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Mass Transfer Enhancement as a Function of Oscillatory Baffled Reactor Design

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Graphical abstract

Highlights:

- Gas-liquid flows were characterised in detail in various OBR designs.
- OBR gas-liquid flow regimes were identified quantitatively and qualitatively.
- Effect of operating conditions on OBR flow patterns determined.
- k_{La} s determined for various OBR designs and operating conditions.

Abstract

Air-water two-phase flow regimes were identified quantitatively and qualitatively for four designs of oscillatory baffled reactor (OBR) over a range of oscillation conditions in semi-batch mode operation (continuous gas phase; batch liquid phase). The baffle designs assessed were helical baffles, smooth periodic constrictions, single orifice plate baffles and multi-orifice plate baffles. Oscillation in a smooth-walled tube was also characterised for comparison purposes. The designs were characterised over a range of oscillatory Reynolds number ($Re_o = 0-8000$) and aeration rates, $vvm = 0-1$. All the reactors had the same geometrical parameters such as diameter, ratio of length to diameter etc. Three distinct flow regimes (bubbly flow, slug flow, and churn flow) were identified, which were similar to those found in conventional bubble columns (BCs), but the bubbly flow regime, which exhibits the highest rates of mass transfer, was observed over a wider range of oscillatory liquid velocities in OBRs. This was due to the flow patterns (usually vortices) and shear engendered by the interactions of the oscillatory flows and the baffle designs, which resulted in coalescence and breakage of the bubbles. The volumetric mass transfer coefficients, k_{La} , were significantly increased in the multi-orifice design, up to 7-fold, compared with that for a steady flow (no oscillatory flow) in a smooth tube (unbaffled column).

Keywords: gas-liquid flow, mass transfer, oscillatory baffled reactor

1. Introduction

Studies on mass transfer enhancement of gas-liquid and liquid-liquid systems have been conducted in many devices including bubble columns, internal and external loop columns, gas-sparged stirred tanks, and pulsed packed column [1, 2, 3]. The bubble column (BC) is a two-phase, gas-liquid, contactor in which a gas flows as bubbles in a liquid

phase. It is commonly used for oxidation, chlorination, and hydrogenation, exploiting design advantages such as good heat and mass transfer, lack of moving parts, ease of operation and low operating and maintenance costs. However, complicated flow structures, the backmixing of the phases, and unpredictable scale up are the key disadvantages of BC [4]. Deckwer (1991) suggested adding internals (baffles) to overcome the backmixing problem in the BC [5]. In addition, pulsation has been applied to various devices, packed columns for example, to enhance mass transfer [e.g. 3]. However, the pulsation incurs greater running costs, at low frequency in particular.

Depending on operating conditions (gas/liquid velocity), four flow patterns can be observed: homogeneous (“bubbly”) flow, heterogeneous (“churn” or “turbulent”) flow, slug flow, and annular flow [4-7]. Numerous studies have been conducted to identify the flow regimes in the BC using different methods or techniques such as visual observation, evolution of global hydrodynamic parameters, Particle Image Velocimetry (PIV), electrical capacitance tomography, Laser Doppler Anemometry (LDA), and γ -ray Computed Tomography (CT), entropy (photon counts) and information entropy [8-12].

The oscillatory baffled column (OBC), a plain tube containing periodic baffles of 1.5 times column diameter, has been shown to significantly enhance k_{LA} [12-16] due to the combined effect of baffles and oscillatory motion, which act to decrease bubble size and increase the gas-liquid contacting area. The k_{LA} in an OBC can be sixfold higher than a BC [1]. The k_{LA} in a stirred tank (ST) was also compared with that in an OBR [17]. It was shown that at the same power density, the k_{LA} was 75% higher in a pulsed baffled bioreactor (PBB) than in an ST fermenter. However the enhancement in the k_{LA} values in the OBR were determined beyond the power density value 1000 W m^{-3} . This may be due to the transition to another flow pattern, but this was not reported. Gas-holdup, ε_G , was also affected by oscillation frequency and amplitude [2, 15] in an air-water system. Increasing frequency and amplitude increased k_{LA} . While mass transfer coefficient is independent of the design of gas sparger due to turbulence, which occurs as a result of the oscillatory motion [14, 16]. Al-Abduly *et al* [16] found that the single-orifice OBR (25 mm i.d.) was more efficient than a baffled column (without oscillation) and an unbaffled column (bubble column) by a factor of 3-5. A more recent study used a series of oscillatory meso-tubes of inside diameter 4.4 mm and lengths of 75 mm and 350 mm to examine the effect of oscillation conditions on the mass transfer enhancement for the gas-liquid system [2]. Two different behaviours of gas hold-up were observed for the effect of f on ε_G due to the different flow patterns were observed such as slug flow which occurred as a result of the OBR’s geometry and the size. Moreover, it was observed that the flow patterns of the gas-water mixture were dominated by the oscillatory flow. The same

behaviour for the gas hold up (ε_G) were observed in an OBC with 50 mm i.d. [18] where the ε_G enhancement appeared beyond $Re_o=4000$. In addition, the calculated k_{La} values obtained from an integral baffled meso-OBR were higher than that obtained in conventional gas-liquid systems (BC), stirred tank (ST), and continuous stirred tank reactor (CSTR) [19, 20]. A multi-orifice baffle column (MOBC) has been used to enhance the dissolution of CO_2 in water [13]. In this study, k_{La} increased 3-fold compared to the unbaffled column.

