

## Article

## Structural parameter optimization for novel internal-loop iron–carbon micro-electrolysis reactors using computational fluid dynamics☆

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## ABSTRACT

It is generally recognized that internal-loop reactors are well-developed mass and heat-transfer multiphase flow reactors. However, the internal flow field in the internal-loop reactor is influenced by the structure parameter of the reactor, which has a great effect on the reaction efficiency. In this study, the computational fluid dynamics simulation method was used to determine the influence of reactor structure on flow field, and a volume-of-fluid model was employed to simulate the gas–liquid, two-phase flow of the internal-loop micro-electrolysis reactor. Hydrodynamic factors were optimized when the height-to-diameter ratio was 4:1, diameter ratio was 9:1, draft-tube axial height was 90 mm. Three-dimensional simulations for the water distributor were carried out, and the results suggested that the optimal conditions are as follows: the number of water distribution pipes was four, and an inhomogeneous water distribution was used. According to the results of the simulation, the suitable structure can be used to achieve good fluid mechanical properties, such as the good liquid circulation velocity and gas holdup, which provides a good theoretical foundation for the application of the reactor.

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## 1. Introduction

Iron–carbon micro-electrolysis consisting of scrap iron and activated carbon is considered a low cost, high-performance pre-treatment option for the purification of wastewater [1–3]. It can degrade the pollutants by the action of redox, flocculation, and adsorption of ferric ions [4].

Because of its advantages, iron–carbon micro-electrolysis has become an efficient pre-treatment tool and the technology is widely applied for wastewater from many industries, including the dye [5], petroleum [6], and coke [7] industries. However, there are practical problems with such reactor over long periods of operation. Two such problems are (1) the bed easily succumbs to passivation due to iron rust, resulting in short circuits and dead zones, and (2) scrap iron is consumed too quickly, causing the filling process to be time-consuming and labour-intensive.

To overcome the problems identified above, we have developed a novel reactor which combines Internal-Loop Airlift Reactors (ILARs) and iron–carbon micro-electrolysis (Fig. 1). In the developed reactor, the driving force is produced by the difference in the average gas

densities between the riser and down-comer, and the multiphase flow inside and outside of the draft-tubes generates circulation. In contrast to mechanical stirring devices [8], ILARs have no rotating parts and reduce cost. More importantly, there is an increase in the rate of particle collision, mixing, and mass transfer [9]. Our research has shown that our novel reactor possesses good hydraulic performance and ability of sewage treatment [5].

Relatively few studies have been devoted to the complexities of the flow field of internal-loop micro-electrolysis reactors. Even though empirical and semi-empirical correlations have been found predicting their hydrodynamic parameters [10–12], their design, operation, and control still present difficulties.

In recent years, a number of studies have focused on the structure optimization of the ILARs, which provides a good foundation for the design and amplification of the reactor. Generally, gas holdup and liquid velocity are usually used to evaluate the hydraulic performance of reactor with the experiment and simulation methods. The influence of operating parameters and physical properties on the hydrodynamic property was investigated by experiments (Tao Yang *et al.* [13], Cao *et al.* [14]). However, if the influences of structure parameters on the hydrodynamic property were only carried out by experiment, a large amount of material and labour force will be wasted. Therefore, the computational fluid dynamics (CFD) simulation methods have become the main way of studying the hydrodynamic property of the ILARs.

Davarnejad *et al.* [15] investigated the overall gas hold-up in the riser and down-comer of three different volume reactors, CFD simulation

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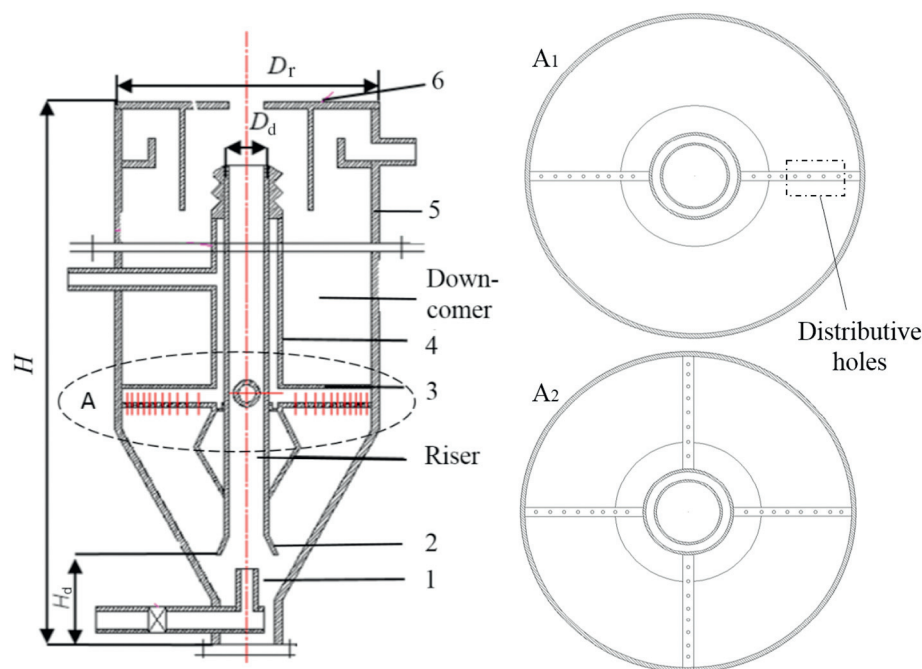


**Fig. 1.** Schematic of the structure of an internal-loop, iron-carbon, micro-electrolysis reactor. 1) gas sparger; 2) horn-mouth; 3) water inlet pipe; 4) draft-tube; 5) reactor wall; 6) reactor cap;  $H$ : height of reactor;  $H_d$ : axial position height of the draft-tube;  $D_r$ : diameter of reactor;  $D_d$ : diameter of draft-tube; A: local enlarged drawing;  $A_1$ : 2 water distribution pipes;  $A_2$ : 4 water distribution pipes.

