Numerical modelling of a three-phase internal air-lift circulating photocatalytic reactor
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ABSTRACT

The photocatalytic degradation process has been recognized as a low-cost, environmentally friendly and sustainable technology for water and wastewater treatment. As a key carrier of the photocatalytic process, the semiconductor TiO₂ has been used in many studies. Analysis and modelling of hydrodynamics in the three-phase flow system can provide useful information for process design, operation and optimization of the three-phase flow photocatalytic reactor, which requires research on the mixing and flow characteristics of the interphase regions in the reactor. In this study, we modelled the hydrodynamics in an internal air-lift circulating photocatalytic reactor using an Eulerian multi-fluid approach. Localized information on phase holdup, fluid flow patterns and mixing characteristics was obtained. The simulation results revealed that the distribution of solid particle concentration depends on the flow field in the internal air-lift circulating photocatalytic reactor. The distance between the draft tube and wall of the reactor and changes in the superficial gas velocity \( (U_g) \) were found to be influential factors in reactor performance. The computational model developed could support optimizing reactor design to improve the hydrodynamics and provide guidance for scale-up.

Key words | air-lift, computational fluid dynamics, photocatalyst, photocatalytic reactor, water treatment

INTRODUCTION

During the past 30 years, research on and applications of advanced oxidation processes (AOPs) for treating gaseous streams and wastewater have considerably increased (Palmisano et al. 2009; Parrino et al. 2014). As one of the AOPs, the photocatalytic oxidation (PCO) technologies using titanium dioxide (TiO₂) with UV irradiation in the water treatment field have shown increasing advantages and received much attention from researchers and practitioners (Leblebici et al. 2015). Commercialized semiconducting TiO₂ in the anatase form has been widely used in the PCO process for wastewater treatment (Bhattacharyya et al. 2004; Fujishima & Zhang 2006). When the TiO₂ particle absorbs a photon with energy greater than the band gap energy (\( E_b \geq 3.2 \) eV), an electron-hole pair is generated and simultaneously remains separated, creating highly oxidizing hydroxyl (\(-\mathrm{OH}\)) and superoxide (\(-\mathrm{O}\)) radicals. Therefore, the process of using TiO₂ to treat wastewater has been a focus of research owing to its high photocatalytic efficiency in degrading a wide range of organic contaminants (Ochuma et al. 2007).

A number of studies have used suspended fluidized bed reactors to investigate the photocatalytic reaction (Kabir et al. 2006; Imoberdorf et al. 2008). Because the catalyst particles are in a suspended state in the reactor in the two-phase flow, increasing the contact area with the gas and liquid reactant in the reactor can improve the area available for illumination, which can greatly enhance the dynamic process of photocatalytic reaction. For three-phase flow (gas-liquid-solid) in the photocatalytic reactor, a region exists characterized by eddy flow, causing the concentration distribution of solid catalyst particles to be non-uniform. In order to make the distribution of photocatalyst in the reactor more uniform, a mixer or draft tube has been used. However, most scaled-up photocatalytic reactors employ air-lift instead to achieve a uniform mixing effect over the whole reactor. The internal air-lift circulating reactor reduces the dead zone in the reactor, while enhancing flow with a simple structure, low cost and economical operation (Razzak et al. 2007, 2009, 2010a, 2010b; Demarchi & Rodrigues...
In addition, the particle circulation rate can be controlled separately. As the flow behavior of the internal circulation system is complicated, being affected by the reactor structure and physical parameters, there is a great need to study the optimization of reactor design and operation (Razzak et al. 2007, 2009, 2010a, 2010b).

Following the development of computer technology, numerical methods and the theory of fluid mechanics, numerical simulation based on computational fluid dynamics (CFD) has been applied broadly in the study of reactor flow fields. There are many publications on using CFD methods for internal air-lift reactors. However, due to the complexity of the multiphase flow process, these studies have focused on the Reynolds averaged fluid mechanics of reactors, which is of very limited use for understanding local hydrodynamic conditions and the axial and radial distribution of phase holdup (Luo et al. 2011). Furthermore, only very few authors have studied the degradation characteristics of internal air-lift photocatalytic reactors using laboratory experiments (Hang et al. 2013). In addition, the hydrodynamics remain unclear when the catalyst is present in the heterogeneous PCO process, where complicated interactions between the gas, liquid and solid phases take place.