Due to the significant enhancement mass transfer by the oscillatory baffled flow, OBRs have been just used in industry, NiTech Solutions Ltd for example, to yield different chemical and biochemical productions.

Although identifying flow regime is an important task in the design and scale-up of OBRs, no study has yet mapped the flow regimes for OBRs. OBRs with four designs of baffles such as helical, integral, and single orifice were characterised and proved that the designs could provide high degree of plug flow over a wide range of operating conditions [21, 22]. However, these studies only focused on single phase flow. This study was to investigate the effect of baffle design, oscillation conditions, and gas velocity on the flow patterns and the mass transfer coefficient, k_{La} , for two-phase flow.

2. Experimental apparatus and procedure

2.1. Experimental setup and k_{La} determination

The experimental setup for identifying the regime maps and calculating the mass transfer coefficient for an air-water system in four designs of oscillatory baffled reactors (OBRs) are shown in Fig. 1a.

Fig. 1: Schematic diagram of: a) the experimental apparatus, and b) OBR configurations.

The setup consists a 10mm inner diameter (i.d.) and 450 mm height tube and mounted vertically. An oscillator was connected at the end of the tube to provide a sinusoidal oscillation with a wide range of oscillation frequencies (f) (0.5–15 Hz) and centre-to-peak amplitudes (x_o) (1–15 mm). Various designs of baffles were used in this study including oscillatory helical baffle reactor (OHBR), oscillatory integral baffled reactor, smooth periodic constriction (OIBR), oscillatory single-orifice baffled reactor (OSBR), and oscillatory multi-orifice baffled reactor (OMBR), as well as an oscillatory reactor (OR), a smooth tube. The baffle spacing (l) for all designs was maintained

at 1.5 times column diameter. An isometric view of the OBRs and their geometries are also shown in Fig. 1b, and the dimensional details of the baffles are given in Table 1. A Mettler Toledo polarographic oxygen probe (response time: 35s and error: $\pm 1\%$ at 25 °C) was located at 345 mm from the gas inlet and positioned diagonally to avoid air bubbles sticking to the membrane [11] to continuously measure dissolved oxygen (DO). The DO probe was connected to a transmitter and the data was transferred to a PC via a portable EL-USB-4 Data Logger. The collected data of DO in water were analysed using EasyLog USB Software, where the profile of DO concentration against time, t , is obtained. The OBR columns were operated in semi-batch mode system (continuous gas flow and no liquid throughput) at atmospheric pressure and 20 °C. The liquid (distilled water) height (H) was fixed at 380 mm, with the free liquid surface, for all the experiments.

The gas (air or nitrogen) was injected into the water using a 1 mm i.d. tube (i.e. no sparger used as its effect in OBRs has been shown to be negligible due to the domination of oscillation on the flow [14, 16]).

Prior to each experiment, nitrogen gas (oxygen free, BOC Ltd) was injected into the system to deoxygenate the liquid until the dissolved oxygen, DO, concentration became zero, and then the three-way valve switched from nitrogen to oxygen. As the OBR operates at atmospheric pressure, (open from the top), the air injected had a constant flow rate during the experiments. The mass transfer process, oxygen to water, starts and continues until the dissolved oxygen concentration in the liquid reaches the saturation (no change in DO concentration). The k_La was then calculated by applying the following procedure:

The mass balance for oxygen in the liquid is written as:

$$\frac{dC}{dt} = k_La (C^* - C) \quad (1)$$

...where C^* and C are, respectively, the oxygen solubility and oxygen concentration in the liquid (g l^{-1}). Assuming the liquid phase is homogeneous and C_o is the oxygen concentration at time (t) = 0, the integration of the previous equation leads to:

$$\ln(C^* - C) = \ln(C^* - C_o) - k_La \cdot t \quad (2)$$

k_La can be determined by plotting $\ln(C^*-C)$ against time (t), where the k_La value is the slope of the linear zone, R^2 of the slope line of the $\ln(C-C^*)$ against t plot was ≥ 0.96 . The variation in the k_La measurements was determined from 5 repetitions.

Table 2 shows the experimental operating conditions, oscillation conditions (frequency, f and amplitude x_o) and superficial gas velocity, U_G , which is represented by the aeration rate, vvm . The oscillatory motion in the OBRs is governed by oscillatory Reynolds number (Re_o) and the Strouhal number (St), defined as:

$$Re_o = \frac{2\pi f x_o \rho D}{\mu} \quad (3)$$

$$St = \frac{D}{4\pi x_o} \quad (4)$$

$$\alpha = \left(\frac{D}{d_o}\right)^2 \quad (5)$$

...where, Re_o describes the intensity of mixing applied to the tube, and St characterizes the effective eddy propagation, ρ is the fluid (gas/liquid mixture) density (Kg m^{-3}), μ is the fluid viscosity (Pa s), f is the oscillation frequency (Hz), x_o is the oscillation amplitude (m), α is the cross sectional area (-), D is the OBR diameter (m), and d_o is the orifice diameter (m).

