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CO₂ Dissolution and Design Aspects of a Multi-Orifice Oscillatory Baffled Column

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ABSTRACT

Dissolution of CO₂ in water was studied for a batch vertical multi-orifice baffled column (MOBC) with varying orifice diameters ($d_0$) of 6.4–30 mm and baffle open area ($\alpha$) of 15–42 %. Bubble size distributions (BSDs) and the overall volumetric CO₂ mass transfer coefficient ($K_{La}$) were experimentally evaluated for very low superficial gas velocities, $U_G$ of 0.12–0.81 mm s⁻¹, using 5 % v/v CO₂ in the inlet gas stream at a range of fluid oscillations ($f = 0–10$ Hz and $x_0 = 0–10$ mm). Remarkably, baffles presenting large $d_o = 30$ mm and $\alpha = 36 \%$, therefore in the range typically found for single-orifice oscillatory baffled columns, were outperformed in respect to BSD control and CO₂ dissolution by the other baffle designs or the same aerated column operating without baffles or fluid oscillations. Flow visualisation and bubble tracking experiments also presented in this study established that a small $d_o$ of 10.5 mm combined with a small value of $\alpha = 15 \%$ generates sufficient, strong eddy mixing capable of generating and trapping an extremely large fraction of microbubbles in the MOBC. This resulted in increased interfacial area yielding $K_{La}$ values up to $65 \pm 12$ h⁻¹ in the range of $U_G$ tested, and represented up to 3-fold increase in rate of CO₂ dissolution when compared to the unbaffled, steady column. In addition, a modified oscillatory Reynolds number, $Re_{o'}$ and Strouhal number, $St'$ were presented to assist on the design and scale-up of gas-liquid systems based on multi-orifice oscillatory baffled columns. This work is relevant to gas-liquid or multiphase chemical and biological systems relying on efficient dissolution of gaseous compounds into a liquid phase.
1. INTRODUCTION

The sequestration of carbon dioxide, CO$_2$ is a topic of major industrial interest motivated by the recent increased need for reducing the greenhouse gas emissions. New biotechnological processes are being developed where microalgae, anaerobic bacteria or cyanobacteria use CO$_2$ to produce bulk chemicals and green fuels.$^{1-3}$ The intensification of dissolution of CO$_2$ and other gases requires generating fine bubbles and reducing the mass transfer resistances around the bubbles surface by means of strong mechanical mixing using e.g. a mechanical impeller, which is not always possible in biological processes involving living cells as the external energy input has also to ensure cell integrity.$^{4,5}$

Conventional gas-liquid contacting technology based e.g. on bubble columns (BCs), stirred tank reactors (STRs) and air-lift reactors (ALRs) are somewhat inefficient and present very modest performances$^6$ in respect to the dissolution of gases with large gas aeration rates ($Q_{gas}$) of 1 vvm (volume of gas per volume of liquid per minute) or above; in the particular case of BCs and ALRs this is due to the intensity of mixing being directly linked to the gas flow rates, therefore the contacting times being extremely short.