The objective of this study is to investigate the effect of the draft tube structure (draft tube diameter) and variations in gas velocity on fluid dynamics in the internal air-lift circulation photocatalytic reactor with regards to the parameters of gas holdup, solid holdup and liquid circulation velocity through the development of a numerical model. The results are expected to provide a basis for optimization and scale-up of the three-phase internal air-lift circulating photocatalytic reactor.

**MATHEMATIC MODEL**

**Hydrodynamic module**

Based on the principles of conservation of mass and momentum, the continuity for each phase in an internal air-lift circulation reactor was described. A three-dimensional CFD module was developed to calculate the local hydrodynamics of the gas-liquid-solid flow reactor. In this study, the Eulerian multi-fluid model was used to describe the flow behaviors of each phase. The phasic volume fraction $\alpha$ must satisfy the relation:

$$\sum_{i=1}^{n} \alpha_i = 1$$

where $n$ is the total number of phases; the subscript $i$ represents the gas, liquid or solid phase. The conservation equations are written by performing an ensemble average of the local instantaneous balance for each phase. The motion of each phase is governed by corresponding mass and momentum conservation equations.

**Continuity equations:**

$$\frac{\partial (\alpha_i \rho_i)}{\partial t} + \nabla \cdot (\alpha_i \rho_i \vec{u}_i) = 0$$

**Momentum equation:**

$$\frac{\partial (\alpha_i \rho_i \vec{u}_i)}{\partial t} + \nabla \cdot (\alpha_i \rho_i \vec{u}_i \vec{u}_i) = -\alpha_i \nabla P_i + \nabla \cdot \left( \alpha_i \mu_i \left( \nabla \vec{u}_i - (\nabla \vec{u}_i)^T \right) \right) + \vec{F}_i$$

**Interphase force models**

There are various interchange forces such as the lift and added mass forces during momentum exchange between the different phases. But these forces can be ignored in comparison with the drag and turbulent dispersion forces (Wiemann & Mewes 2005; Panneerselvam et al. 2008). Hence to reduce the computational complexity and time, only the drag and turbulent dispersion forces were considered in this study. Accordingly, the interfacial force $\vec{F}_i$ is approximated as the drag and interphase turbulent dispersion forces:

$$\vec{F}_i = \vec{F}_i^D + \vec{F}_i^T$$

where $\vec{F}_i^D$ and $\vec{F}_i^T$ are the drag and turbulent dispersion forces, respectively.

The drag force between the gas and liquid phases is calculated by the following equation:

$$\vec{F}_{lg} = C_D \rho \frac{3}{4} \frac{\alpha_l \bar{g}}{d_g} (\bar{\vec{v}}_g - \bar{\vec{v}}_l) (\bar{\vec{v}}_g - \bar{\vec{v}}_l)$$

where $C_D$ is the drag coefficient. The Grace relation is chosen for the drag force coefficient because bubbles are
experimentally observed to be spherical and dispersed, which is given as (Clift et al. 1978):

\[ C_D = \frac{4g d_b \Delta \rho}{3 \mu_T^2 \rho_l} \]  
\[ (6) \]

where \( d_b \) stands for the mean bubble diameter, \( \Delta \rho \) is the difference in density between the liquid and gas phases, and \( \mu_T \) is the bubble terminal rise velocity. It can be calculated as follows:

\[ \mu_T = \frac{\mu_l}{\rho_l d_b} M^{-0.149}(J - 0.857) \]  
\[ (7) \]

where \( M \) is the Morton number related to the fluid property, which is defined by:

\[ M = \frac{\mu_l^2 \Delta \rho}{\rho_l \sigma^3} \]  
\[ (8) \]

in which \( \sigma \) is surface tension. \( J \) is given by:

\[ J = \begin{cases} 0.94 H^{0.757} & 2 < H \leq 59 \\ 3.42 H^{0.441} & H > 59.3 \end{cases} \]  
\[ (9a,b) \]

where \( H \) is expressed as:

\[ H = \frac{4}{3} E_O M^{-0.149} \left( \frac{\mu_l}{\mu_{ref}} \right)^{-0.14} \]  
\[ (10) \]

where \( \mu_{ref} \) is the molecular viscosity of tap water at a reference temperature and pressure (Clift et al. 1978). \( E_O \) stands for Eotvos number as below:

\[ E_O = \frac{g \Delta \rho d_b^2}{\sigma} \]  
\[ (11) \]