results and experimental data showed that the gas holdup increases with enlarging of reactor scale, and the simulation results are in good agreement with the experimental data, and numerical simulation can further study the microscopic flow of gas-liquid two-phase flow. Chen and Bai [16] simulated the hydrodynamics in a cyclohexane oxidation airlift loop reactor using a two-fluid model. The results indicated the key hydrodynamic parameters are liquid circulation velocity and liquid circulation flux, and liquid circulation flux can enhance gas holdup in the down-comer. Wadaugorn *et al.* [9] systematically study hydrodynamic behaviors and mixing characteristics with a computational approach. Simulation results showed structural parameters (height-to-diameter ratio, down-comer diameter and reactor scale) have significant effects on hydrodynamic parameters, and the detailed distribution of flow field in the reactor is obtained [17]. However, in specific fields, the range of the structural parameters is certain scope, but not universally applicable to any ILARs.

In this study, a two-dimensional (2D) numerical simulation was used to study the influence of the height-to-diameter ratio ( $H/D$ ), diameter ratio of riser to down-comer ( $D_r/D_d$ ), and the axial position height of the draft-tube ( $H_d$ ) water distribution ways to the hydrodynamic parameters of a novel internal-loop iron-carbon micro-electrolysis reactor. Due to the influence of the number of distribution pipes, a 2D model cannot be carried out in 2D model; thus, a three-dimensional (3D) model was employed to discuss this structure parameter and verified the reliability of 2D simulation results through the height-to-diameter ratio.

## 2. Methodology

### 2.1. Geometry and meshing

The geometry was created with the software GAMBIT for Fluent Pre-processing. The initial structural dimensions were a height ( $H$ ) of 600 mm, a reactor diameter ( $D_r$ ) of 300 mm, and the  $H/D$  of 2:1, 3:1, and 4:1. The draft-tube had a diameter ( $D_d$ ) of 30 mm and a height ( $H_d$ ) of 400 mm. The water distributor comprised two pipes, with

uniform water distribution, located 90 mm from the draft-tube's bottom. The diameter of the distributor holes was 3 mm.

The quality of the CFD simulation grid has a critical impact on the simulation results, as does the efficiency of computing [18]. During the meshing process, an ideal division method not only produces high-quality grids, but also conserves computational resources. Close attention must therefore be paid to the division of grids.

In order to maintain a high-quality grid for the reactor analysis, polygonal mesh generation was used. The "Map" meshing method was used to mesh the geometry with the exception of the cone part. As shown in Fig. 2, the mesh interval size was 3 mm (cell number 16502, 13925, and 14942).

### 2.2. Boundary condition

In this simulation, the boundary conditions at the gas-inlet of the model were set as the velocity inlet. The turbulence intensity was 5%, the hydraulic diameter was 20 mm, and the gas volume fraction was one. The outlet was set to a pressure outlet with a turbulence intensity of 5%, and a hydraulic diameter of 20 mm. The water distributor was used as the velocity inlet boundary. Its turbulence intensity was 5%, and the hydraulic diameter was 3 mm. The moving wall was applied at the top two sides of the reactor to better simulate the impact of water flow on the top cap. The other walls were defined as the no-slip boundary. Based on subsequent calculation, the liquid velocity was set to  $0.04 \text{ m} \cdot \text{s}^{-1}$ , and the gas velocity was  $0.7 \text{ m} \cdot \text{s}^{-1}$ .

### 2.3. Mathematical model

The Reynolds number was used to select the turbulence model for the fluid flow. When the Reynolds number is less than 2100, the flow in the pipe is a laminar flow [19]. When the Reynolds number is greater than 4000, the flow in the pipe is turbulent. According to Eq. (1), the Reynolds number at the entrance of the reactor was 11045.

