Gas-liquid flow regimes and effective interfacial area in a solid foam block stirred tank

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Abstract: Recently, a solid foam block (SFB) stirred tank has attracted wide attention owing to its excellent mass transfer efficiency. However, more fundamental information is needed to deepen the understanding of the strengthening mechanism and application of this novel stirrer. Therefore, in this work, the flow regimes and the effective interfacial area as the crucial information for the mass transfer process in the SFB stirred tank are investigated by high-speed imaging and the Danckwerts-plot technique. Four typical flow regimes are revealed in the SFB stirred tank, which corresponds to the three changes in the effective interfacial area when adjusting the operating conditions. Correlations are respectively developed with the deviation of ±10% and ±15% to distinguish the flow regimes and predict the effective interfacial area in SFB stirred tank. Compared with the Rushton stirred tank, the SFB stirred tank is able to generate two times effective interfacial areas at fixed power input, showing superior performance and the promising prospect in some certain multiphase application fields.

Key words: stirred tank; flow regime; effective interfacial area; multiphase process

1. Introduction

Mechanical agitation is the most common method to enhance mixing/ mass transfer in industries [1, 2]. It refers to forcing fluids flow in a vessel by mechanical means, or to the random distribution of two or more separate phases between one another, e.g., by mixing two miscible liquids, dissolving a solid in a liquid, and dispersing a gas in a liquid to form fine bubbles [3, 4]. These processes are commonly used in chemical engineering processes including hydrogenation, polymerization, crystallization, fermentation, wastewater treatment, etc. [5], where the mechanical agitation devices, especially the stirred tanks play an essential role. However, due to the fact that many fast reactions, crystal, material preparation processes are developed over the years, the stirred tank faces a great challenge for realizing the industrializations of these novel reactions [6, 7]. The stirrer is a core component of the stirred tank and has a direct impact on the quality and quantity of products, mixing time, and power consumption during the chemical processes. Optimizing the stirrer will therefore be a straightforward and effective way to enhance the mixing and mass transfer efficiency of the stirred tank.

One of the most recent innovations of the stirrer design is the solid foam-based stirrer [8, 9]. Solid foam is a highly open-celled material consisting of a reticulated structure of struts. It combines a large specific surface area (160-8500 m^2/m^3) with a high porosity (80-97 %). In some early studies, the struts were applied as static mixers for the fluid streams, which split and recombined passing the struts, and exhibited a low pressure drop and high mass transfer efficiency [10, 11]. The Chemical Reactor

Engineering group at Eindhoven University of Technology firstly used the solid foam block (SFB) as stirrers [8, 9]. The strong centrifugal and shear forces generated by the rotation of the porous stirrer can cut and break gas in the tank into fine bubbles, as well as achieve high gas holdup and strong turbulence for interphase contact [12-14]. Furthermore, the solid foam block can be used as a support for catalyst deposition, and thus allows simple re-use of the catalyst and avoids attrition and agglomeration of the catalyst in it [14-16]. In these studies, the SFB stirred tanks have exhibited excellent mixing and mass transfer efficiency compared to the Rushton stirred tank (the RT stirred tank). For example, Yang et al. [17, 18] demonstrated that the SFB stirred tank in tanks exhibited excellent micromixing efficiency, and the micromixing time can reach 10^{-4} s, which is faster than many conventional reactors [19, 20]. Tschentscher et al.'s results [9, 21] showed that the solid foam stirred tank achieved the high gas-liquid mass transfer coefficient of 0.19 s^{-1} in the oxygen-water system and a liquid-solid mass transfer coefficient of 0.6 s^{-1} in copper dissolution system, which is substantially higher compared to the RT stirred tank and the slurry reactor. Therefore, the SFB stirred tank has been considered promising improvements to conventional stirred tanks and slurry reactors.