Table 1: Specifications of baffle designs

OBR's designs	Baffle thickness mm	Orifice diameter, d_o (mm)	Open cross-sectional area, α , %
OR	-	-	100
OHBR	2	8	64
OIBR	2	5	25
OSBR	1	5	25
OMBR	2	1.5	56

Table 2: Operating conditions

Condition	Range
f (Hz)	0-10
x_o (mm)	0, 2, 4, 6, 8
vvm	0-1
U_G (mm s^{-1})	0-21
Re_o	0-8000
St	0, 0.1, 0.13, 0.2, 0.4

2.2. Power dissipation

Applying an oscillatory motion to the OBR will add more energy and cost because the oscillation is usually provided by a mechanical motor [1, 2, 12, 13, 14, 23]. However, it plays an important role in improving the flow behaviour with presence of baffles by generating strong eddies and vortices resulting in bubble size reduction, high contacting

area, thereby enhancing mass and heat transfer. The power dissipation (P/V) which is estimated by using the quasi-steady flow model as shown in Eq. (6) [23], should be added to Eq. (7) [1, 23], which represents the total energy consumed by the flowing fluid, kinetic energy, and by the fluid static pressure, potential energy.

$$\left(\frac{P}{V}\right)_o = \frac{2\rho N_b}{3\pi Z C_D^2} \frac{1-\alpha^2}{\alpha^2} x_o^3 \omega^3 \quad (6)$$

$$\left(\frac{P}{V}\right)_B = \rho g U_G \quad (7)$$

...where ρ is the fluid (gas/liquid mixture) density (kg m^{-3}), N_b the number of baffles per unit length (m^{-1}), C_D is the orifice discharge coefficient (taken as 0.7 for the OBRs [1, 14, 15], and 1.0 for the smooth tube), α is the baffle free cross-sectional area defined as $(d_o/D)^2$, x_o is the oscillation amplitude (m), and ω is the angular frequency of oscillation defined as $2\pi f$, g is the gravitational constant (m s^{-2}), Z is the system length, and U_G is the superficial gas velocity (m s^{-1}). Therefore, the overall power consumed in gas-liquid system is given by:

$$\frac{P}{V} = \left(\frac{P}{V}\right)_o + \left(\frac{P}{V}\right)_B \quad (8)$$

2.3. Flow regime identification

The gas-liquid flow patterns inside the OBRs were determined qualitatively by visual observation using a high-speed CCD camera (Photron FastCam) and quantitatively using the flow regime maps. Photron FASTCAM Viewer software (PFV) was applied to control the high-speed camera and save data, also for image processing. The high-speed camera was set on high frame rate, 4000 frames per second (fps), which was sufficient to follow any slight change in the flow pattern precisely. A Perspex-optical box (see Fig. 1a) was also fitted halfway up the OBR and filled with glycerol to remove optical distortion due the OBR's wall curvature. In addition, a powerful light source was applied in 90° with cameral position to gat clear 2D-images (bubble shapes) and videos (see videos in the supplementary materials). The effects of the operating conditions and the baffle geometry designs on the bubble shapes and the flow regimes were also studied. A sequence of image snapshots second were taken to track bubbles over a wide range of aeration rate, $vvm = 0-1$ ($U_G = 0-21 \text{ mm s}^{-1}$), and oscillation conditions, $Re_o = 0-8000$. The collected images and videos (see the supplementary materials) were used to characterise the flow regimes and pinpoint the specific experimental points for each condition on the related flow regime map. Then these experimental points were validated quantitatively using the gas hold-up and drift flux data. The flow pattern on the map was plotted on a graph of maximum oscillatory velocity of liquid (Re_o) vs. superficial gas velocity (vvm). As the system worked in semi-batch mode (continuous gas flow and no liquid throughput), the max velocity of liquid

(Re_o) was used instead of superficial liquid velocity which is used to classify the flow regimes in the conventional bubble column (BC). In order to determine the transition regime conditions, liquid and gas velocity of transition ($(Re_o)_{tr}$ and $(vvm)_{tr}$, from homogeneous (bubbly) flow to heterogeneous (churn), the gas hold up, ε_G , (Eq. (9)) was measured using visual observation with variation in the ε_G measurements based on 3 repetitions. ε_G plotted against vvm and the drift flux velocity, j_{GL} , (Eq. (10)) [24], was plotted against ε_G over a wide range of vvm . The sharp change in the gas hold-up and drift flux slopes represents the transition from homogeneous to heterogeneous flow.

$$\varepsilon_G = \frac{H^* - H}{H} \quad (9)$$

$$j_{GL} = U_G(1 - \varepsilon_G) \pm U_L \varepsilon_G \quad (10)$$

...where ε_G is the gas holdup, H is the liquid height in the absence of gas (m), H^* is the liquid height with the presence of gas (m), j_{GL} is the drift flux velocity (m s^{-1}), and U_L is superficial liquid velocity (m s^{-1}). The positive or negative sign indicates counter-current or co-current flow of liquid relative to the gas phase, respectively.