The turbulent dispersion force, \( \mathbf{F}_{\text{td}}^T \), is calculated by the model of Lopez de Bertodano (Lopez de Bertodano 1991):

\[ \mathbf{F}_{\text{td}}^T = C_{\text{td}D} \frac{v_{tl}}{\sigma_{tl}} \left( \frac{\nabla a_g}{a_g} - \frac{\nabla a_l}{a_l} \right) \]  
\[ (12) \]

where \( C_{\text{td}D} \) is the momentum transfer coefficient for the interphase drag force, \( C_D \) is the drag coefficient as described above, \( v_{tl} \) stands for turbulent viscosity, and \( \sigma_{tl} \) is the liquid turbulent Schmidt number. \( a_g \) and \( a_l \) are the gas and liquid phase volume fractions, respectively.

The liquid-solid interfacial drag force is similar to that of gas-liquid:

\[ \mathbf{F}_{\text{ls}}^D = C_{\text{d},l} \frac{3 \rho_l a_s}{4} \left( \frac{\bar{v}_l - \bar{v}_s}{\bar{v}_l - \bar{v}_l} \right) \]  
\[ (13) \]

The drag coefficient is exerted by the solid phase on the liquid phase, where the drag coefficient \( C_{D,l} \) is proposed as calculated by the Schiller–Naumann drag model:

\[ C_{D,l} = \max \left( \frac{24(1+0.15R_e^{0.687})}{R_e}, 0.44 \right) \]  
\[ (14) \]

where

\[ R_e = \frac{\rho_l d_b |\bar{u}_l - \bar{u}_l|}{\mu_l} \]  
\[ (15) \]

The turbulent dispersion force, \( \mathbf{F}_{\text{ls}}^T \) for liquid-solid interaction is similar to that of gas-liquid:

\[ \mathbf{F}_{\text{ls}}^T = C_{\text{td}} \frac{v_{tl}}{\sigma_{tl}} \left( \frac{\nabla a_s}{a_s} - \frac{\nabla a_l}{a_l} \right) \]  
\[ (16) \]

**Turbulent model**

The turbulent dispersion force is the result of the turbulent fluctuations of liquid velocity. It approximates the diffusion of the gas phase from a higher fraction region to a lower fraction region by turbulent fluctuations of liquid velocity. The standard \( k-\varepsilon \) model for single-phase flows has been extended for the three-phase flows for simulating the turbulence in this study, which can be described as:

\[ \frac{\partial}{\partial t} (\alpha p_i k_i) + \frac{\partial}{\partial x_i} (\alpha p_i \bar{u}_i k_i) = \frac{\partial}{\partial x_i} \left[ \alpha_i \left( \mu_i + \frac{\mu_l}{\sigma_k} \right) \frac{\partial}{\partial x_i} k_i \right] + \alpha_i \rho_i - \alpha_i \rho_i \varepsilon_i \]  
\[ (17) \]

\[ \frac{\partial}{\partial t} (\alpha p_i \varepsilon_i) + \frac{\partial}{\partial x_i} (\alpha p_i \bar{u}_i \varepsilon_i) = \frac{\partial}{\partial x_i} \left[ \alpha_i \left( \mu_i + \frac{\mu_l}{\sigma_\varepsilon} \right) \frac{\partial}{\partial x_i} \varepsilon_i \right] + \alpha_i \varepsilon_i (C_{e1p} - C_{e2p\varepsilon}) \]  
\[ (18) \]

where \( C_{e1}, C_{e2}, \sigma_k \) and \( \sigma_\varepsilon \) are parameters in the standard \( k-\varepsilon \) model and the following values are selected: \( C_{e1} = 1.44, C_{e2} = 1.92, \sigma_k = 1.0 \) and \( \sigma_\varepsilon = 1.3 \). The turbulent viscosities \( \mu_l \) are computed from the following
equations (Sato et al. 1981):

\[ \mu_{t} = \mu_{t,s} + \mu_{t,b} \]  

