Gas Holdup and Gas-Liquid Mass Transfer in Three-Phase Circulating Fluidized-Bed Bioreactors

Sung-Mo Son, Suk-Hwan Kang, Tae-Gyu Kang, Pyung-Seob Song, Uk-Yeong Kim, Yong Kang†, and Hyoung-Ku Kang∗

School of Chemical Engineering, Chungnam National University, Daeduck Science Town, Daejeon 305-764, Korea
*Youn San Corporation, Daejeon 306-010, Korea

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Abstract: Characteristics of gas holdup and gas-liquid mass transfer were investigated in the riser of a three-phase circulating fluidized-bed bioreactor where the nitrogenous component was removed from the synthetic wastewater. The diameter and height of the riser were 0.102 and 1.0 m, respectively. Anion polymer resin (diameter: 0.4 mm) was used as a fluidized biofilm media upon which the microorganism was adhered. The effects of the gas and liquid velocities and the holdup of the biofilm media on the gas holdup and gas-liquid volumetric mass transfer coefficient were examined. The gas holdup was determined by means of a dual electrical resistivity probe method. The gas-liquid mass transfer coefficient was recovered from the concentration profile of dissolved oxygen in the axial direction of the riser by adopting the axial dispersion model. We found that the gas holdup increased with increasing gas velocity, but decreased slightly with increasing the liquid velocity or the holdup of the biofilm media. The mass transfer coefficient increased with increasing the gas velocity or the holdup of the biofilm media, but did not change considerably with respect to the liquid velocity in the riser. The gas holdup and volumetric gas-liquid mass transfer coefficient correlated well in terms of the operating variables.

Keywords: gas holdup, mass transfer coefficient, three-phase, circulating fluidized-bed, bioreactor

Introduction

A fluidized bed can be utilized as a multiphase reactor, such as a bioreactor, because of its easy and continuous operation as well as the good contact among the multiphases [1-3]. In particular, a three-phase circulating fluidized bed has been proposed to circumvent the conventional three-phase fluidized bed, which is often restricted of its use when the fluidized solid particles are relatively small or light. The particles used as the catalyst, adsorbent, absorbent, or catalyst carrier in the beds are usually porous or small, so that they increase the contact area with the continuous and dispersed phases. In addition, the three-phase circulating fluidized bed, which can increase the heat and mass transfer coefficients at a higher range of liquid velocity, can be useful for regenerating the deactivated catalyst, adsorbent, or absorbent continuously and minimizing the dead zone in the reacting or contacting system by means of the circulating fluidization mode [4-9].

For practical applications of three-phase circulating fluidized beds, it has been understood that the values of the gas holdup and gas-liquid mass transfer are essential for the determination of the performance and efficiency of a bioreactor or contactor. The gas phase, which is needed to maintain the oxygen concentration in the bio-film reactor, exists as a dispersed phase in the continuous liquid medium. Although some information, such as the hydrodynamics and heat transfer, has been reported recently for three-phase circulating fluidized beds, little attention has so far been focused on the gas-liquid mass transfer in the bioreactors [10-15].

In the present study, the gas-liquid mass transfer has been investigated in the riser of a three-phase circulating fluidized-bed bioreactor. The gas holdup was also measured to analyze the effects of bubbling phenomena on the volumetric gas-liquid mass transfer coefficient in the...
bioreactor. The effects of the gas and liquid velocities and the holdup of the biofilm media on the volumetric gas-liquid mass transfer coefficient have been determined to obtain prerequisite knowledge for the design and scale-up of three-phase circulating fluidized-bed bioreactors or contactors.

Experiments

Experiments were carried out in the riser of a three-phase circulating fluidized bed that comprised three main sections, a riser column, solid recycle device, and down-comer, as can be seen in Figure 1 [6,7].