3. Results and Discussion

3.1. Flow regime maps

Figures 2 (a)-(e), show the images for the gas-liquid flow regimes in different designs of OBRs which exhibited more bubbly flow than that for BCs [4, 22, 27]. In addition, The flow patterns in all oscillatory reactors vary greatly compared to those reported by Taitel *et al* [6] using conventional tubular reactor. This is due to combination effect of the oscillatory flow and baffles led to enhancing the coalescence and break-up processes that govern average bubble size thereby affecting the gas-liquid flow behaviour [2, 18]. Break-up occurs as a result of shear generation at the baffles, the edge of the orifice baffle in particular, which reduces the size of gas bubbles and the liquid drops [13]. Moreover, the residence time of the small bubbles, radial mixing, and gas hold-up were increased by the vortices.

Fig. 2: Optical visualisation of the gas-liquid flow patterns in OBRs.

The observed regimes were divided by transition regimes, bubbly-slug, bubbly-churn, and slug-churn, that can be seen as lines or curves on the plots and their details are summarised in Table 3. It has been concluded from the observation that the transition regimes were strongly dependent on superficial gas velocity, oscillation condition, and baffle design where the oscillatory has the major effect (Fig. 2 (a)-(e)). This agrees with Reis *et al* [2] study which concluded, according to the quantitative results of the gas hold up, that the slug flow could be avoided by increasing the oscillation intensity.

In order to decrease the errors resulting from the identification of the bubbly-churn transition regimes due to the high oscillatory flow, the bubbly-churn transition gas velocities $((vvm)_{tr})$ and oscillatory velocities $((Re_o)_{tr})$ for all

the OBR designs were located at all conditions applied using the gas hold-up curves for determining $(vvm)_{tr}$ and drift flux plots for determining $(Re_o)_{tr}$ as shown in Fig. 3 (a) and (b). The marked points on the curves represent the $(vvm)_{tr}$ values, Fig. 3(a), and $(Re_o)_{tr}$ values, Fig. 3(b). The gas hold up, ε_G , and drift flux, j_{GL} , increased linearly with the gas velocity, vvm , and ε_G respectively below the transition point while above it ε_G and j_{GL} changed slightly. A sudden change in the slopes occurred when the small bubbles combined at $vvm \geq 0.41 \pm 0.033$ and formed very large bubbles (churn) across the column, causing ε_G to flatten off, even at high gas velocities, $vvm = 0.64 - 1.0$ [5, 6, 9]. The data of the sudden changes (bubbly-churn transition points) slopes of the gas hold-up and drift flux plots were matched that collected by the visual observation with agreement of more than 90%.

Fig. 3: Typical plots for the OBRs, a) gas hold-up curves, without oscillation, and b) drift flux plots.

$(vvm)_{tr}$ and $(Re_o)_{tr}$ are the aeration rate and the oscillation condition, respectively, at transition regime, where OR is the oscillatory reactor (Unbaffled reactor), OHBR is the oscillatory helical baffled reactor, OIBR is the oscillatory integral baffled reactor, OSBR is the oscillatory single-orifice baffled reactor, and OMBR is the oscillatory multi-orifice baffled reactor. Average standard deviation from experimental replicas was $\pm 3.2\%$

Table 3: Summary of the transition regimes

OBRs / Transition	bubbly-slug	bubbly-chum	slug-chum
OR	$0 \leq Re_o \leq 3900$ $vvm = 0.43 \pm 0.025$	$3900 \leq (Re_o)_{tr} \leq 8000 \pm 41$ $(vvm)_{tr} = 0.43 \pm 0.031$	$Re_o = 3900$ $0.43 \pm 0.042 \leq vvm \leq 1.0$
OHBR	$0 \leq Re_o \leq 3650$ $vvm = 0.41 \pm 0.023$	$3650 \leq (Re_o)_{tr} \leq 8000$ $(vvm)_{tr} = 0.41 \pm 0.031$	$Re_o = 3650$ $0.41 \pm 0.046 \leq vvm \leq 1.0$
OIBR	$Re_o = 200 e^{4.57 vvm}$ $0 \leq vvm \leq 0.53 \pm 0.028$	$2250 \leq (Re_o)_{tr} \leq 8000 \pm 35$ $(vvm)_{tr} = 0.53 \pm 0.032$	$Re_o = 2250$ $0.53 \pm 0.046 \leq vvm \leq 1.0$
OSBR	$Re_o = 150 e^{3.5 vvm}$ $0 \leq vvm \leq 0.56 \pm 0.026$ and $1050 \leq Re_o \leq 4050$ $vvm = 0.56 \pm 0.026$	$4050 \leq (Re_o)_{tr} \leq 8000 \pm 35$ $(vvm)_{tr} = 0.56 \pm 0.031$	$Re_o = 4050$ $0.56 \pm 0.043 \leq vvm \leq 1.0$
OMBR	$Re_o = 93 e^{5.0 vvm}$ $0 \leq vvm \leq 0.64 \pm 0.025$	$2230 \leq (Re_o)_{tr} \leq 8000 \pm 36$ $(vvm)_{tr} = 0.64 \pm 0.0312$	$Re_o = 2120$ $0.64 \pm 0.043 \leq vvm \leq 1.0$

...where $(vvm)_{tr}$ and $(Re_o)_{tr}$ are the aeration rate and the oscillation condition, respectively, at the transition regime.