$$Re = \frac{\rho v d}{\mu} \quad (1)$$

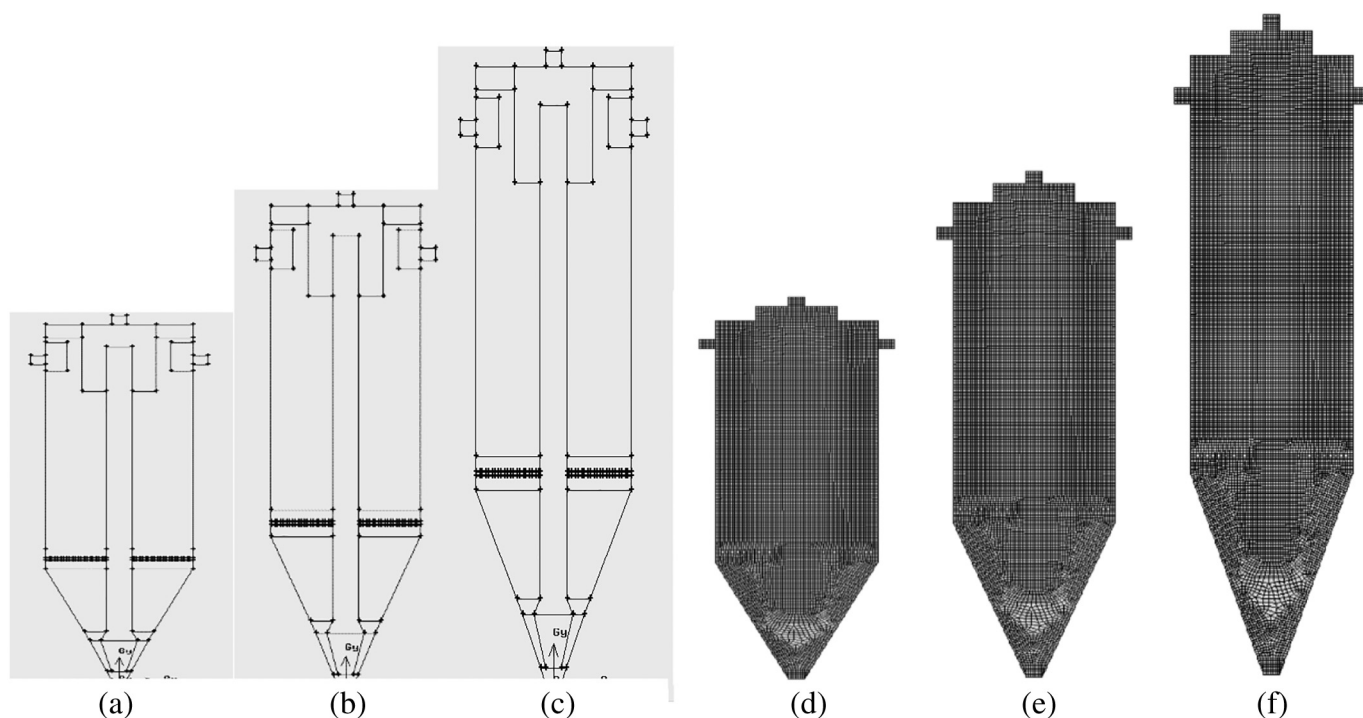


Fig. 2. Reactor geometry for  $H/D$ : a) 2:1; b) 3:1; c) 4:1. The geometry's meshing  $H/D$  ratios were d) 2:1; e) 3:1; and f) 4:1.

where  $\rho$  is the liquid phase density,  $v$  is the liquid velocity,  $d$  is the diameter of inlet,  $\mu$  the liquid phase viscosity.

The factors affecting simulation results come mainly from the shear stress, eddy, and separation produced by the two-phase circulation flow. However, the effect of the Near-Wall Grid Size of ILARs on the fluid flow can be ignored. Hence, the standard  $k$ - $\varepsilon$  model of turbulence was selected. This model was proposed by Launder and Spalding [20] in 1972, owing to its advantageous simulation of a slightly diffused flow fluid as well as its economic efficiency and relative stability. It has already become the primary tool for the calculation of engineering flow fields.

The turbulent kinetic energy  $k$  and the dissipation rate  $\varepsilon$  for the standard equation  $k$ - $\varepsilon$  model are as follows [21]:

$$\frac{\partial}{\partial t}(\rho_k) + \frac{\partial}{\partial x_j}(\rho_k u_j) = \frac{\partial}{\partial x_j} \left[ \left( \mu + \frac{\mu}{\sigma_k} \right) \frac{\partial k}{\partial x_j} \right] + G_k + G_b - \rho \varepsilon - Y_M + S_k \quad (2)$$

$$\frac{\partial}{\partial t}(\rho_\varepsilon) + \frac{\partial}{\partial x_j}(\rho_\varepsilon u_j) = \frac{\partial}{\partial x_j} \left[ \left( \mu + \frac{\mu}{\sigma_\varepsilon} \right) \frac{\partial \varepsilon}{\partial x_j} \right] + C_{1\varepsilon} \frac{\varepsilon}{k} (G_k + C_{3\varepsilon} + G_b) - C_{2\varepsilon} \rho \frac{\varepsilon^2}{k} \quad (3)$$

where coefficient  $C_{1\varepsilon} = 1.44$ ,  $C_{2\varepsilon} = 1.92$ ,  $C_{3\varepsilon} = 0.09$ ,  $\sigma_k = 1.0$ , and  $\sigma_\varepsilon = 1.3$ .

In numerical simulations, the multiphase flow model should be introduced to describe the force and motion between two or more phases. The hydrodynamic modelling of two-phase flow uses a dual approach: a) the Euler-Lagrange formula and b) the Euler-Euler formula. If the two phases are treated as a continuous phase, the first approach is chosen; otherwise, the latter is selected. The two phases are composed of a primary phase (water) and secondary phase (air) within the ILAR. Both are treated as continuous (similar to Xu *et al.* [17]). This volume-of-fluid (VOF) simulation under the Euler-Euler approach, which can model two or more immiscible fluids, relies on solving a single set of momentum equations and tracking the volume fraction of each of the fluids throughout the domain [22].

### 3. Results and Discussion

#### 3.1. Grid independence

This simulation used differently sized grids, which helps exclude the influence of grid size on the simulation results. In other words, this reinforces the idea that hydrodynamic parameter differences were caused by structural or operating parameters, rather than the grid quality [23]. The simulated liquid velocities with different cell sizes are shown in Fig. 3. As the sizes of the cells increase from 1 mm to 5 mm, the velocity changes approximately  $0.02 \text{ m} \cdot \text{s}^{-1}$  for every 1 mm. For a cell size less than 3 mm, the hydrodynamic parameters were predominantly stable. Therefore, this simulation was conducted with a cell size of 3 mm.

