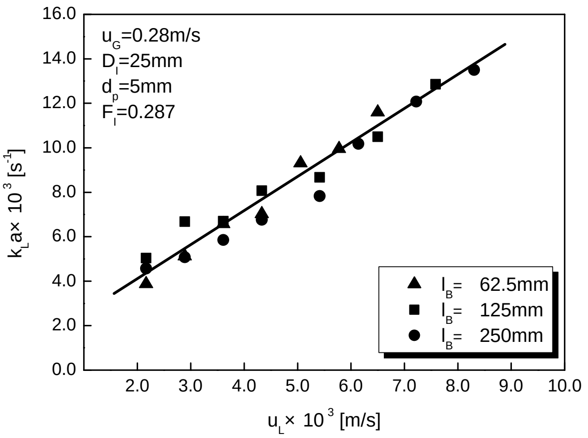

Fig. 3. Relationship between mass transfer and the height of the stages.

Even though the decreases in the mass transfer coefficients are due to the internal, on the other hand, the internal can avoid excessive pressure drop and “flooding”. A larger volume fraction of the internal results in a lower pressure drop [3,5]. Therefore, a suitable volume fraction is determined by a compromise between the hydrodynamics and mass transfer for each individual process.

3.2.2. The height of the stages

Fig. 3 presents the relationship between the mass transfer coefficient per packing bed volume and the superficial liquid velocity for different heights of the stages ($l_{\mathrm{B}} = 62.5$, 125, 250 mm). As indicated in the figure, a decrease in the height of the stages has no clear effect on gas–liquid mass transfer. This is because the change in the height of the stages mainly affects the gas flow behavior [3,5] and the gas velocity has no significant effect on mass transfer.

However, if the height of the stages is too large, some gas may bypass the packing bed through the spring; if the height of the stages is too small, operational flexibility will be reduced [3]. Thus, the height of the stages should also be determined according to each individual process.

3.2.3. The diameter of the spiral springs

The study of the effect of the diameter of the springs on the mass transfer was performed using three internals whose volume fractions are nearly 30%, which were made of nine spiral springs of diameter 25 mm, five spiral springs of diameter 35 mm, and three spiral springs of diameter 45 mm, respectively.

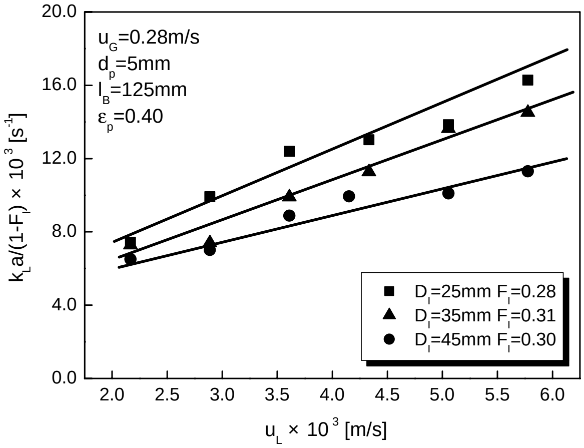

Fig. 4. Effect of the diameter of the spiral springs on mass transfer.

Fig. 4 presents the effect of the diameter of the spiral springs on mass transfer. The mass transfer increases with a decrease of the outer diameter of the spiral springs when the volume fraction of the internal parts, the liquid flux and the height of the stages are almost the same. For the internal which consists of more springs of smaller diameter, the gas distributes more evenly and contacts the liquid well, which benefits mass transfer and operational flexibility [3]. Therefore, a smaller diameter of the springs is preferred.

3.3. Effect of the particle size and packing bed porosity

Fig. 5 shows the effect of the particle size and packing bed porosity on mass transfer and liquid hold-up. When the average diameter of the porcelain balls are 2.5, 3.5 and 5 mm, the corresponding bulk void fractions are 0.35, 0.38, and 0.4, respectively. As shown in Fig. 5, the volumetric mass transfer coefficient per column volume decreases with an increase in the particle size and packing bed porosity, and this is in agreement with the literature. Larcachi et al. [14] stated that the gas–liquid interfacial area is the dominant factor and it will decrease with an increase in the particle size and packing bed porosity.

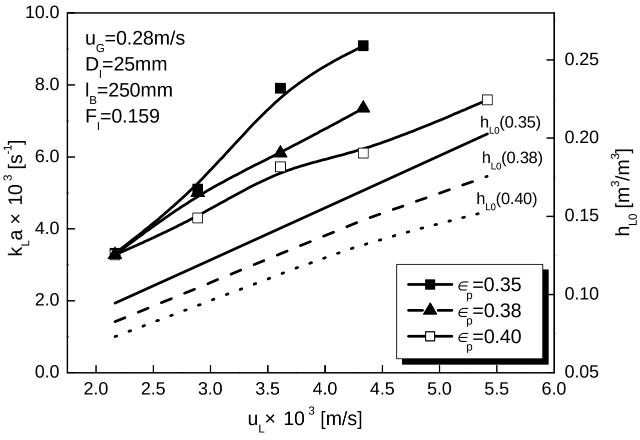

Fig. 5. Effect of the particle size and packing bed porosity on mass transfer and liquid hold-up.