The overall volumetric mass transfer coefficient ($K_{La}$) for CO$_2$ has been experimentally measured only in a small number of studies.$^7-10$ Calderbank and Lochiel$^7$ investigated $K_{La}$, bubble’s velocity and shape for CO$_2$ freely rising in distilled water, and showed that $K_{La}$ remained constant along the height of the column for bubbles with an equivalent spherical diameter, $d_e$ in the range 4–31 mm. Boogerd and co-authors$^8$ showed that $K_{La}$ for CO$_2$ can be predicted from the known $K_{La}$ values measured for O$_2$, using the following relation: $K_{La,CO2} = 0.893 * K_{La,O2}$ which has been derived from a diffusion coefficient correction factor. Based on that same relationship these authors have predicted a maximum possible $K_{La}$ value for CO$_2$ in the order of 140 h$^{-1}$, based on $K_{La}$ values measured for O$_2$ at a $Q_{gas} = 1$ vvm in a lab scale fermenter operating at pH = 2, which has yet to be demonstrate
experimentally. Hill\textsuperscript{10} determined the dependence of $K_La$ for CO\textsubscript{2} with the temperature, stirring speed and $Q_{gas} = 0.08–0.8$ vvm in a 2.45 L STR using distilled water, and obtained $K_La$ values of 20–120 h\textsuperscript{-1} using a 10 \% v/v CO\textsubscript{2}, so well below the typical $K_La$ values measured for O\textsubscript{2}-water mass transfer in well-mixed vessels. It is however unclear from that study what were the specific conditions that allowed Hill\textsuperscript{10} achieving the highest $K_La$ values reported. Nevertheless, this stresses the difficulty in predicting or comparing performance of different gas contacting systems in respect to CO\textsubscript{2} dissolution.

The oscillatory baffled column (OBC)\textsuperscript{11} is a new mixing technology that has been successfully applied to the intensification of a wide range of chemical and biological processes, including gas-liquid and multiphase systems. The eddy mixing in the periodic baffles or constrictions delivers a good degree of radial mixing and secondary flow that is very effective for controlling the bubble/drop size distribution in the column enhancing the contact between immiscible phases. Few studies have previously used OBCs for O\textsubscript{2} and CO\textsubscript{2} dissolution in water,\textsuperscript{9,12–19} all based on single-orifice OBCs as overviewed in Table 1. Reis \textit{et al.}\textsuperscript{20} reported values of $K_La$ up to 576 h\textsuperscript{-1} for O\textsubscript{2} dissolution in a meso-OBC using a very low value for superficial gas velocity ($U_G$) of 0.37 mm s\textsuperscript{-1} (equivalent to $Q_{gas} = 0.064$ vvm). The superior gas-liquid performance of the meso-OBC resulted mainly from the enhanced gas hold-ups associated with the trapping of microbubbles in the periodic eddies generated in the space between the narrow constrictions, as well as the enhanced shear and velocity fluctuations in the gas-liquid interface. Only in one occasion the dissolution of CO\textsubscript{2} has been experimentally studied in OBCs, but in this instance using pure CO\textsubscript{2} in a continuous 94 mm i.d. column by Taslim and Takriff;\textsuperscript{9} $K_La$ values up to $\sim$100 h\textsuperscript{-1} were reported for $Q_{gas}=1.3–3.6$ vvm. Overall, OBCs are very efficient in respect to gas-liquid mass transport, and the large values of $K_La$ reported were obtained with a 5 to 10-fold reduction in $Q_{gas}$ when compared to the gas aeration rates typically used for BCs, ALRs or STRs. An additional feature perhaps unique to OBCs is its linear
scale-up in some particular applications,\textsuperscript{15,21–23} however no rules have yet been established in respect to scale-up of gas-liquid mixing in OBCs.

In this work, the dissolution of CO\textsubscript{2} on a vertical 150 mm i.d. batch multi-orifice baffled column (MOBC) was experimentally studied and three baffle configurations with different $\alpha$ and orifice diameter ($d_0$) were developed and tested, and the impact of baffle design and $Q_{\text{gas}}$ on $K_{L\alpha}$ quantitatively evaluated. Optical flow visualisation and image analysis was applied for quantifying the impact of oscillatory flow mixing on the Sauter mean diameter ($D_{3,2}$) and BSDs. For the first time the connection between microbubbles trapping and the toroidal vortices in OBCs is quantitatively illustrated. In addition, the main governing dimensionless numbers use for characterising the oscillatory flow mixing intensity were revisited, which should establish the principles for the design of MOBC and scale-up from single-orifice to multi-orifice OBCs.