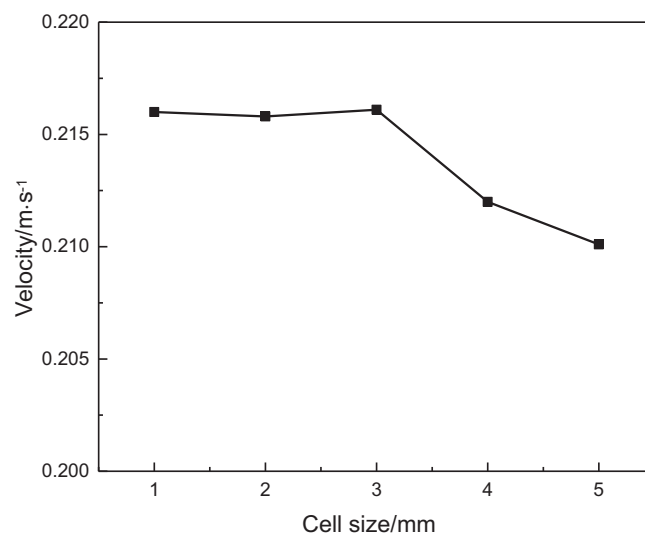


Fig. 3. Verification of grid independence.

### 3.2. CFD results

#### 3.2.1. Effects of height-to-diameter ( $H/D$ ) ratio

Fig. 4 shows the radial distribution of liquid velocities at different  $H/D$  ratios at an axial height of 300 mm. We can see that with an increasing  $H/D$  ratio, the liquid circulation velocity increases in the riser zone, and the liquid velocity decreases in the down-comer zone. This makes the liquid velocity uniform at radial and axial distributions, increases the bubble residence time and total gas holdup [24], and improves the degree of liquid circulation. Fig. 5 shows the velocity distribution of the liquid phase at different  $H/D$  ratios. With an increasing  $H/D$  ratio, the velocity distribution of the gas–liquid phases is more uniform in the down-comer zone, which is useful to the iron–carbon micro-electrolysis.

When the  $H/D$  ratio is 2:1, the maximum liquid velocity in the riser is about  $0.7 \text{ m} \cdot \text{s}^{-1}$ , which is half at the  $H/D$  ratio of 4:1. The liquid velocity outside the draft-tube was higher, up to  $0.5 \text{ m} \cdot \text{s}^{-1}$ , and the average liquid velocity on the left side was higher than the right side. This suggested that the liquid velocity distribution is not uniform. Fig. 5 illustrates the liquid velocity contour of different  $H/D$  ratios. It can be seen that there is obvious bias flow at the bottom of the reactor, a

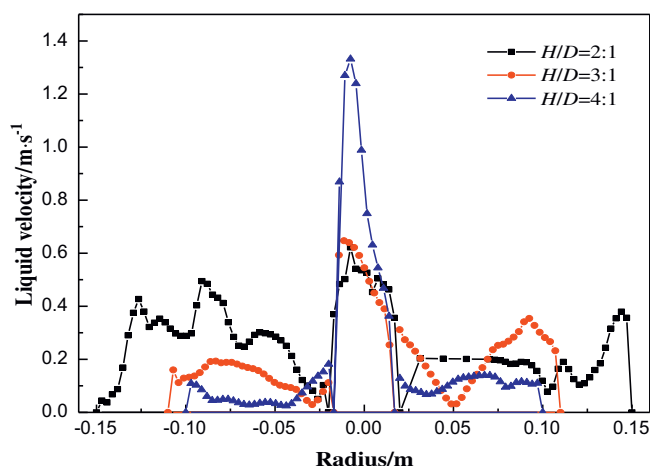


Fig. 4. Radial distribution of liquid velocity of different  $H/D$  ratios.

portion of the air spilling directly through the two sides of the draft-tube (see from Fig. 5(a)) under the wall effect. As a result, the liquid velocity in the riser is reduced, which ultimately affects the gas–liquid circulation performance.

With an increase in the  $H/D$  ratio to 3:1, the reactor fluid flow form was better, and the speeds within the draft-tube considerably improved. The liquid velocity distribution is more uniform here, but a portion of the gas is still entering the down-comer (as shown in Fig. 5 (b)). When the  $H/D$  ratio is approximately 4:1, the maximum liquid velocity in the draft-tube is about  $1.4 \text{ m} \cdot \text{s}^{-1}$ , which is about 75% higher than that at 3:1. The flow state is similar to the plug flow type, with the maximum velocity of the gas–liquid mixture concentrated in the draft-tube. The gas holdup is more uniform in the reactor, and the refracted flow is virtually eliminated. This is the ideal state of mass transfer. Therefore, the best  $H/D$  ratio is 4:1.

#### 3.2.2. Effects of the diameter ratio of riser to down-comer ( $D_r/D_d$ )

In this section, we examine the effect of the  $D_r/D_d$  ratio using an  $H/D$  ratio of 4:1. Three draft-tube diameters were used: 28 mm ( $D_r/D_d = 7:1$ ), 25 mm ( $D_r/D_d = 8:1$ ), and 22 mm ( $D_r/D_d = 9:1$ ). These diameters were examined with the value of the other structural parameters unchanged. From Fig. 6(a), we can see that when a 7:1 diameter ratio is used, much gas enters the down-comer zone, inducing that it is difficult for the wastewater to form a cycle inside the reactor.

It can be seen from Fig. 7 that as the ratio increases, the average liquid velocity rises in the riser. When the ratio is 9:1, the maximum velocity is about  $2.13 \text{ m} \cdot \text{s}^{-1}$ , which was almost one time higher than that of the  $D_r/D_d$  ratio of 7:1. A good flow regime can generate in the down-comer zone when the diameter ratios are 8:1 and 9:1. The distribution of the gas–liquid two-phase flow is especially symmetric when the diameter ratio is 9:1.

#### 3.2.3. Effects of the axial position height of the draft-tube ( $H_d$ )

The axial position height of the draft-tube ( $H_d$ ) refers to the vertical distance from the reactor's bottom to the draft-tube and mainly affects the gas and liquid circulation in the reactor. Models with  $H_d$  at 50 mm, 90 mm, and 150 mm were studied in our simulation. From Fig. 8, more gas enters the down-comer zone when the  $H_d$  was increased. Especially at 150 mm, only a small portion of the gas can enter the riser zone. Hence, a large amount of gas that enters the down-comer zone increases the resistance of fluid flow.