In addition, Figures 4(a)-(e), below, illustrate the flow regime maps that are identified based on the observation using the high-speed camera images and videos over wide ranges of Re_o (0-8000) and vvm (0-1) as well as the gas hold up and drift flux velocity plots. Devices in oscillatory flow differ, as observed, substantially from conventional bubble columns in the regimes obtained (BC) [25, 27]. No annular flow regime observed in any designs of OBRs within the tested conditions could be (i) because the gas flow rates were not sufficient and/or (ii) the oscillatory flow prevented a film of liquid around a large bubble.

Fig. 4: Flow pattern maps for the vertical flow of an air/water mixture in 10 mm in diameter OBRs at $x_o=0-8\text{mm}$, $f=0-12\text{Hz}$, and $v_{vm}=0-1.0$ ($U_G=0-21\text{ mm s}^{-1}$): a) oscillatory reactor (OR), b) oscillatory helical baffled reactor (OHBR), c) oscillatory integral baffled reactor (OIBR), d) oscillatory single-orifice baffled reactor (OSBR), and e) oscillatory multi-orifice baffled reactor (OMBR).

Bubbly (homogeneous) flow regime. This regime was observed in all the OBR designs at $v_{vm} \leq 0.64$ and at all values of Re_o . It was identified by small and uniform bubbles dispersed along the column. It has been observed in this regime that bubble size, breakage, and the distance between bubbles were strongly dependent on Re_o , v_{vm} , and the baffle design. According to the error observed from the experimental data as well as the visual observation, this regime was more stable (i.e. less error values by 32%) than the other regimes (slug, and churn flow regime) as the latter were significantly affected by the baffle design. As the bubble size distribution plays a major role in controlling k_{La} and Nu_t [4] in the gas liquid systems, this flow regime, bubbly flow, is often recommended in industry to achieve high rates of mass and heat transfer, even at low gas flow rates. The small bubbles that are characteristic of this regime have high contact areas which increases transport. Therefore, many applications operate

in the bubbly flow regime including Fischer-Tropsch synthesis, water purification, oil transportation, nuclear cooling systems, bioreactors, and oxidation reactions [4, 5, 6, 25, 26].

According to the qualitative, visualisation, and quantitative, hold up, results the order of the area of bubbly flow regime $OMBR > OSBR > OIBR > OHBR > OR$ (Fig. 4 (a)-(e)). At the same operating conditions, the OMBR exhibited a greater area of the bubbly flow pattern, a wider range of operating conditions, Fig. 4(e), than the other OBR designs. This was associated with smaller diameter of the orifice which leads to high shear by interacting with the oscillatory flow thereby increasing the population of the small bubbles as well as forming a large proportion of very small bubbles, $\sim 0.26 \pm 0.01$ mm, (Fig 2(e)-bubbly flow) [2, 12, 14, 15, 27]. The OSBR and OIBR exhibited similar areas of bubbly flow, as shown in Fig. 4(c) and (d). However, the OSBR exhibited a wider range of operating conditions due to the sharp edge of the orifice that acts as a bubble breaker and shear generator. Consequently, very small bubbles were observed in the inter-baffle zone of the OSBR (Fig. 2(d) - bubbly flow). In contrast, for the SPC, large bubbles, $D > 5$ mm, were impeded when passing through the narrow section [2], nevertheless, they returned to the original shape after this section (Fig. 2(c)- bubbly flow), as it does not have sharp edges and the consequent increased shear. The OHBR and OR had similar flow regimes which confirms that the intensity of swirling flow generated by the oscillatory flow and helical baffle [28] was not strong enough to promote coalescence and break-up phenomena, exhibiting little effect on the bubble size comparing with the other baffle designs. Therefore the sharp-edged helical baffle could be more effective than the rounded helical baffle for two-phase flow to induce the break up phenomenon [29].

Slug flow regime. This regime was observed in all the OBRs at low oscillation conditions, $Re_o < 4050$, and high aeration rate, $vvm \geq 0.41$. The slugs typically occupied the inter baffle-zone with a diameter nearly that of the tube (D) and length of 1-2 D . At $Re_o < 4050$, the slug flow was dominated by the gas velocity and the baffle design where the slug bubbles increased with vvm . In addition, the size of the formed slug bubble was depending on the cross-section of the baffles. Although the slug shapes were very similar for all designs, their sizes were affected by the baffle configurations (Fig. 2- slug flow), whereas the largest slug diameter was at the largest open cross-sectional area, $\alpha=100\%$, (OR) (Fig. 2 (a)-slug flow). This implies that the baffle geometry plays an important role in forming slug flow [2]. Consequently, the slug flow occurred earlier in OMBR, OSBR, and OIBR than in OHBR and OR. Slug flow is preferred occasionally as it enhances heat and mass transfer at high liquid velocity, i.e. higher than gas velocity, compared to the other regimes. However, it increases liquid evaporation, leads to damping of fluid oscillations due to the gas compressibility, and increases the pressure drop [2, 30, 31].