However, due to the special structure and novelty of the stirrer, the sufficient fundamental information such as hydrodynamics and mass transfer in this stirred tank should be revealed, and then its operating and application ranges can be presumed. Previous studies employed the γ-ray tomography and camera to investigate the multiphase fluid flow in the SFB stirred tanks [12, 13]. Results showed that the fluid zone in the tank was separated into three sections with different flow characteristics: the center section, where gas bubbles with holdup higher than 50% are trapped and broken up; the foam block section, where phases flow outward by strong centrifugal force while gas bubbles are further cracked into more bubbly shape; and the outer reactor section between the solid foam block and reactor wall, where the phases are transported and affected by baffles flow back to the center section, further enhancing mixing efficiency. Although the visual observation was discussed, the gas-liquid flow regimes, which is fundamental information of the reactor to discriminate the mass transfer capacity and then set the operating parameters in industrial application, were not described in detail.

Moreover, the gas holdup in the tank and the bubble size distribution in the out section were studied as well [12, 13], but the gas-liquid interfacial area as a key indicator to determine interphase mass transfer efficiency were still not exhibited. One of the major reasons is that it is still difficult to obtain the detailed state of bubbles in the solid foam block. Therefore, applying the indirection method such as chemical method to obtain the specific interfacial area will be an effective approach. The Danckwerts-plot technique, proposed by Danckwerts et al. [25], is a recognized chemical method for simultaneous determination of the gas-liquid mass transfer (k_L) and the effective specific interfacial area (a_e) [22-24], and has been widely applied to characterize the mass-transfer performance in many conventional multiphase reactors such as the stirred tank and the packed bed [26-29] and some novel reactors such as the rotating packed bed and the microchannel [30, 31]. This method is based on the pseudo-first-order reaction in which CO_2 is absorbed in the aqueous alkaline solution. The changes in CO_2 concentration between gas inlet and outlet is measured, and then the mass transfer parameters can be obtained by drawing the Danckwerts-plot.

In this work, the flow regime and the effective interfacial area as the crucial fundamental information in the SFB stirred tank are firstly investigated by a high-speed camera and the Danckwerts-plot technique. The effects of the agitation speed, gas flow rate, working liquid volume, and stirrer size are studied, and the mass transfer performance for the SFB stirred tank is compared to the Rushton stirrer as a benchmark. Based on the visual results, several criteria are developed to discriminate the flow regimes in the reactor. The correlations are proposed to predict the effective interfacial area under various operating conditions. The fundamental data provided in this study will promote the applicant of the SFB stirred tank in the relevant practical processes.

2. Experimental Section

2.1 The SFB stirred tank

The ceramic foam material is used for the stirrer material, which is calcined as a donut-shaped block with a height of 5 cm (H_s), an inner diameter of 5 cm (D_i), and three sizes of the outer diameter of 15, 13, and 11 cm (D_o). The SFB stirrer is manufactured by the supplier (Juyifei ceramic Technology Cooperation) with the pore size of 10 pores per linear inch (ppi), the porosity of 0.900, the tortuosity of 1.391, and the interfacial area per unit volume of 598 m^{-1} calculated by previous work [14].The stirrer is mounted in a transparent PMMA tank with the inner diameter (D_R) and height (H_R) of the same size of 20 cm. The distances between the stirrer bottom and the tank bottom (H_D) are set as 1 cm. The inner wall and lid of the tank are equipped with four baffles of 1 cm both in thickness and width as an attempt to improve mass transfer efficiency. The gas inlet and outlet are respectively installed in the center of the bottom and the tank lid.

Meanwhile, a 6-blade Rushton (RT) stirrer made of stainless steel is equipped in the same baffled tank for the comparison study. The RT stirred tank is a standard design [5] with a diameter of 7 cm, a blade size of 1.75×1.4 cm^2, and a distance of 67 mm to the bottom of the tank. It treats the same volume of working fluids, and the comparison is based on the same power input range. The schematic structure of two stirred reactor are shown in **Figure 1**.