2. EXPERIMENTAL METHODS AND PROCEDURES

2.1 Multi-Orifice Oscillatory Baffled Column (MOBC)

The 150 mm internal diameter MOBC used in this work is presented in Figure 1. The total volume of the column was 10.6 L, with a working volume ($V_L$) of 9.6 L, and a total column height ($h$) of 540 mm. All experiments have been carried out at atmospheric pressure and room temperature (20 °C).

The gas phase consisted of 5% v/v of CO\textsubscript{2} in air sparged from the bottom of the MOBC. The composition of the gas phase was chosen to prevent changes in the bubbles size due to CO\textsubscript{2} absorption and to minimise the effect of response time of the dissolved CO\textsubscript{2} probe. The sparger consisted of a circular plastic tube perforated with a 0.6 mm diameter needle to deliver an even bubble formation within the column. $Q_{\text{gas}}$ was controlled by a needle valve and measured with a calibrated in-line gas flow meter. The range of $Q_{\text{gas}}$ values herein tested was 0.01–0.1 vvm, corresponding to a range of $U_G$ of 0.12–0.81 mm s\textsuperscript{-1}.
The liquid phase (distilled water) in the MOBC was kept at a constant volume, with the free liquid surface always kept well above the top baffle in order to avoid air entrapment from the headspace. Sinusoidal fluid oscillations were imposed on the fluid using a servo-hydraulic system that controlled a 125 mm o.d. piston attached to the bottom of the column. This moving base piston was capable of delivering fluid oscillation frequency \( f \) and centre-to-peak amplitude \( x_0 \) in the ranges of 0–10 Hz and 0–10 mm, respectively. Due to the nature of design of the servo-hydraulic system a maximum value of \( f = 8 \) Hz could be used with \( x_0 = 3 \) mm.

The batch oscillatory column was equipped with equally spaced multi-orifice baffles with unique designs. Three stainless steel rods (6 mm diameter) were placed inside the column to support the set of baffles. Baffles were designed to fit closely to the column wall. Three different baffles configurations were used in this study (described as designs 1, 2, and 3), with significant differences in \( d_o \) and \( \alpha \) as detailed in Table 2. Design 1 was initially tested as it had been successfully applied to liquid-liquid systems and photochemical oxidation in recent times in the same column (unpublished data). The baffle design with \( d_o = 30 \) mm and \( \alpha = 36 \% \) mimicked that of single-orifice OBCs used in liquid mixing studies.\(^{12,13}\) Baffle designs 2 and 3 were developed using smaller values for \( d_o \) and \( \alpha \) which were observed to be beneficial for enhancing gas-liquid contacting. In all experimental sets, baffles were stacked inside the column at an equal baffle spacing \( L \) of 50 mm (design 1 and 2) or 40 mm (design 3). The asymmetrical configuration of baffle designs 1 and 2, regarding holes distribution in the plate, resulted in selecting a value for \( L \) of 50 mm, which was selected based on other studies in MOBCs\(^6,19,24\). Design 3 aimed at replicating a set of single orifice baffled tubes working at same peak oscillatory liquid flow velocity, where a stack of baffles is fixed and the liquid moved by the action of a piston, following the OBC scale-up rule established by Smith and Mackley.\(^{21}\) Thus, for baffle design 3 the value of \( L \) was adjusted to 40 mm, based on the optimisation studies reported in literature\(^{15,23}\), which suggested \( L \) being in the range of 1.5–1.8 times
the column diameter. This design used a fully symmetrical distribution of holes with a constant distance of 24 mm between any adjacent holes.