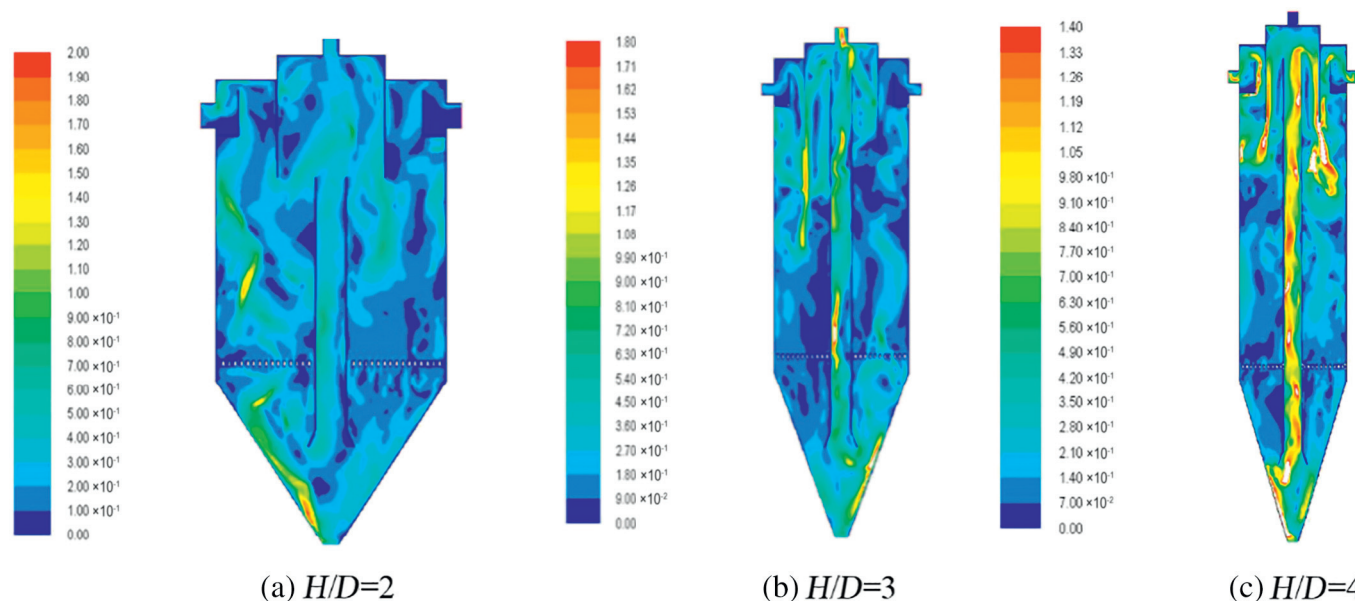


Fig. 5. Liquid velocity contour of different  $H/D$  ratios.

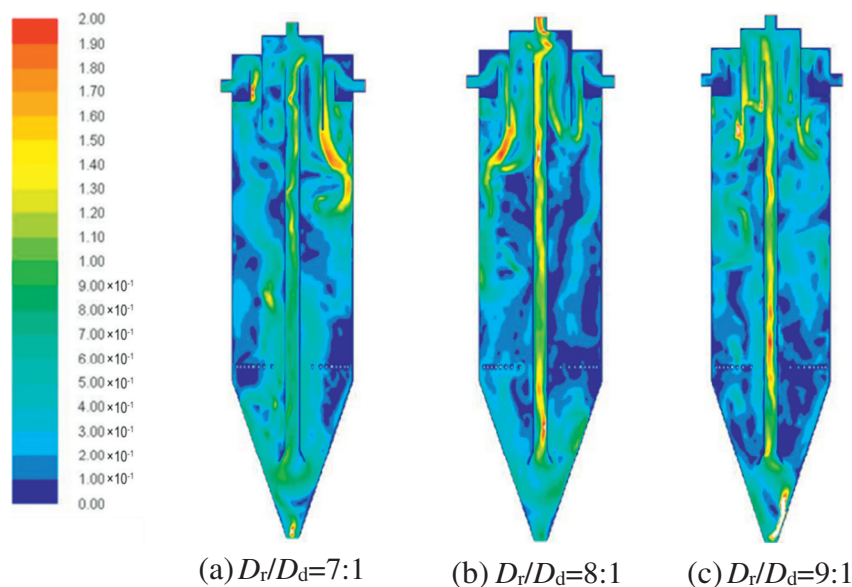


Fig. 6. Liquid velocity contour of different  $D_r/D_d$  ratios.

Hence the height of the draft-tube can affect the flow direction of the gas. In order to quantitatively investigate the effect of  $H_d$  on the liquid velocity, three cross sections of difference positions ( $H_1$ : the connection area of draft-tube and horn-mouth;  $H_2$ : the end edge of the horn-mouth;  $H_3$ : the middle position between  $H_1$  and the bottom of the reactor) were selected. Although  $H_d$  is different, three pictures (Fig. 9 (a)–(c)) have the same trend that the average liquid velocity possesses high level (about  $0.7 \text{ m} \cdot \text{s}^{-1}$ ) at the location of  $H_3$ . However, the liquid velocity of Fig. 9(a) is obviously lower than (b) and (c) at  $H_1$ . As mentioned earlier, with the decrease of  $H_d$  more gas will enter the draft-tube and then improve liquid velocity. However, the maximum liquid velocity is only  $1.8 \text{ m} \cdot \text{s}^{-1}$ . It may be that the  $H_d$  is too low to raise the circulating resistance of the down-comer.