2.2 Flow regimes measurements

Figure 2 illustrates the experimental setup to observe the gas-liquid flow in the solid foam block stirred tank. A high-speed camera (PHANTOM v1840) is adopted to capture the bubbles thrown out from the outer section of the SFB stirred tank. Based on the previous observation [14], the hydrodynamics of bubbles are different at various agitation speeds, gas flow rates, and working liquid volumes. Therefore, in this work, the density of thrown bubbles and their location in the block stirrer are observed at different agitation speeds (N = 100 to 500 rpm), the gas flow rates (G = 1 to 5 L/min), and the working liquid height (H_L = 11 to 17 cm), which lead to the discrimination of the flow regimes. Meanwhile, as the technology was mainly used to qualitatively capture the apparent flow of bubbles and liquid in this study, the cylinder shell has little influence on the observational results. Moreover, a dyeing method is adopted to measure the vortex depth. In this method, a white cloth is packed the shaft to measure the distance between the vortex tip and the reactor bottom. Until the rotation of the block stirrer reaches steady operation, the colored liquid with the liquid height from 11 to 17 cm, and the corresponding liquid volume (V_L) from 3 to 5 L is gradually injected into the reactor through a tube with a diameter of 0.6 cm centered on the bottom of the tank. The gas with different flow rates is further introduced into the reactor from the bottom. After reaching the steady state, the gas inlet valve is closed and liquid is discharged. During this process, the cloth is stained, and its dyeing length is measured. Meanwhile, a camera is installed on the upper of the reactor to capture the vortex shape in the tank. Based on the information of bubbles and vortex flow at different operating conditions, the various flow regimes are distinguished.

2.3 Effective interfacial area measurements

2.3.1 Reactions of CO_2 in K_2CO_3/ $KHCO_3$ and Danckwerts-plot method

An absorption system, in this work, occurring during absorption of CO_2 into aqueous carbonate solution is used to measure the effective interfacial area (a_e) in the reactor. The reaction can be described by:

$$CO_2 + H_2O \longleftrightarrow HCO_3^- + H^+ \tag{1}$$

Reaction (1) is catalyzed by NaClO, which allows for a wide variation of the reaction rate by varying the amount of the catalyst [32]. The reaction rate is given by

$$r_1 = \left(k_{H_2O} + k_B C_{cat}\right) C_{CO_2} \tag{2}$$

where k_{H_2O} is the reaction rate constant (0.026 1/s at 25 °C [33]) for the reaction without catalyst, k_B is catalyst rate constant, which can be calculated from the equation [34]:

$$\ln\left(k_B\right)=15.86-\frac{2741}{T}+0.3234I+0.3224\frac{C_{HCO_3^-}}{C_{CO_3^{2-}}} \tag{3}$$

where I is ionic strength calculated according to the definition

$$I=\frac{1}{2}\sum_j c_j z_j \tag{4}$$

Due to the alkaline condition in the buffer solution, CO_2 also reacts with OH^- in the solution, described by:

$$CO_2+OH^- \longrightarrow HCO_3^- \tag{5}$$

Reaction (5) is a second-order reaction and the reaction rate is therefore described by

$$r_5 = k_{OH^-}C_{OH^-}C_{CO_2} \tag{6}$$

The equilibrium concentration of OH^- in the buffer solution can be determined from the expression [33]:

$$C_{OH^-} = \frac{K_W}{K_2}\frac{C_{CO_3^{2-}}}{C_{HCO_3^-}} \tag{7}$$

where K_W is the ionization constant of water, and K_2 is the second dissociation constant of H_2CO_3. In this work, the concentrations of $CO_3^{2-}$and HCO_3^- were the same, and the overall reaction rate of CO_2 can be expressed as

$$r_{CO_2} = \left(k_{H_2O} + k_B C_{cat} + k_{OH^-}\frac{K_W}{K_2}\right)C_{CO_2} \tag{8}$$