The diameter and height of the riser was 0.102 m (ID) and 1.0 m, respectively. A perforated plate, which was designed to introduce the liquid and gas phases into the riser at the same plane, was used as a distributor. The distributor was situated between the main column section and a 0.2-m-high stainless-steel distributor box into which water was introduced through a 0.025-m pipe from the liquid reservoir. Oil-free compressed air was fed to the column through a pressure regulator, filter, and a calibrated rotameter. The gas and liquid velocities were 0.0005 ∼ 0.005 and 0.01 ∼ 0.03 m/s, respectively. The biofilm media were returned to the bottom of the riser through the solid recycle device. An anion polymer resin (DIAIONSA 20AP), whose diameter and density were 0.4 mm and 1130 kg/m³, respectively, was used as the biofilm media.

A screen was installed at the top of the bioreactor to prevent the biofilm media from overflowing out of the reactor. Two down-flow connectors were situated between the riser and the down-comer to circulate the biofilm media, as well as the liquid medium, continuously [9-11]. Filtered compressed air and synthesized wastewater, whose composition is summarized in Table 1, were used as the gas and liquid phases, respectively.

Gas holdup in the riser of a three-phase circulating fluidized-bed bioreactor was determined by means of an electrical resistivity probe method [6,7]. The heights of the measuring gas holdup in the reactor were 0.1, 0.3, 0.5, and 0.7 m axially from the distributor. The local gas holdup was obtained using equation (1).
Table 1. Composition of the Concentrated Synthetic Wastewater

<table>
<thead>
<tr>
<th>Concentration (g/L)</th>
<th>Component</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.6</td>
<td>Glucose</td>
</tr>
<tr>
<td>9.7</td>
<td>CH₃COONa ⋅ 3H₂O</td>
</tr>
<tr>
<td>1.32</td>
<td>KH₂PO₄</td>
</tr>
<tr>
<td>0.006</td>
<td>FeCl₃ ⋅ 6H₂O</td>
</tr>
<tr>
<td>0.075</td>
<td>CaCl₂</td>
</tr>
<tr>
<td>2.8</td>
<td>(NH₄)₂SO₄</td>
</tr>
<tr>
<td>1.0</td>
<td>MgSO₄ ⋅ 7H₂O</td>
</tr>
<tr>
<td>0.1098</td>
<td>MnSO₄ ⋅ H₂O</td>
</tr>
<tr>
<td>2.1</td>
<td>NaHCO₃</td>
</tr>
</tbody>
</table>

Figure 2. Effects of gas velocity on the gas holdup in three-phase circulating fluidized bed bioreactor (εₛ = 0.015).

(εₑ) = \sum_{i} \frac{t_i}{t}  \hspace{1cm} (1)

The average value of the gas holdup was determined by averaging the local gas holdup throughout the axial direction. The individual liquid and solid holdups were obtained using equations (2) and (3) and the known value of the gas holdup.

\[ -\frac{\Delta P}{\Delta Z} = \left( \rho_G \varepsilon_G + \rho_L \varepsilon_L + \rho_S \varepsilon_S \right) g \hspace{1cm} (2) \]

\[ \varepsilon_G + \varepsilon_L + \varepsilon_S = 1 \hspace{1cm} (3) \]

The volumetric gas-liquid mass transfer coefficient in the riser of a three-phase circulating fluidized bed was determined by employing the axial dispersion model [5,16,17], which is based on the oxygen balance of the liquid phase in the axial coordinate. The final form of this model can be written as equation (4).

\[ \frac{1}{Pe} \frac{d^2 C}{dX^2} - \frac{dC}{dX} + St(C^* - C) = 0 \hspace{1cm} (4) \]

with the following boundary conditions

at X = 0, \[ C = C_0 + \frac{1}{Pe} \frac{dC}{dX} \bigg|_{X=0} \hspace{1cm} (5) \]

at X = 1, \[ \frac{dC}{dX} \bigg|_{X=1} = 0 \hspace{1cm} (6) \]

where,

\[ Pe = \frac{U_L L}{D_G \varepsilon_L}, St = (K_a L \varepsilon_L) \frac{L}{U_L}, X = Z/L \hspace{1cm} (7) \]

\[ C^* = a + bX = \frac{Py}{H} \hspace{1cm} (8) \]

\[ P = P_T[1 + \alpha (1 - X)], \alpha = \frac{\rho L e L_g L}{P} \hspace{1cm} (9) \]