Churn (heterogeneous) flow regime. This regime was observed in all the OBRs at high gas flow, $vvm > 0.41$, and oscillatory motion, $Re_o \geq 2250$. It was observed that long, $\sim 3-4D$, large cap bubbles appeared surrounded by a thin film and small bubbles as a result of high bubble coalescence [25, 30, 31]. The mass transfer coefficient in this regime is low compared to that in bubbly flow [32]. However, churn flow is preferred in some industrial fields such as nuclear reactors as it increases gas hold-up. The baffle geometry had a significant impact on the shape of the large bubbles in churn flow. As shown in Fig. 2(a) - churn flow, the churn bubble in the OR has essentially the same shape as that in the BC even at high oscillatory flows (which increase the bubble break up rate). However, in Fig. 2(b) the churn bubble was twisted around the helix in the OHBR due to the swirling flow generated by the helical design. The integral (SPC), single-orifice, and multi-orifice designs impede large bubbles and cause three dimensional oscillations during the narrow sections, before returning to their original shapes in the inter-baffle zone (Fig. 2 (c), (d), and (e)-churn flow). However, OIBR exhibited a smaller churn bubble than the other designs, where the churned bubble cannot occupy the whole area of the wide section of the integral. Consequently, the film around the bubble was thicker, which would enhance mass transfer due to the high contacting area. Microbubbles around the churn bubble were observed in all the designs except for OR due to the interaction between the high oscillatory flow and the baffles.

3.2. Effect of operating condition on k_La

3.2.1. Effect of superficial gas velocity and baffle design

Figures 5 (a), (b), and (c) show the impact of the superficial gas velocity, vvm , and the baffle design on k_La for non-oscillation, $Re_o=0$, (Fig. 5 (a)) and with oscillation, $Re_o=3020$ ($f=8\text{Hz}$ and $x_o=6\text{mm}$) and $Re_o=5030$ ($f=10\text{Hz}$ and $x_o=8\text{mm}$), (Fig. 5 (b) and (c) respectively). Where the dotted lines represent the transition regimes based on the operating conditions, vvm and Re_o .

Fig. 5: Effect of superficial gas velocity on the mass transfer coefficient for different designs of OBRs at a) $Re_o=0$, b) $Re_o=3020$, and c) $Re_o=5030$, average standard deviation from experimental replicas was $\pm 0.0013s^{-1}$.

Similar trends apparent for both cases: k_{La} increases with vvm which is expected based on the definition of the aeration rate, at the same volume, where an increase in vvm leads to increasing the population of bubbles thereby the contacting area increases. According to the k_{La} enhancement the order of the designs is the same, $OMBR > OSBR > OIBR > OHBR > OR$, k_{La} affected in a manner by the flow behaviour as the interfacial area is dependant strongly on the bubble size.

In Fig. 5(a), k_{La} increased insignificantly with vvm due to the limiting interfacial area which usually is very associated with the coalescence and break-up processes where the latter processes are very poor in the absence of oscillatory flow. The maximum enhancement in k_{La} was achieved by OMBR, up to 3.6-fold over a steady flow in a smooth tube (Table 4), while OSBR, OIBR, and OHBR displayed 2.5-fold, 2.9-fold, and 2-fold enhancement in mass transfer respectively.

In Fig. 5 (b) and (c), k_{La} increased with vvm under the effect of oscillatory flow, $Re_o = 3020$ and $Re_o = 5030$. Fig. 5(b) and (c), showed that in the bubbly flow regime, $0.05 \leq vvm \leq 0.64$, k_{La} increased significantly with vvm in all

the OBR designs except OR which shows approximately a constant value due to the poor break-up process, as observed. However, around and beyond the transition regimes, bubbly-slug, bubbly-churn, and slug-churn, (Table 3) the increased k_{LA} has affected clearly in all the OBRs designs. For instant, k_{LA} in OMBR has decreased from $0.03 \pm 0.0014 \text{ s}^{-1}$ to $0.024 \pm 0.0012 \text{ s}^{-1}$ this is because in churn flow and slug flow regimes, the bubbles size is much greater than that in bubbly flow, so there is less surface area and hence less scope for enhancement of the mass transfer (without changing regime). Moreover, it can be observed from Fig. 5(c) that the k_{LA} values in the churn flow decreases significantly with $vvm, \geq 0.6$. This was due to the sudden change in temperature ($>20 \text{ }^{\circ}\text{C}$) as a result of the high friction between the fluid oscillated at $Re_o > 5000$ and the foam formation as observed above the liquid which affected the O_2 dissolution as well as air-water contacting area.

Compared with the other designs, the multi-orifice OBR had the highest enhancement in the k_{LA} values over a steady flow in the smooth tube, bubble column, by approximately 6.7-fold at $vvm = 0.53$ while it, k_{LA} enhancement, is 6-fold, 6.1-fold, and 5-fold for OSBR, OIBR, and OHBR respectively as shown in Table 4. This is associated with the increased interfacial area due to bubble size reduction [7]. The OSBR and OIBR had similar values of k_{LA} at all vvm , superficial gas velocity, applied. This could be because, as observed in the visualisation, in both designs some of the large bubbles were not broken down when travelling through the constrictions in these designs. The k_{LA} was similar for the OHBR and OR. Based on the experimental results and the visual observation, the helical baffle design is not suitable for gas-liquid flow, as the enhancement in k_{LA} over the unbaffled design is small or insignificant. These results behaviour agree very well with other mass transfer studies using integral baffled design, single orifice baffled design, and multi-orifice baffled design in particular, in terms of the effect of the gas and oscillatory flows on k_{LA} [1, 2, 13, 14, 15, 16].