The apparent rate constant (k_{app}) of the reaction of CO_2 in K_2CO_3/ $KHCO_3$ buffer solutions with catalyst is shown as:

$$k_{app} = k_{H_2O} + k_B C_{cat} + k_{OH^-}\frac{K_W}{K_2} \tag{9}$$

According to the Danckwerts surface renew model[35], the rate of absorption accompanied by a (pseudo) first-order reaction can be expressed as:

$$R = (C_{Ai} - C_{Ae}) a_{\mathrm{e}} \sqrt{k_L^2 + D k_{app}} \tag{10}$$

where $C_{A\mathrm{i}}$ and C_{Ae} respectively represent the average concentrations and equilibrium concentration of CO_2 at the interface. D is the diffusivity of CO_2 in the liquid. Based on a criterion developed by Danckwerts [33], the concentrations of all ions in this work meet this requirement for the pseudo-first-order assumption. Meanwhile, for relatively fast reaction, the value of $C_{A\mathrm{i}}$ far outweigh the value of C_{Ae}, hence the value of C_{Ae} can be ignored [31]. Finally, the effective interfacial area (a_{e}) can be calculated by rearranging the Eq. 10 to Eq. 11:

$$\left(\frac{R}{C_{Ai}} \right)^2 = a_{\mathrm{e}}^2 D k_{app} + a_{\mathrm{e}}^2 k_L^2 \tag{11}$$

where the absorption rate (R) is determined based on the change of the concentration of CO_2 between gas inlet and outlet. $C_{A\mathrm{i}}$ is the logarithmic average of inlet and outlet CO_2 concentration at the interface. By adjusting the catalyst concentration and measuring the absorption rate, the Danckwerts plot (Eq. 11) is then constructed. In the plot, the slope equals the squared specific gas-liquid interfacial area, and the intercept matches the square of the volumetric mass transfer coefficient.

2.3.2 Experimental procedure

This part of the experimental setup includes a gas premix section, a reactor, and an analysis section, which is also schematically shown in **Figure 2**. In the premix section, the feed gas consisting of CO_2 and N_2 is mixed in a 5 L premix tank, and the initial volume ratio of CO_2 to N_2 is set at 10% (v/v) by controlling CO_2 and N_2 gas flow rates. The 0.5 mol/L K_2CO_3/ 0.5 mol/L $KHCO_3$ buffer solutions with hypochlorite anions as the catalyst are poured into the reactor with the different working liquid height (H_L = 11 and 14 cm), which submerge the stirrer. The hypochlorite anion concentration ranges from 0 - 0.1 mol/L. The agitation speed ranges from 100 to 500 rpm for the SFB stirred tank and 100 to 1300 rpm for the RT stirred tank respectively. Their energy consumptions are calculated based on the applied torque measured by a torque sensor (Beijing Zrn Instrument Technology Co., LTD). After the agitation reaches steady conditions i.e. the value of torque and rotating speed become steady, the feed gas is introduced at the bottom of the reactor. The unabsorbed carbon dioxide and nitrogen leave the reactor at the top, and the concentration of CO_2 in the outlet is analyzed and recorded versus time by a CO_2 concentration analyzer (Jishunan company).

Overall, in this work, a high-speed camera and a dyeing method are designed to measure the flow regime in the SFB stirred tank, and the Danckwerts-plot technique is used to obtain the value of a_e in the tank. The effects of agitation speed (N = 100 to 500 rpm), gas flow rate (G = 1 to 5 L/min), working liquid height (H_L = 11 to 17 cm), and stirrer size (D_0 = 11 to 15 cm) on flow regimes and a_e are studied. All experiments are conducted at a room temperature of 25 ℃. Each experiment is repeated at least three times under the same operating condition, and the most relative error in these repeated experiments is less than 5%.

3. Results and discussion

3.1 Flow regimes

Figure 3 shows the bubbly flow in the SFB stirred tank at various agitation speeds. At low agitation, no bubble is thrown from the outer of the SFB stirred tank, indicating that at this condition bubbles are not drawn into the stirrer leading to the insufficiency breakage. With increasing agitation speed, bubbles start to be drawn into the SFB stirred tank, broken by the stirrer and finally thrown out from the outer of the SFB stirred tank. Meanwhile, the zone of thrown bubbles gradually sinks from the top to the bottom of the SFB stirred tank as the agitation speed increases. With further increasing the agitation speed to a certain value, the whole outer region of the SFB stirred tank appears the thrown bubbles, indicating that at this condition the whole SFB stirred tank is involved in the breaking process of the bubbles. Moreover, **Figure 3** shows that at the agitation speed of 130 rpm, the main size of bubbles ranges from 7.0 to 1.5 mm. improving the agitation speed to 170 rpm, the range of bubble size is 4.0 to 1.0 mm, and when the agitation speed reaches to 250 rpm, the main amount of bubbles were from 2 mm to 0.5 mm. The size of main bubbles decreases obviously with an increase of the agitation speed, hence it proves the SFB stirrer can efficiently break gas to fine bubbles.