In equation (8), \( C^* \) is the equilibrium concentration of oxygen. The constant values of \( a \) and \( b \) can be written as equations (10) and (11), respectively.

\[ a = \frac{y}{H} (\rho_T + \rho_T g e_L L) \hspace{1cm} (10) \]

\[ b = -\frac{y}{H} \rho_T g e_L L \hspace{1cm} (11) \]

Results and Discussion

The effects of the gas velocity on the mean value of the gas holdup can be seen in Figure 2. The gas holdup increases with increasing gas velocity in all cases studied, because the amount of gas injected into the riser increases with increasing gas velocity. However, the value of the gas holdup decreases slightly with increasing liquid velocity (Figure 3) or holdup of the biofilm media (Figure 4). It is understood that the increase of the liquid velocity leads to the increase of the liquid holdup in the riser; therefore, the gas holdup decreases slightly to compensate for the increased liquid holdup under a given operating condition. Similarly, the increase of the solid holdup due to the increase of the holdup of biofilm media results in the slight decrease of the gas holdup in the riser. It has been reported that the increase of the solid holdup in the bed can help the fluidized bed system to increase the turbulence by increasing the contacting effects between the solid phase and the liquid or gas phase [17,18].

A typical concentration profile of dissolved oxygen in
Gas Holdup and Gas-Liquid Mass Transfer in Three-Phase Circulating Fluidized-Bed Bioreactors

Figure 3. Effect of liquid velocity on the gas holdup in a three-phase circulating fluidized bed bioreactor ($U_L = 0.003$ [m/s]).

Figure 4. Effect of $\varepsilon_S$ on the gas holdup in a three-phase circulating fluidized-bed bioreactor ($U_L = 0.02$ [m/s]).

the riser of a three-phase circulating fluidized-bed bioreactor can be seen in Figure 5. The profile of dissolved oxygen fits the axial dispersion model well [16,17]. The volumetric gas-liquid mass transfer coefficient was obtained by fitting the analytical solution of equation (4) to the dissolved oxygen concentration profile in the axial direction of the riser [5,17].

The effect of the gas velocity on the gas-liquid mass transfer coefficient in the riser can be seen in Figure 6. The mass transfer coefficient increased with increasing $U_G$ in all of the cases studied. This result is due primarily to the increased contacting area between the gas and liquid phases for mass transfer arising from the increased gas holdup with increasing gas velocity.

Figure 5. Dissolved oxygen concentration profile in the axial direction of a column ($\varepsilon_S = 0.1$).

Figure 6. Effect of gas velocity on the value of $k_{La}$ in a three-phase circulating fluidized-bed bioreactor.

The effect of the liquid velocity on the volumetric gas-liquid mass transfer coefficient can be seen in Figure 7. The value of the mass transfer coefficient did not change considerably, but only slightly increases, with increasing
Figure 7. Effect of liquid velocity on the value of $k_L\alpha$ in a three-phase circulating fluidized bed bioreactor.

Figure 8. Effect of $\varepsilon_S$ on the value of $k_L\alpha$ in a three-phase circulating fluidized-bed bioreactor.

Figure 9. Correlation between the experimental and calculated values of the gas holdup in a three-phase circulating fluidized-bed bioreactor.

UL. It has been reported that in the beds of conventional three-phase fluidized beds, the mass transfer coefficient tends to decrease in the higher UL range owing to a significant decrease of the solid holdup in the beds. However, in the riser of the three-phase circulating fluidized-bed bioreactor, the solid holdup did not decrease considerably even in the higher UL range [1, 2, 19-21]. Therefore, the value of the mass transfer coefficient did not decrease, but increased only slightly, with increasing the liquid velocity.