(19)

where \( \mu_{t,s} \) is the conventional shear-induced turbulent viscosity, which is obtained by the standard k – \( \varepsilon \) model as:

\[ \mu_{t,s} = 0.09 \rho \frac{k^2}{\varepsilon} \]  

(20)

\( \mu_{t,b} \) is the bubble-induced and \( \mu_{t,p} \) is the particle-induced component of turbulent viscosity, which are given by:

\[ \mu_{t,b} = 0.09 \rho_l \alpha_g \frac{d_g}{d_l} |\vec{u}_g - \vec{u}_l| \]  

(21a)

\[ \mu_{t,p} = 0.09 \rho_l \alpha_s \frac{d_s}{d_l} |\vec{u}_s - \vec{u}_l| \]  

(21b)

The gas- and solid-phase turbulence is modelled using a zero equation model, in which gas turbulent viscosity is proportional to liquid-phase turbulent viscosity (Fan 1989):

\[ \mu_{t_g} = \frac{\rho_g \mu_t}{\rho_1 \sigma_t} \]  

(22a)

\[ \mu_{t_s} = \frac{\rho_s \mu_t}{\rho_1 \sigma_t} \]  

(22b)

where \( \sigma_t \) is a turbulent Prandtl number relating the dispersed phase kinematic eddy viscosities \( \mu_{t_g} \) and \( \mu_{t_s} \) to the continuous phase kinematic eddy viscosity \( \mu_t \).

**NUMERICAL PROCEDURE**

**Geometry and grid generation**

The reactor shown in Figure 1 was simulated in this study. It is conical bottomed, internally irradiated with the light source being placed in the central core axis, and has a draft tube with the internal circulation occurring around the light source. The position of the draft tube was set to give four different proportions (0.2, 0.3, 0.4, 0.5) of \( r/R \), where \( r \) is the distance between the draft tube and the light source, and \( R \) is the distance between the reactor wall and the light source. The \( r/R \) ratio determines the area ratio of the up-flow and down-flow regions in the reactor, but the area of aeration does not change in proportion to \( r/R \). First, the gas flows directly into the draft tube by aeration. After the contact of liquid and gas in the draft tube, the gas holdup inside is higher than outside of the draft tube, because the density of the multi-phase mixture in the draft tube is low, forming a density difference to drive the circulation in the reactor. Afterwards, the inner flow is upward while external flow is downward.

Given the characteristics of the axially symmetric structure of the reactor, the flow field simulation can be reasonably simplified by a 2D axial symmetry model. An unstructured numerical grid has been implemented with a total number of 14,017 elements, where the basic size of the grid is 1.5 mm and the boundary size is 0.2 mm.

**Initial and boundary conditions**

For the initial conditions, the reactor was filled with the liquid phase; the solid phase was uniformly distributed in the whole system at a concentration of 10 mg/L; and gas was fed from the bottom of the reactor. A uniform velocity \( U_g \) (0.2–0.5 m/s) was simulated for the gas inlet, while the liquid velocity was set at zero. The gas and liquid phases were air and water in the standard state, and the solid phase was photocatalyst particles. Detailed phase properties
used for the simulation are listed in Table 1. The viscosity of solid particles was obtained by the simple Brinkman formula and the diameter of solid particles was 0.02 mm. A pressure boundary condition was applied to the top of the reactor with an average static reference pressure of $1.01 \times 10^5$ Pa. In addition, the periphery and inner area was defined as the opening and degassing boundary condition, where only the gas phase can leave the computation domain. The solid was kept within the reactor (Jia et al. 2007). For the reactor walls and the light source boundary, the gas and solid phase were treated as free-slip, while for the liquid phase a no-slip boundary condition was applied (Sozzi & Taghipour 2006).