In addition, the vortex flow was observed. The effect of the different conditions on the distance between the vortex tip and the reactor bottom is shown in **Figure 4**. The result shows the distance decreases with increasing the agitation speed, and the vortex in the middle of the reactor starts to appear at the low agitation. This Phenomenon was also found in the previous work [8], which proves the SFB stirred tank with a slow rotation still has strong the gas self-inducing capacity [36]. When the agitation speed closes to about 200 rpm, the vortex tip reaches the top of the stirrer. The phenomenon corresponds to the operational condition where the whole outer section of the novel stirrer appears the thrown bubbles. When the agitation speed further improves to about 350 rpm, the volume of the vortex gradually increases, and its tip falls to the bottom of the stirrer. Based on the observation, the vortex affected by the construct of the solid foam block presents a similar annular shape in the hollow part of the stirrer. The gas in this region will be easily drawn into the foam block by centrifugal force and then broken frequently by the rotational porous material, finally generating fine bubbles. **Figure 4** also shows that by increasing working liquid height or decreasing the size of the stirrer, a higher agitation speed is needed for providing more energy to achieve the falling of the vortex tip and introduce gas.

Based on the above discussion, three critical operation lines are proposed to divide the four flow regimes in this work, as shown in **Figure 5**. The red line represents the initial appearance of bubbles at the outer of the stirrer. Therefore, at the regime (I) which is below the red line, no gas is introduced in the stirrer, and then bubbles escape without passing stirrer. Bubbles are gradually drawn into the stirrer, and thrown from the outer of the SFB stirred tank at the regime (II). The blue square represents the whole outer of the stirrer with bubbles appearing, meanwhile at similar conditions, the vortex tip reaches the stirrer top (seen the blue triangle), hence this condition is unified as the blue line in this work. Above this line, it is the regime (III), where a gas void region is gradually generated in the hollow of the stirrer. The black line represents the gas is filled in the hollow of the stirrer, and a complete gas void region is formed. Further improving the operational condition in this work, the gas-liquid flow has no further obvious changes, hence this regime above black line is set as the regime (IV).

The effects of the agitation speed, gas flow rate and liquid height on the transformation of the flow regimes are also shown in **Figure 5**. Results show that the gas flow rate in this work has little effect on the flow regime transformation but increasing the working liquid height or changing the structure such as reducing the size of the stirrer (shown in the **Supplement materials**) should raise the agitation speed to alter the flow regime. Normally, the agitation speed is the effective operation for the transformation, suggesting changing the agitation speed can control the flow regime in the SFB stirred tank to introduce more gas in the liquid.