When the  $H_d$  is 150 mm, the maximum liquid velocity at the  $H_1$  position is about  $2.6 \text{ m} \cdot \text{s}^{-1}$ , which is better than that (about  $1.8 \text{ m} \cdot \text{s}^{-1}$ ) when the  $H_d$  is 50 mm. However, it is lower than that when  $H_d$  is 90 mm. This phenomenon can also be seen from the liquid velocity contour of different  $H_d$ , as shown in Fig. 8. It may be due to the gas enters into the down-comer zone when the  $H_d$  is higher than a certain value,

which will increase the resistance of circulating flow. Therefore, the  $H_d$  has a significant impact on the circulation driving force, as the results reported by Chen and Bai [16]. The  $H_d$  was set at 90 mm in the follow-up study.

### 3.2.4. Effects of water distribution

The function of the water distributor is to distribute the water evenly within and make full use of the packing layer. The flow rate of the fluid within the water distributor should be uniform, neither too fast nor too slow. In this section, only the radial distribution of liquid was investigated, therefore, two-dimensional simulation with 2 pipes was selected. We found that uneven distribution often appears in the simulation as a result of different structural changes, short circulation currents, and bias within the current. In Fig. 10 (where the liquid velocity of the distribution pipe constitutes the vertical axis and the radial distance is shown as the horizontal), we can see that the average liquid velocity of the uniform water distribution (the inlet holes in the pipes have an equal distance) is low, at approximately  $0.3 \text{ m} \cdot \text{s}^{-1}$ . This makes it difficult to

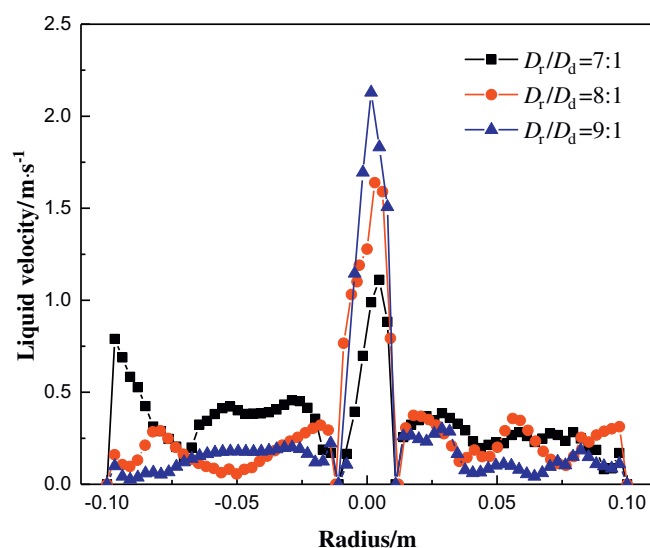


Fig. 7. Radial distribution of liquid velocity at different  $D_r/D_d$  ratios.

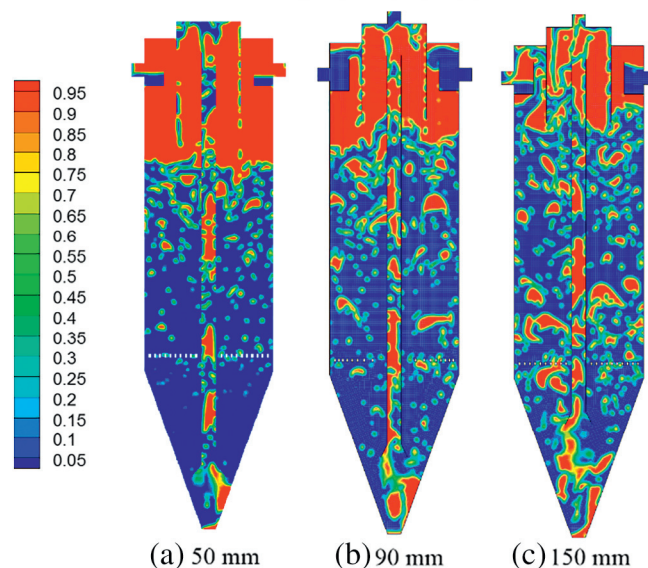


Fig. 8. Liquid velocity contour of different  $H_d$ .