The effect of the holdup of the biofilm media on the volumetric gas-liquid mass transfer coefficient can be seen in Figure 8. The value of the mass transfer coefficient increases with increasing the value of the solid holdup in all of the cases studied. This result is due to the increase of the holdup of the biofilm media helping the system to increase the hindrance effects on the continuous upward liquid flow, by increasing the solid holdup in the bed. Therefore, the mass transfer coefficient increases with increasing the value of $\varepsilon_S$.

The gas holdup and the volumetric gas-liquid mass transfer coefficient correlate well as a function of the operation variables in the riser of a three-phase circulating fluidized bed bioreactor, as represented by eqns (12) and (13), respectively (Figures 9 and 10).

$$\varepsilon_G = 0.15 U_L^{-0.047} U_G^{0.303} \varepsilon_S^{-0.05} \quad (12)$$

$$k_L\alpha = 0.069 U_L^{0.042} U_G^{0.162} \varepsilon_S^{-0.130} \quad (13)$$

The correlation coefficients of Eqns. (12) and (13) are 0.986 and 0.930, respectively. The eqns. (12) and (13) cover the following range of variables: $0.01 \text{ m/s} \leq U_L \leq 0.03 \text{ m/s}, 0.005 \text{ m/s} \leq U_G \leq 0.05 \text{ m/s}, \text{ and } 0.05 \leq \varepsilon_S \leq 0.21$. 

\[ \frac{d\varepsilon_S}{dt} = \frac{\varepsilon_S}{\tau_S} - \frac{\varepsilon_S}{\frac{U_L}{U_G}} - \frac{\varepsilon_S}{\frac{U_L}{U_G}} \frac{1}{\frac{1}{\varepsilon_B} - 1} \]

\[ \frac{d\varepsilon_B}{dt} = \frac{\varepsilon_B}{\tau_B} - \frac{\varepsilon_B}{\frac{U_L}{U_G}} - \frac{\varepsilon_B}{\frac{U_L}{U_G}} \frac{1}{\frac{1}{\varepsilon_B} - 1} \]
Conclusion

The gas holdup increases with increasing gas velocity, it does not change considerably with liquid velocity, and it decreases slightly with increasing the holdup of the biofilm media in the riser of a three-phase circulating fluidized-bed bioreactor. It has been found that the gas velocity is an important factor to determine the value of the mass transfer coefficient in the riser. The volumetric gas-liquid mass transfer coefficient increases with increasing gas velocity or holdup of the biofilm media, but it does not change considerably with variation of the liquid velocity in the riser. The gas holdup and the mass transfer coefficient correlate well in terms of the operation variables in three-phase circulating fluidized-bed bioreactors.

Nomenclature

- $C$: concentration of oxygen, mol/m$^3$
- $C^*$: equilibrium concentration of oxygen, mol/m$^3$
- $C_0$: initial oxygen concentration, mol/m$^3$
- $g$: gravitational acceleration, m/s$^2$
- $H$: Henry's constant, kPa L/mol
- $k_L a$: mass transfer coefficient, 1/s
- $L$: column height, m
- $\Delta P/\Delta Z$: static pressure drop in the riser, kPa/m
- $P$: total pressure, kPa
- $Pe$: Peclet number (dimensionless), $U_L L/D_Z \cdot \varepsilon_L$
- $P_T$: pressure at the top of the column, Pa
- $St$: Stanton number (dimensionless), $k_L a \cdot L/U_L$
- $t$: time, s
- $t_i$: exposure time of the probe to bubble, s
- $U$: fluid superficial velocity, m/s
- $X$: dimensionless distance defined in Eq. (7)
- $y$: oxygen mole fraction in gas phase
- $Z$: distance from the distributor, m

Greek letters

- $\alpha$: dimensionless parameter defined in Eq. (9)
- $\rho$: density, kg/m$^3$
- $\varepsilon$: holdup

Subscript

- $G$: gas phase
- $L$: liquid phase
- $S$: solid phase

References