3.2 Effective interfacial area

3.2.1 Effect of agitation speed

Figure 6 shows the effects of the agitation speed and the working liquid height on the effective interfacial area in the SFB stirred tank. The result shows that the effective interfacial area increases with increasing agitation speed. As the agitation speed increases, more gas was introduced into the middle of the stirrer due to lower pressure by stronger centrifugal force. Meanwhile, based on Darcy's law, porous foam block at higher agitation speed has stronger resistance to the fluid, and then accelerates the velocity of the fluid, and increases the turbulent kinetic energy and dissipation rates in the agitation zone, finally resulting in the higher gas holdup, the smaller size of bubbles and the larger effective interface area. Based on the growth rate of the line of N-a_e, the line can be divided into three parts: (1) At the agitation speed ranging from 50 to 150 rpm, the effective interfacial area is relatively small, and its growth rate is low. Based on the above visualization results, within this range no gas is introduced into the stirrer, hence most of the bubbles do not get the effective breakage resulting in the low value of a_e. (2) When the agitation speed reaches 150 rpm, a_e starts to increase significantly because gas begin to be gradually introduced into the stirrer. Further increasing the agitation speed, more gas as the vortex is introduced into the inside of the stirrer resulting into higher gas holdup in the center section of the stirrer. Previous researches [9, 12, 13] show that the introducing gas flows with liquid through the foam block by the centrifugal forces, resulting into broking to fine bubbles. Meanwhile more zone of the stirrer is involved in breaking bubbles, eventually rapidly increase the gas-liquid interfacial area. Together with visualization data, the results shows that the solid foam material and the structure of the SFB stirred tank are the crucial factors in achieving effective intensification. (3) When the agitation speed further increases to about 350 rpm, the main features of the SFB stirred tank, such as the whole outer of the stirrer with bubbles appearing, and the complete annular vortex in the stirrer, have generated. Therefore, further increasing the agitation speed will not affect the flow regimes, resulting in the increase of a_e at a relatively stable rate. On the whole, the experimental results have similar trends with previous results [8, 9], while the increase of effective interfacial area in the part (2) is more obvious. Moreover, from **Figure 6** a higher effective interfacial area is achieved at 11 cm working liquid height compared to 14 cm. This is because, at the same agitation speed, the driving energy from the rotation of the stirrer is constant, thus a higher working liquid height or more liquid volume means less gas is drawn into the stirrer, meanwhile less energy is dissipated per unit volume. With a smaller gas and lower energy dissipation, the gas holdup and the ability to break bubble declines [37] , causing a smaller effective interfacial area.

3.2.2 Effect of gas flow rate

Figure 7 shows the effect of the gas flow rate on the effective interfacial area in the SFB stirred tank. At the same gas flow rate, higher agitation speed increases the interfacial area, corresponding to the results in **Figure 6**. The increase in gas flow rate will lead to an increase in fluid disturbance in the SFB stirred tank, which is conducive to the breaking of bubbles and the increase of effective interface area. However, the increase in gas flow rate leads to a decrease in the residence time, which means that some bubbles may have been discharged from the SFB stirred tank without breaking, especially at a low agitation speed. With the combination of the mutually exclusive effects, the effective interface area shows a smaller correlation with the gas flow rate at low stirring speeds. However, the effect of gas flow rate on the effective interfacial area is more significant at the higher agitation speed. Due to the higher agitation speed and high gas flow rate, more gas is drawn into the stirrer, and the turbulence in the tank is intense, which can maintain a complete bubble breaking process, finally resulting to an increase in the effective interface area. The results also show that at the agitation speed of 400 and 500 rpm, the SFB stirred tank in this work can effectively process gas at the ranges of gas flow rate from 1L/min to 5L/min, while maintaining the ability to generate a high interfacial area.

3.2.3 Effect of block stirrer size

Figure 8 shows the effect of the stirrer size on the effective interfacial area of gas in the SFB stirred tank. The three different sizes of stirrers shown in the figure have a similar trend, which can be attributed to the similar transition of the flow regimes and mechanism of introducing gas. With increasing the stirrer size, the value of a_e increases at a fixed agitation speed. Meanwhile, the difference of the a_e value between D_o = 15cm & D_o = 13 cm is larger than the difference between D_o = 13cm & D_o = 11 cm. This may be caused by an increase in the centrifugal force and energy dissipation rate as the stirrer size increases. Consequently, more gas would be drawn in the stirrer, meanwhile more energy would be imparted to the dispersion of the gas, resulting in an increase in the effective interfacial area [38]. In addition, the distance between the outer of the larger stirrer and the baffles is smaller, hence the thrown bubbles are more likely to interact with the baffle. These collisions result in the violent turbulent fluctuations of fluid [39], and then the bubbles get further breakage resulting in a larger difference in the interfacial area between the jump from *Do*= 13 cm to *Do*=15 cm and the jump from *Do*=11 to *Do*=13 cm.