**Numerical procedure**

The numerical solution was obtained using the ANSYS Fluent 16.2 package. For all scenarios, unsteady and pressure-based solvers were used, and a least-squares cell based method was adopted to calculate gradients. The Euler method with a $k - \epsilon$ model for turbulence simulation was employed. The standard single-phase parameters were used for the $k - \epsilon$ model. The momentum equation was solved with a second-order upwind scheme. The pressure-velocity coupling was realized through the phase-coupled SIMPLE algorithm. A fully implicit upwind finite differencing scheme was used for the time integration. The time step was set at 0.1 s. A maximum residual convergent target of $1.0 \times 10^{-4}$ was set for all simulations. The monitoring section was set at the axial height of $H = 10, 50, and 90$ cm, and the change of average liquid velocity as well as the average gas velocity were observed, which is shown in Figure 1.

**RESULTS AND DISCUSSION**

**Distribution of gas holdup**

The reactor was initially full with liquid, and solid was well dispersed in the whole reactor. From the bottom part, gas was supplied with a certain $U_g$. Catalyst solid particles started to flow into the liquid phase with the gas. To check the distribution of gas holdup in the riser and downcomer of the reactor, an operating superficial gas velocity in the range of 0.2–0.5 m/s was used. Figure 2 shows the contours of the radial distribution of three axial zones of gas holdup for different $r/R$ ratios ($r/R = 0.2, 0.3, 0.4, 0.5$ expressed as A, B, C, D in the figure, respectively). As can be seen from the figure, gas holdup shows a pronounced increase at the center of the draft tube and on both sides of the light source following the increase of $U_g$. This is because the reactor aeration holes are mainly located in the center area of the bottom of the draft tube. When gas enters from the bottom, the joint effects of gas up-flow and liquid back-flow make the bubbles concentrate in the narrow center region.

Section-1 is 10 cm up from the bottom of the reactor. It can be seen that the gas holdup near the sidewall was the smallest for $r/R = 0.4$ (Figure 2(c)) among the four different $r/R$ ratios. For $r/R = 0.4$, the minimum value was 0.000161 when the gas velocity was $U_g = 0.5$ m/s, and the maximum value was 0.012500 when the gas velocity was $U_g = 0.3$ m/s. This indicates that good gas-liquid separation cannot be achieved with too large or too small an $r/R$ ratio, even with the assistance of the draft tube, which causes the decrease of liquid density in the downcomer region and thus affects the mixing efficiency in the internal air-lift circulating reactor. An appropriate proportion of $r/R$ can separate the gas and liquid effectively in the downcomer and thus improve the mixing performance of the reactor. In addition, high gas velocity is also advantageous for reducing gas holdup in the downcomer.

Section-2 is 50 cm up from the bottom of the reactor. It is in the main part of the reactor, where the light source is installed. In general, the gas holdup at Section-2 increases with $U_g$, which in turn decreases the gas holdup in the downcomer. However, it can be seen from Figure 2(d) that gas holdup in the downcomer increases with $r/R$ ratio when $U_g$ is higher than 0.4–0.5 m/s. Combining the results of Section-1 and Section-2, it is concluded that a small $r/R$ is not conducive to fluid circulation, while a large $r/R$ causes part of the air to flow upward through the downcomer due to the separation of the draft tube. Therefore, $r/R = 0.4$ is the optimal distance for the reactor in this study.

Section-3 is the gas outlet of the reactor, which is located at 90 cm from the bottom. The radial distribution of gas holdup in Section-3 is relatively uniform. This is because the momentum of the gas jet degrades along the

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<th>$r$ (mm)</th>
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axial height and the width of the gas jet increases; hence the gas holdup decreases in the central region and becomes uniform radially.

The distribution of radial gas holdup indicates that the mixing efficiency between gas and liquid is greatly improved by the downcomer tube inside the internal air-lift circulating reactor. The gas holdup is higher in the center than near the wall, which is similar to the traditional bubble reactor (Zhang & Zhao 2006).

**Distribution of solid holdup**

The axial and radial distribution of the solid holdup in the reactor is shown in Figure 3. For all the different axial heights (Section-1, Section-2 and Section-3), it can be found that the axial distribution of solid holdup decreases with increasing $U_g$; the radial distribution of solid holdup is low in the center while high near the sidewall. Both the axial distribution and radial distribution patterns of solid holdup are opposite to those of gas holdup. The reason is that when gas enters from the bottom of the reactor, the gas jet has large initial momentum, thus a high gas holdup but low solid holdup area is formed in the central region. The axial solid holdup decreases with increasing $U_g$ because high $U_g$ can cause high gas holdup, resulting in a more uniform distribution of solid particles. In addition, due to the sidewall effect, the solid holdup is relatively high near the sidewall. Part of the solid particles collide with the side wall and produce radial motion; however, their momentum is not strong enough to allow them to pass through the axially moving fluid, thus agglomeration occurs near the side wall. The difference of r/R ratio has little effect on the axial and radial distribution of solid holdup.