Table 4: The max, min, enhancement values of k_{LA} in Fig. 5 (a) -(c) for each OBR design

	OR		OHBR		OIBR		OSBR		OMBR	
	$Re_o=0$	$Re_o=5030$	$Re_o=0$	$Re_o=5030$	$Re_o=0$	$Re_o=5030$	$Re_o=0$	$Re_o=5030$	$Re_o=0$	$Re_o=5030$
min k_{LA}	0.0046	0.01	0.0046	0.011	0.0044	0.0156	0.0046	0.0158	0.0034	0.0176
max k_{LA}	0.0094	0.05	0.009	0.0226	0.0134	0.028	0.0116	0.0272	0.0164	0.0306

3.2.2. Effect of fluid oscillation on k_{LA}

The effect of baffle configuration and fluid oscillation on k_{LA} is shown in Fig. 6 (a) and (b) for different aeration rates, $vvm = 0.28$ and 0.64 .

Fig. 6: Effect of oscillation condition and baffle configuration on the mass transfer coefficient at a) $vvm = 0.28$, and b) $vvm = 0.64$, average standard deviation from experimental replicas was $\pm 0.0013s^{-1}$.

All the OBRs had the same behaviour: $k_{L,a}$ increased with increasing Re_o in the bubbly flow regime, $vvm \leq 0.64$ (Fig. 6(a)) to achieve mass transfer enhancement of 3.1-fold, 3.1-fold, 3.1-fold, and 2.6-fold by OMBR, OSBR, OIBR, and OHBR respectively. However, beyond the transitions, $vvm \geq 0.41$, the $k_{L,a}$ values fluctuated significantly in all the OBRs designs (Fig. 6(b)). This is because the slug and churn bubbles are larger which might divide the OBR into multiple sections above and below the slug or churn bubbles resulting in different amounts of O_2 dissolution in water in each section.

At $Re_o \leq 500$ ($x_o \leq 2$ mm), k_{LA} enhancement was insignificant, 1.4-fold as max enhancement achieved by OMBR. This was probably due to insufficient eddy formation, which is usual at low amplitudes. A further increase in either Re_o or amplitude (>500 , $x_o > 2$ mm) led to significant increases in k_{LA} due to the transition from slug flow to bubbly flow regime as a result of the oscillation intensity, x_o in particular which controls the eddy length and increases the gas hold up [1, 2, 14, 15]. The behaviour agrees well with a previous study using a single-orifice OBR [14] where k_{LA} increased with the oscillation frequency, however, the enhancement became noticeable when x_o increased to 8mm to achieve ~12.7-fold in k_{LA} enhancement. In addition, the sudden change in the slope of k_{LA} versus oscillation occurs earlier, at lower f , due to the transition with increasing x_o . The behaviour in Fig. 6 also agrees with various studies [2, 12, 13, 15].

It was observed that the OMBR had the highest values of k_{LA} , $0.0084 \pm 0.0005 - 0.03 \pm 0.0008 \text{ s}^{-1}$, at all Re_o due to the huge number of periodic eddies generated, as observed, which lead to holding up the bubbles in the inter-baffle zones (in recirculating vortices), microbubbles formation, and increasing radial mixing [12]. Moreover, the bubble size reduction observed at oscillation conditions $Re_o > 2500$ led to an increase in interfacial area. The OSBR and OIBR designs exhibited similar values of k_{LA} over the Re_o range 0-1500, then k_{LA} increased in the OSBR by a factor of ~ 1.4 at higher Re_o . This correlates well with the observations (Fig. 2 (c) and (d)): both designs exhibit the same flow behaviour at low oscillation conditions ($Re_o \leq 1500$). However, at higher Re_o (>1500), the interaction between the sharp edge of the single orifice baffle and the oscillatory flow enhances bubble breakage when oscillation frequency increases, and a subsequent reduction in the Sauter mean diameter [12, 13].

The OHBR exhibited little improvement in mass transfer because the helical baffle does not break bubbles well, consequently larger bubbles were observed, in the bubbly flow regime, than for the other baffle designs. The OR exhibited the lowest enhancement in mass transfer (Table 4) where the k_{LA} values, 0.0076 s^{-1} and 0.01 s^{-1} at $v_{vm}=0.28$ and 0.64 respectively, at no oscillation, $Re_o=0$ (BC), were lower than that for the other designs in all cases, and increased monotonically with Re_o . This confirms the importance of baffles in a gas-liquid contactor.

According to the data in Fig. 5 and 6, the experimental results of k_{LA} ($0.0046 \text{ s}^{-1} - 0.031 \text{ s}^{-1}$) were not influenced by the dynamics of the oxygen electrode where the mass transfer time (32.3s-217.4s) and the DO probe response time is 30s.

Figure 7 is a typical plot (2D-contour) demonstrating the dependency of the air-water flow pattern and mass transfer coefficient on the oscillatory flow (Re_o) and superficial gas velocity (v_{vm}) in the OBRs. The maximum mass transfer enhancement is in the bubbly flow regime, where the mass transfer coefficient increases with the oscillation conditions and gas velocity. However, beyond $Re_o > 6000$, k_{LA} is affected by the foam formation, which

leads to lower gas liquid contacting area. In addition, slug flow has exhibits fluctuations in the enhancement, while the churn flow exhibits a lower enhancement.