3.3 Compared with the Rushton stirrer

Figure 9 shows the performance of the SFB stirred tank compared to the RT stirred tank based on the obtained effective interfacial area and power input. It can be found that at low power input, the value of a_e in the SFB stirred tank is similar to that in the RT stirred tank. However, by further increasing the power input, the SFB stirred tank performs significantly better than the RT stirred tank. When the power input in the ST stirred tank is the maximum in this work, the value of a_e in the SFB stirred tank is almost two times that in the RT stirred tank. This indicates that compared to the RT stirrer which is considered the main stirrer for gas-liquid dispersion, the SFB stirrer has a stronger capacity for breaking bubbles with the same given energy input. The rotation of porous foam packing has a higher dissipated energy rate and bubble collision frequency than the six flaky blades on the disk. Moreover, in the RT stirred tank, the gas flow rate in this work has little effect on the effective interfacial areas proving that the rate reaches the processing limit of this RT stirred tank. However, in the SFB stirred tank, the higher gas flow rate shows a larger interfacial area, indicating that besides the strong breakup capacity, the SFB stirred tank also has fine self-inducing capacity. More gas can be drawn in the SFB stirrer to dispersion, hence it can treat more gas even at high gas throughput. The result shows that the novel stirrer has not only strong breakup capacity, but also fine introducing-gas capacity, so it will have potential to treat the hazardous and/or expensive gas which is desirable to totally absorption. In addition, the value of a_e in the SFB stirred tank is larger than some novel gas-liquid reactors such as the rotating packed bed [30], and impinging steams reactor [40].

3.4 Model development

Based on the above results, the multiple linear regression (MLR) method is used to develop several correlations to discriminate the flow regimes and predict the effective interfacial area, and the calculated results are verified by linear coefficient (R^2) and experimental results. Firstly, for the four fluid regimes, three correlations (Eq.12-14), including the agitation speed, the gas flow rate, the liquid height and the size of the stirrer, respectively represent the transition of the initial gas introducing, the compete bubbles thrown / vortex tip reaching stirrer top, and the vortex tip reaching stirrer bottom situations.

1) The critical correlation of the flow regime from I to II:

$$Fr_{c_1} = 0.062 Fl^{-0.155} \left(\frac{H_R}{H_L} \right)^{-1.622}, \ R^2 = 0.945 \tag{12}$$

2) The critical correlation of the flow regime from II to III:

$$Fr_{c_2} = 0.252 Fl^{-0.024} \left(\frac{H_R}{H_L} \right)^{-1.111}, \ R^2 = 0.967 \tag{13}$$

3) The critical correlation of the flow regime from III to IV:

$$Fr_{c_3} = 1.111 Fl^{0.054} \left(\frac{H_R}{H_L} \right)^{-1.361}, \ R^2 = 0.983 \tag{14}$$

As the effect of the agitation speed on the transition of flow regimes in the SFB stirred tank is obvious, the critical Froude number (Fr_c), representing the ratio between the fluid-inertia force and gravity, is set at the left side of the correlation as the judgment value of different regimes. The effect of the liquid height is more significant compared to the gas phase flow number (*Fl*), because it is relate to the difficulty of gas self-introducing and the formation of gas vortex, which are critical factor for the flow regimes in this tank. The influence of the gas flow rate on the transition is less pronounced, hence the powers of *Fl* in the three correlations are small and the largest one appears in the critical bubbles drawn line, which may due to the influence of the diameter of stirrer. **Figure 10** shows the deviation between the experimental critical Froude number and the predicted results is within ±10% using the above correlations, suggesting a reasonable accuracy.