2.2 Flow visualisation and BSDs

For optical imaging of gas bubbles and particle tracing experiments in the MOBC, a Perspex-optical box was fitted at mid-height of the MOBC and filled with glycerol as shown in Figure 1. The gap between the external and internal walls of the jacketed glass column was also filled with glycerol in order to reduce optical distortion.25

A fluorescent lamp attached to a light diffusor provided the necessary illumination for tracking of bubble size using a low speed (60 fps) or high-speed (1,000 fps) CCD cameras. For liquid flow visualisation, polyamide particles having mean size of 20 µm were dispersed in the liquid phase and illuminated at 90 degrees to the camera by a mercury vapour lamp to give a bright illuminated field. A high-speed CCD camera (Photron FastCam) with a faster shutter speed was used to continuously acquire 512×512 pixels images. Images were saved to a PC in TIF format at a frequency of 1,000 fps. A sequence of at least 600 image snapshots was taken at different combinations of \( x_0 \) and \( f \), which provided more than 2,000 bubbles for image analysis at each condition. This number of bubbles was concluded to be sufficient for the BSDs to be independent of the number of bubbles analysed (results not shown).

Bubble image analysis was carried out using ImageJ software (NHI Image, USA). A set of 600 images for each experimental condition was converted to 8-bit binary images by applying a threshold. The binary images were then treated through a number of image processing steps in order to obtain a clear edge and area for each individual bubble, which included filling holes, erosion and dilation. Finally, bubbles with minimum size higher than 0.02 mm\(^2\) and circularity in the range of 0.7–1.0 were measured on the entire image sequence. Two important bubble diameters are usually relevant for gas-liquid mass transfer studies: the equivalent spherical bubble diameter (\( d_e \)) and the
Sauter mean diameter ($D_{3,2}$). The size of each individual bubble was quantified from $d_e$ which was calculated from the projected area ($A_{proj}$) according to Eq. 1:

$$d_e = \sqrt{\frac{4 \cdot A_{proj}}{\pi}}$$  

(1)

In this equation it is assumed that all bubbles have spherical shape. This might had resulted in underestimated equivalent bubble size for the larger bubbles, which are less spherical and more likely to be oblate ellipsoids. Nevertheless, for the purpose of comparing baffle performances, the use of $D_{3,2}$ provides a good approximation resulting and reduced error propagation from Eq. 1.

Given the restrictions in the flow visualisation and post-processing of imaged bubbles, the minimum value of $d_e$ that could be resolved was 0.16 mm. As CO$_2$ dissolution involved mass transfer through an interfacial area, $D_{3,2}$ was used and calculated using Eq. 2:

$$D_{3,2} = \frac{\sum d_{e_i}^3}{\sum d_{e_i}^2}$$  

(2)

2.3 Measurement of $K_La$ for CO$_2$ dissolution

The dissolved CO$_2$ concentration in water was continuously monitored for each set of experiments using a dissolved CO$_2$ probe (InPro5000, Mettler Toledo) installed at a fixed position at the centre of the MOBC column, with the tip located at half-column height. Because of the large oscillatory Reynolds numbers used in the study, the estimate mixing times were in the range of few seconds$^{26}$ which is insignificant compared to the response time of the probe (150–180 s) and the long aeration times with 5 % v/v CO$_2$ gas mixture. For that reason, the batch column was assumed to be well mixed.

The dynamic gassing-out method with instantaneous gas interchange, from pure nitrogen, N$_2$ to 5 % CO$_2$ mixture was used to estimate $K_La$ values for CO$_2$ in the batch MOBC. Before each set of experiments the column was filled with fresh distilled water. Nitrogen was then sparged for at least
60 minutes to promote degassing of the liquid and to set the reference 0 % CO₂ saturation whilst starting data acquisition. The gas phase was then switched to 5 % v/v CO₂ mixture and the gas flow rate adjusted using a calibrated rotameter. The percentage saturation of dissolved CO₂ was then monitored until it reached a perfect plateau (i.e. 100 % saturation). The pH electrode of the probe was calibrated in buffer at pH 7.00 and pH 9.21 as recommended by the manufacturer.