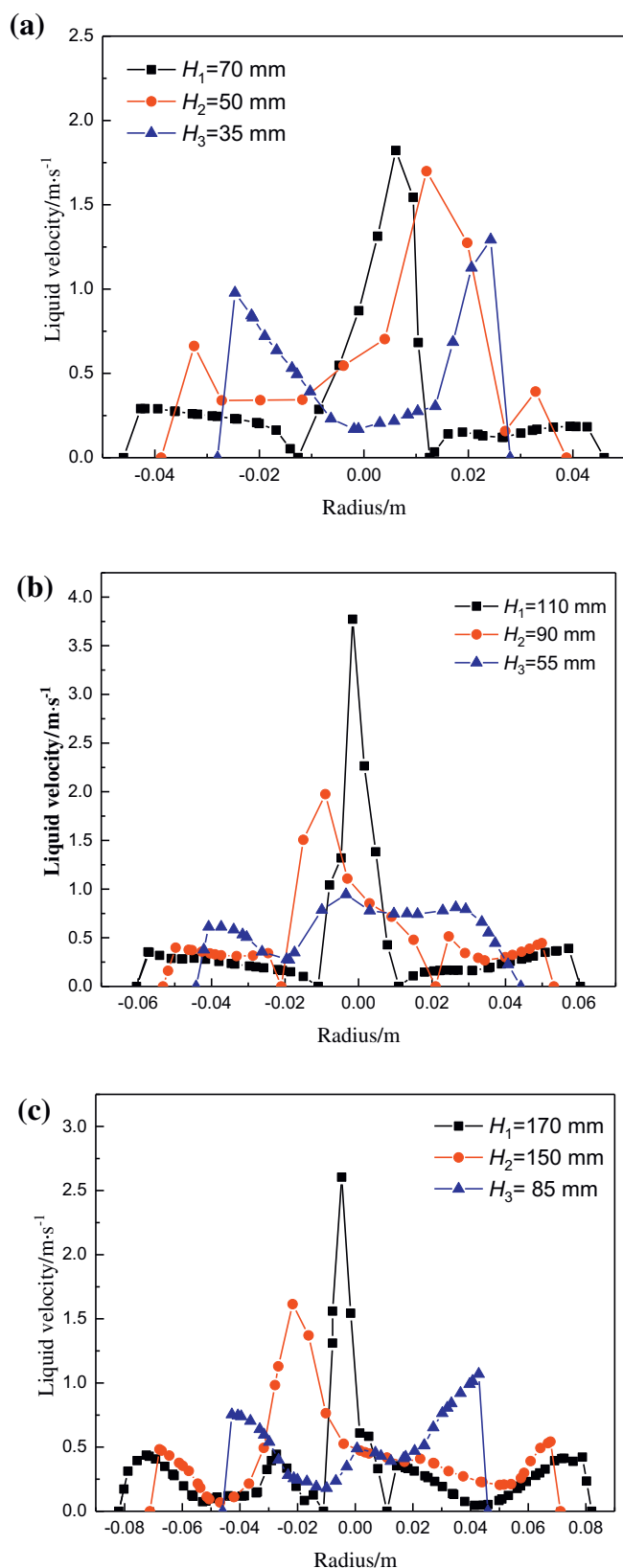


Fig. 9. Radial distribution of liquid velocity at three different axial heights in reactor. ((a)  $H_d = 50$  mm; (b)  $H_d = 90$  mm; (c)  $H_d = 150$  mm.)

form a good cycle. By changing the uniform distribution of water to a calculated non-uniform distribution (non-uniform water distribution has an equal distribution area), the orifice spacing is the largest at the

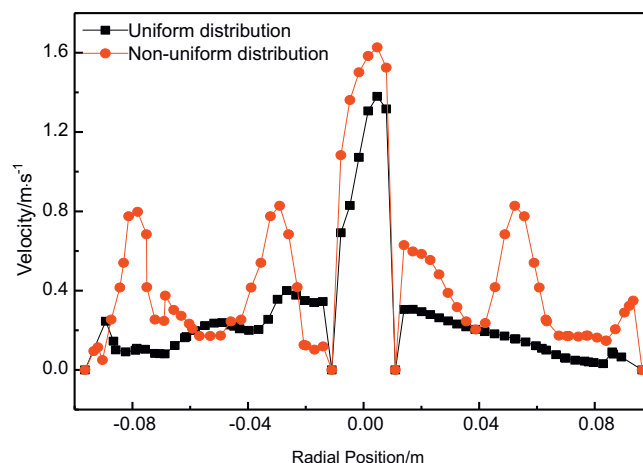


Fig. 10. Velocity distribution of uniform and non-uniform distribution of water.

centre of the draft-tube and gradually decreases towards the reactor's side wall. The liquid velocity is above  $0.4 \text{ m} \cdot \text{s}^{-1}$ , hence uniform and full circulation of water can be achieved.

### 3.2.5. Effects for the number of water distribution pipes

The water distributor, located at the bottom of the reactor, is one of the most important parts of the internal circulation of iron-carbon micro-electrolysis reactors. Liquid outflows from the water distribution pipes into the reactor and mixes with air bubbles; then, gas-liquid cycling occurs in the reactor. Therefore, the flow number and distribution holes of the water distribution pipe reactor also have an important impact on the water distributor [25].

The uniform distribution of wastewater in the down-comer zone is desired, which involves three-dimensional space. Therefore, the GAMBIT software for three-dimensional modelling and mesh generation was employed to investigate the influence of number of water distribution pipes on the wastewater distribution. A high-accuracy hexahedral mesh division was used to allow for better simulation regarding how the number of water pipes influences the reactor. The overall liquid phase circulation speed represents the hydraulic performance of the reactor to a certain extent and is related to the apparent gas velocity, the geometric structure, and the physical properties (viscosity, density, etc.) of the liquid phase. As can be seen from Fig. 11, the overall liquid speed of four tubes is clearly greater than that of two (because of the greater number of water holes). An increase in the

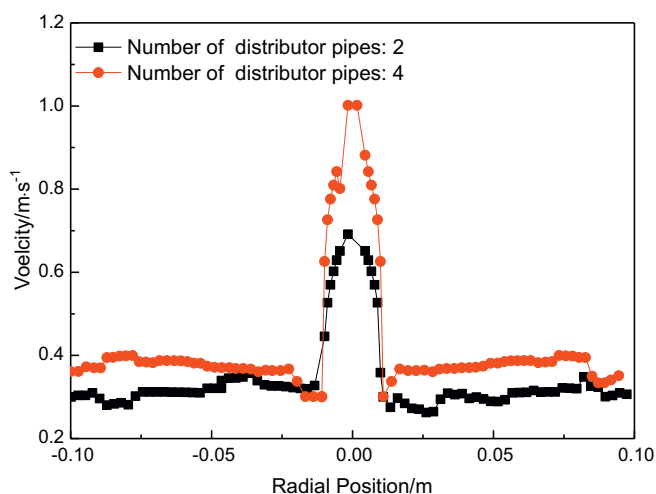


Fig. 11. Effect of the number of water distributing pipes on the overall liquid velocity.