Furthermore, the model to predict the effective interfacial area at different flow regimes is developed. As the operation condition in regime (I) of no bubbles drawn is disadvantageous to the mass transfer process, the predicted value of a_e in this regime is meaningless. The above experimental results show that in regime (II) and regime (III), the value of a_e has a similar quick increase rate, hence the predicted equation (Eq. 15) is unified as one in these two regimes. And after that, the value of a_e increases at a relatively steady speed, thus another equation (Eq. 16) is developed in regime (IV).

$$\frac{a_e}{a_s}=11.787Fl^{0.159}Fr^{1.671}\left(\frac{H_R}{H_L}\right)^{1.534}\left(\frac{\mathrm{D_R}}{\mathrm{D_0}}\right)^{-3.086}, \quad R^2=0.957 \tag{15}$$

$$\frac{a_e}{a_s}=164.067Fl^{0.293}Fr^{0.752}\left(\frac{H_R}{H_L}\right)^{1.080}\left(\frac{\mathrm{D_R}}{\mathrm{D_0}}\right)^{-3.046}, \quad R^2=0.939 \tag{16}$$

where a_s is the specific surface area of the solid foam block as 598 m^2/m^3. Comparing these power number of these dimensionless factors, the agitation speed, liquid height and size of stirrer have more obvious influences on the value of a_e than the gas flow rate. However, For Eq. 15, the power of the fround number is larger compared with Eq. 16, showing that at the section of Eq.16, the effect of the agitation speed on a_e is slight, while at the section of Eq. 15 the influence become obvious. The result shows that the most of the deviations in **Figure 11** using Eq.15 and 16 are within ±15% representing a reasonable accuracy of the proposed correlations. Moreover, these correlations used to distinguish the flow regimes and predict the effective interfacial area can also be applied for optimization of SFB structure such as the size of stirrer and operation conditions such as the agitation speed, the gas flow rate, the liquid height design at laboratory scale in the future.

4. Conclusion

In this work, the flow regimes and the effective interfacial area as the crucial information for mass transfer between phases in the SFB stirred tank are investigated by a high-speed camera and the Danckwerts-plot technique. The influences of agitation speed, gas flow rate, liquid height, and stirrer size on them are studied. The results show that four flow regimes are distinguished by visual observation: no bubbles drawn in the stirrer, bubbles gradually thrown from the stirrer, columnar vortex gradually generating in the stirrer, and columnar vortex completely generated in the stirrer. In the transition of the flow regimes, the effect of the agitation speed is the dominating factor, and increasing the working liquid height or reducing the size of the stirrer should raise the agitation speed to improve the flow regime.

Meanwhile, the results show that the transition of the flow regimes corresponds to the variation of a_e, hence, with increasing the agitation speed, the changes of a_e can also be divided into several parts: in the first part, the value of a_e increases slowly, then increases rapidly in the second part, and finally increases linearly in the third part. Increasing the gas flow rate and the diameter of the stirrer improve the value of a_e. Moreover, compared with the Rushton stirred tank, the SFB stirred tank shows a superior performance based on the power input and the obtained effective interfacial area.

At last, three critical operational equations with the deviation of ±10% are developed to distinguish the transition of the flow regime, and two correlations with the deviation of ± 15% are developed to predict the value of a_e. This work gives a fundamental understanding of the intensification mass transfer mechanism of the SFB stirred tank.

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Nomenclature

a_e	effective specific interfacial area, m^2/m^3
a_s	specific surface area of the SFB stirred tank, m^2/m^3
C	concentration of reactants, mol/L
D	diffusivity of CO_2 in the liquid, m^2/s
D_i	inner diameter of the SFB stirred tank, cm
D_R	inner diameter of the reactor, cm
D_o	outer diameter of the stirrer, cm
Fl	flow number, $G/[(N/60)D_o^3]$
Fr_c	critical Froude number, $(N/60)^2D_o/g$

G	gas flow rate, L/min
g	gravitation constant, m/s^2
H	distance between the vortex tip and the reactor bottom, cm
H_D	distance between the stirrer bottom and the reactor bottom, cm
H_L	height of working liquid, cm
H_R	height of reactor, cm
H_s	height of the SFB stirred tank, cm
I	ionic strength, mol/L
K	ionization constant
k	reaction rate constant
N	agitation speed, rpm
R	absorption rate, mol/(L·s)
r	reaction rate, mol/(L·s)
T	temperature, K
V_L	working liquid volume, L
z_j	charge of each ionic species
σ	surface tension, N/m

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