Fig. 7: Typical visualisation for the flow regimes and mass transfer coefficient in the OBRs.

3.3. Power consumption and OBRs' performance

Figure 8 (a) and (b) show the comparison of the calculated power dissipated in this study by all the OBR designs at $vvm = 0.28$ (Fig. 8(a)) and $vvm = 0.64$ (Fig. 8 (b)).

Fig. 8: Effect of power density on the mass transfer coefficient in all the OBRs at a) $vvm = 0.28$ and b) $vvm = 0.64$

Although extra power was added to oscillate the fluid (gas/liquid) inside the OBRs, the oscillatory flow improves the fluid flow behaviour significantly especially with the presence of baffles to achieve the required transfer

characteristics which cannot be achieved in conventional mass transfer devices at the same net flow [1, 17]. This is because the interaction between the oscillatory flow and baffles induces the bubble breakage and coalescence processes, thereby resulting in higher hold up and longer bubble residence time [2, 12, 15, 18] and therefore enhancing mass transfer.

Generally, in the bubbly flow regime, k_{La} increased with the power density or oscillation condition. Some fluctuations in the k_{La} observed from Fig. 7 (a) and (b) can be explained due to the transition regime i.e. between bubbly and slug flow at the P/V range of 20 – 200. k_{La} decreased slightly in the P/V range of 800 – 1200 due to the churn flow regime. The trend of k_{La} versus P/V in Figs. 7 (a) and (b) agrees well with a previous studies using a 20 mm i.d. integral baffle design (OFR-SPC) [15], a 26 mm i.d. single-orifice OBR [1], and 50 and 100 mm i.d. pulsed baffled reactors [12].

Conclusions

Two phase flow regimes for an air-water system were described and mapped for the first time for 5 designs of Oscillatory Baffled Reactor, and an unbaffled column, as functions of oscillation and gas velocity. Most of the OBR designs exhibited significant enhancement in k_{La} over conventional bubble columns. The exception was the helically baffled design, which exhibited no significant improvement and is therefore not recommended for gas-liquid mass transfer applications.

Transitions in k_{La} correlated very well throughout with visually observed transitions in flow regime. The (generally desirable) bubbly flow region was demonstrated to be much wider for the OBRs than for conventional BCs. This indicates that OBRs could be used for applications where conventional BCs could not, particularly at lower flow rates.

Baffle design had a major impact on the transition regimes, by controlling the bubble size and shape. Discrete microbubbles were observed in the OMBR much more often than in the OSBR and OIBR. Consequently, a wider bubbly flow region was observed and higher mass transfer coefficient was obtained for this design.

The highest k_{La} value, 0.0306 s^{-1} , was for the OMBR operating at $Re_o=5030$. The k_{La} for the OMBR was a factor of 7 higher than that, for the unbaffled column (0.0046 s^{-1}) without oscillation (i.e. a BC). Generally, for gas-liquid mass transfer duties the multi-orifice design would be recommended. Also, this design could be applied to study the effect of variation in the liquid phase properties such as densities, viscosity and surface tension. This work demonstrates that baffles and oscillation can significantly enhance gas-liquid mass transfer, and the flow maps and

other data presented here allow important initial design decisions to be made, such as baffle geometry, oscillation conditions and gas flow rate.

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Nomenclature

D	OBR diameter, m	μ	viscosity, Pa s
f	oscillation frequency, Hz	ρ	density, kg/m ³
x_o	oscillation amplitude, m	ε_G	gas hold up, dimensionless
l	baffle spacing/helical pitch, m	α	open cross-sectional area, dimensionless
$k_L a$	volumetric mass transfer coefficient, s ⁻¹	ω	angular frequency ($2\pi f$)
U_G	superficial gas velocity, m s ⁻¹	OBR	oscillatory baffled reactor
U_L	liquid velocity, m s ⁻¹	OR	oscillatory reactor (un-baffled)
j_{GL}	drift flux velocity, m s ⁻¹	OHBR	oscillatory helical baffled reactor
H	liquid height in the absence of gas, m	OIBR	oscillatory integral baffled reactor
H^*	liquid height with the presence of gas, m	OSBR	oscillatory single-orifice baffled reactor
C^*	the saturated dissolved oxygen concentration, g l ⁻¹	OMBR	oscillatory multi-orifice baffled reactor
C	the dissolved oxygen concentration, g l ⁻¹	BC	bubble column
C_o	initial oxygen concentration at time $t = 0$ s, g l ⁻¹	OBC	oscillatory baffled column
t	time, sec	PIV	particle image velocimetry,
N_b	the number of baffles per unit length, m ⁻¹	CT	γ -ray Computed Tomography
P/V	power density, W m ⁻³	LDA	Laser Doppler Anemometry
St	Strouhal number, dimensionless	PBR	pulsed baffled reactor
Re_n	net flow Reynolds number, dimensionless	ST	stirred tank
Re_o	oscillatory flow Reynolds number, dimensionless	STF	stirred tank fermenter
C_D	the orifice discharge coefficient, dimensionless	CSTR	continues stirred tank reactor
g	the gravitational constant, m s ⁻²	DO	dissolved oxygen
10	orifice diameter, m		
vvm	aeration rate, the volumetric flowrate of air per volume of liquid per minute		

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