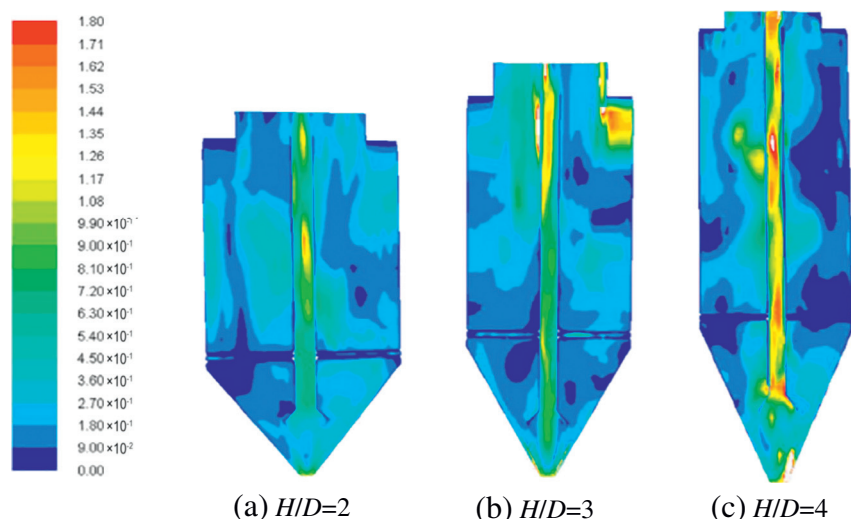


Fig. 12. Liquid velocity contour of different  $H/D$  ratios.

number of water holes corresponds to a more uniform liquid inflow. This means that less gas is carried into the down-comer by the liquid phase, lowering the gas content of the down-comer, increasing the gas content within the riser, and making the gas–liquid circulation more uniform.

### 3.3. Three-dimensional verification

3D simulation of the height-to-diameter ratio was carried out to verify the reliability of the 2D model. The same model and boundary conditions as the 2D simulation were used. Fig. 12 shows the effect on the liquid velocity at  $H/D$  ratios of 2:1, 3:1, and 4:1. At the same superficial gas velocity, the velocity in the draft-tube increases with increasing  $H/D$  ratio. When the  $H/D$  ratio is 4:1, the average liquid velocity within the draft-tube is approximately  $1.35 \text{ m}\cdot\text{s}^{-1}$  for an increase of approximately 35% over an  $H/D$  ratio of 2:1. It can also be seen from Fig. 13 that as  $H/D$  increases, the liquid velocity in the down-comer zone gradually increases.

In the three groups of simulations, the results indicate that the liquid velocity value of the riser is directly proportional to the liquid velocity of the down-comer zone, with the velocity of the down-comer zone being highest at 4:1. The circulation effect is most obvious at these

levels. This phenomenon shows that the  $H/D$  ratio has an obvious influence on the flow field and that the influence law is the same as in the 2D simulations. Hence, the 2D simulation can be replaced by the 3D simulation under certain conditions, reducing time and ensuring accurate results.

### 4. Conclusions

For internal-loop iron–carbon micro-electrolysis reactors, the VOF two-fluid model is an efficient CFD mathematical model for the complexities of a flow field inside the reactor. The model accurately reflects the numerical values of the gas–liquid, two-phase flow characteristics within the reactor. It can, therefore, be used as the basis of understanding for the fluid mechanics performance of the reactor, and as a basic tool for industrial amplification design.

The numerical calculations show that the  $H/D$  ratio makes the flow of the gas and liquid more uniform. The ratio of the reactor to the draft-tube diameter ( $D_r/D_d$ ) predominantly affects the axial velocity within the internal circulation tube and the circulation effect of the reactor. The diameter ratio is optimized at 9:1. The axial height of the draft-tube predominantly affects the degree of gas–liquid mixing in the mixing zone. If it is too high, the gas will enter the down-comer zone and will affect circulation. If the diameter ratio is too low, it will increase the resistance of the circulation. The bias current may be reduced to a certain extent by uneven water distribution.

The number of water distribution pipes was analysed via 3D simulation, which determined that a four-pipe water distribution system is better than a two-pipe system. The simulation results confirm that an inhomogeneous water distribution is obtained when the  $H/D$  ratio is 4:1,  $D_r/D_d$  is 9:1,  $H_d$  is 90 mm, and the number of water pipes is four.

Through simulation, we ascertained that the relationship between the flow field distribution, velocity variation, and mass transfer offers a good foundation for future research on the internal reaction mechanism. CFD numerical simulation is useful for the continued exploration of different structural designs and operational parameters regarding increasing flow models for industrial production.

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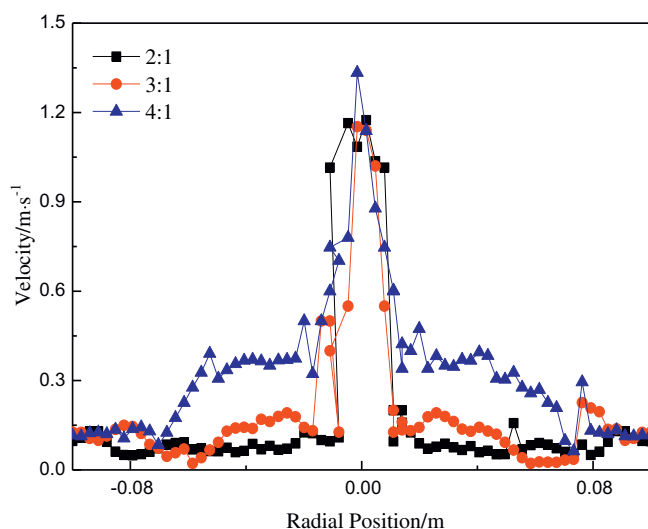


Fig. 13. Effect of  $H/D$  ratio on the overall liquid velocity.

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