Figure 2 | The axial and radial distribution of gas holdup at different gas inlet velocities ((a)–(d) in the figure indicate r/R = 0.2, 0.3, 0.4, 0.5, respectively).
Distribution of liquid velocity

Figure 4 shows the axial and radial velocity of liquid under different $U_g$ in the riser and downcomer regions. It is observed that liquid velocity follows a centrally symmetric distribution for different $U_g$ and different r/R ratios in both the riser and downcomer regions. The maximum liquid velocity is located at the center of the symmetry axis, near the light source, and the peak value is 1.54 m/s (Section-3, $r/R = 0.3$, $U_g = 0.5$ m/s). The liquid velocity gradually decreases from the riser to the draft tube, up to $\sim 0.0957$ m/s. Although the liquid flow is not obvious and even downward liquid flow is observed in the riser region, its contribution to mixing efficiency is significant in the reactor.

Section-3 has the strongest gas momentum, and the maximum liquid velocity reaches 2.07 m/s (Section-1, $r/R = 0.5$, $U_g = 0.5$ m/s). In this section, the axial liquid velocity increases with $U_g$, but the radial liquid velocity in the downcomer region does not change along the axis. In Section-3, the liquid can pass through the light source area and reach the outlet of the reactor due to the comparatively strong momentum of gas. The axial liquid velocity increases with $U_g$ in this section. The radial liquid velocity is high in the center, and gradually decreases towards the sidewall in a non-uniform way.

Further analyses were conducted to reveal the mechanism behind the liquid velocity pattern. In the lower part of the reactor, the radial velocity of gas is high in the central area of the draft tube, leading to strong interphase forces between gas and liquid, hence high liquid velocity. Following the increase of the axial height, gas disperses toward the draft tube and sidewall of the light source, making the radial velocity of the liquid more uniform. The gas holdup is very low in the downcomer, with only a small number of air bubbles or even no air bubbles. Consequently, the interphase force between gas and liquid is quite small, and the radial velocity of liquid in the downcomer shows little variation with axial height.
Fluid flow patterns

Hydrodynamics, circulation and mixing properties in bubble reactors depend strongly on the flow pattern (Fan 1989). The flow of three-phase internal circulation can be observed from the upward movement of the bubbles in the reactor (Qi et al. 2009). The flow in the reactor under \( r/R = 0.4 \) and \( U_g = 0.5 \text{ m/s} \) reaches steady state after 40 seconds, and the velocity vectors of gas and liquid are presented in Figure 5. As can be seen from Figure 5(a), the gas starts from the inlet at the bottom, flows up around the side of the light source in the center of the reactor and then reaches the outlet. However, the liquid re-enters the downcomer near the outlet (Figure 5(b)) and maintains the liquid-phase circulation.

There is a pair of large vortices of liquid next to the light source when air bubbles flow up. The vortices are due to the gas momentum from the bottom and the liquid backflow in the downcomer. The presence of vortices clearly proves the internal circulation structure of the liquid phase, which can make the solid photocatalyst particles more uniformly distributed and achieve a better photocatalytic reaction with the irradiation from the light source.

CONCLUSION

The three-phase flow of gas-liquid-solid in an internal-loop air-lift photocatalytic reactor has been simulated. The results demonstrate that the highly efficient circulation and mixing in the reactor makes it suitable for application in photocatalytic reactions. From the study, it is also concluded that the CFD model can not only capture the general patterns of the three-phase flow in the reactor, but also provide detailed information on each section for different \( r/R \) ratios and \( U_g \). Therefore, the model can be used to optimize the
design of reactors and guide scaling-up strategy. Since the distribution of UV radiation is closely related to the adsorption and scattering of water, air, catalysts and pollutants, in a future study the fluid dynamic model will incorporate the distribution of light inside the reactor.

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