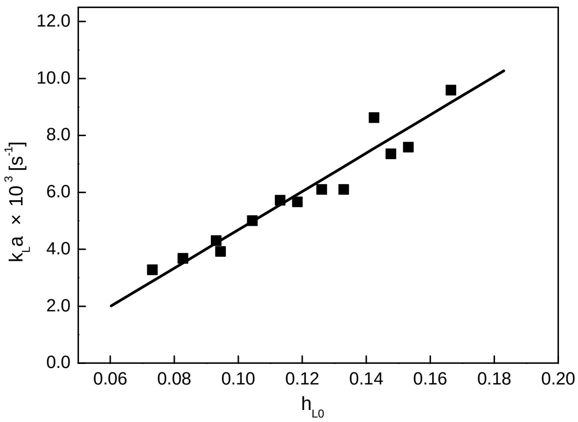

Fig. 6. Relationship between liquid hold-up and mass transfer.

In addition, the liquid hold-up depends on the porosity of the packing bed. With a decrease in the porosity, the liquid hold-up increases (Fig. 5). Fig. 6 presents the relationship between the mass transfer coefficient and the liquid hold-up (h_{L0}). It indicates that the mass transfer coefficient is directly proportional to h_{L0}. Obviously, with an increase in liquid hold-up, the gas–liquid interfacial area will increase, as does the mass transfer coefficient.

3.4. Correlation of the mass transfer coefficient

Many authors have studied mass transfer in a catalytic distillation column and have given different correlation equations to correlate the liquid phase mass transfer coefficient for different internals or packings, e.g. [7],

$$\frac{k_L a \hat{d}_p}{a_t D_L} = 0.149 \left(\frac{4\rho_L u_L}{a_t \mu_L}\right)^{0.30} Sc_L^{0.5} \tag{2}$$

However, for our new internal, the mass transfer depends on the internal structure, so we propose a mass transfer correlation as follows:

$$\frac{k_L a \bar{d}_I}{a_t D_L} = a_0 \left(\frac{4\rho_L u_L}{a_b \mu_L (1 - F_I)}\right)^{a_1} \left(\frac{\mu_L}{\rho_L D_L}\right)^{0.5} \tag{3}$$

F_I is the volume fraction of internals, and $\bar{d}_I$ the equivalent diameter distance, is twice the distance of gas flow between two adjacent springs [3].

$$\bar{d}_I = \sqrt{\frac{1}{N}}(D_c - D_I) \tag{4}$$

The parameters in Eq. (3) can be obtained by nonlinear regression, giving $a_0 = 0.4280$, $a_1 = 0.8748$, error $= \frac{1}{n}\sum_1^n |Y_{exp} - Y_{cal}|/Y_{exp} = 11.38\%$. Fig. 7 shows the comparison of the experimental values of the mass transfer coefficient with calculated values from Eq. (3). The experimental values are in agreement with calculated values.

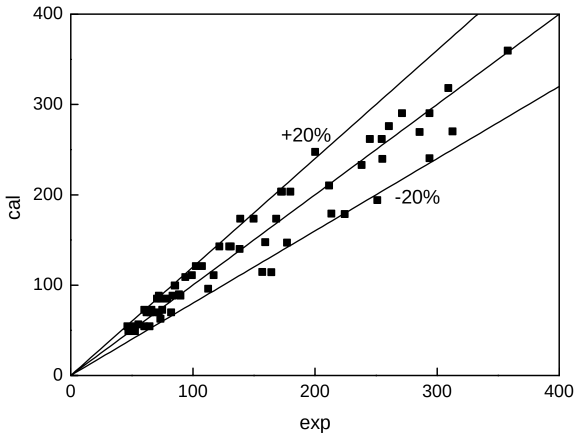

Fig. 7. Comparison of the experimental and calculated values of $k_L a \bar{d}_I / a_t D_L$.

4. Conclusions

For two-phase countercurrent flow in a packed column with the novel internal, which is made of several structured porous passages (springs), the volume fraction and the diameter of the springs have significant effects on the mass transfer coefficient, but the height of the stages has almost no effect on it. A suitable internal should be made of many springs of small diameter. The mass transfer coefficient is directly proportional to the liquid hold-up, which depends on the structural parameters of the internal, the superficial liquid flow velocity and the porosity of the packing bed.

Acknowledgements

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