A time-lag on dissolved CO₂ probe response was detected which has been associated by other authors with the time required for replacement of the gas in the connection tubing (connecting gas valves in the cylinder to the sparger), in the bubbles, in the liquid phase, and in the headspace. Consequently, a floating coordinate system \((t - t_0)\), set as constant for each gas flow rate used, was defined during data analysis, in which the time delay \((t_0)\) was an arbitrary parameter determined by best-fitting the experimental data with the model using as objective function the minimum square of the difference. The value of \(t_0\) determined for each \(Q_{\text{gas}}\) was within ±10 % of the gas residence time that can be calculated based on the gas flow rate, headspace volume and gas holdups in the column.

In order to compensate for the effect of gas and liquid dynamics in the probe response, only values corresponding to 10–95 % of the saturation dissolved CO₂ concentration \((C_L^*)\) were considered during the best-fitting procedure. According to Oliveira and Ni a first order model and a step change in concentration technique can be used to evaluate probe dynamics. Hence, the constant of the probe \((K_P)\) was determined using a first order model in the column in a step change in CO₂ concentration, which could be determined from a mass balance to CO₂ dissolved in the liquid phase in the batch column:

\[
\frac{C_L^* - C_L(t)}{C_L^* - C_{L,0}} = \exp(-K_P \cdot t)
\]  

(3)

The probe constants, \(K_P\) determined were 18 ± 2 h⁻¹ for the set of experiments using baffle designs 1 and 2, and 23 ± 1 h⁻¹, for the set of experiments shown with baffle design 3. These constants were
different as these sets of experiments have been performed in different instances, and therefore some alteration to the membrane of the probe could have occurred.

Once $K_p$ value was determined, it was then used to determine the volumetric CO$_2$-water mass transfer coefficient, $k_{L'A}$ from the CO$_2$ dissolution plots, assuming a steady-state behaviour for the gas dynamics (i.e. no significant decrease in partial pressure of CO$_2$ pressure in the gas phase) and perfectly mixed liquid phase. A mass balance to the gas phase combined with the first order model for probe dynamics defined in Eq. 3 yields:

$$C_L(t) = C_L^* - \frac{C_{L0}}{K_p - K_{L'A}} \cdot \left( K_p \cdot \exp\left[-K_{L'A} \cdot (t-t_0)\right] - K_{L'A} \cdot \exp\left[-K_p \cdot (t-t_0)\right] \right)$$

Equation (4) was then used to determine the $K_{L'A}$ values for each experiment by best-fitting the experimental CO$_2$ dissolution profiles data to the model using Excel Solver, being the objective function the minimum root-square difference between the two curves in the range of CO$_2$ saturation levels of 10–95 % of $C_L^*$.

### 2.4 Modified oscillatory flow dimensionless numbers

In OBCs the oscillatory motion is complex and traditionally the mixing intensity and mass transfer rates in the inter-baffle regions of small diameter single-orifice OBCs is assumed as governed by two dimensionless numbers, the oscillatory Reynolds number ($Re_o$) and the Strouhal number ($St$):

$$Re_o = \frac{2 \pi f x_0 \rho d_c}{\mu}, \quad (5)$$

$$St = \frac{d_c}{4 \pi x_0}, \quad (6)$$

where $d_c$ is the internal diameter of the column (m), $f$ the fluid oscillation frequency (s$^{-1}$), $\mu$ is kinematic fluid viscosity (kg m$^{-1}$ s$^{-1}$), $\rho$ is the specific mass of the fluid (kg m$^{-3}$) and $x_0$ is the centre-to-peak fluid oscillation amplitude (m).
The $Re_o$ in Eq. (5) was described in analogy to net flow Reynolds number where the product $(2\pi x_0 f)$ represents the peak fluid velocity (m s$^{-1}$) during an oscillation cycle which occurs halfway the piston full stroke. The $St$ and $Re_o$ dimensionless numbers in Eqs. (5) and (6) are routinely used in studies involving single-orifice OBCs where there is a direct link between $d_c$ and the open diameter of the orifice ($d_o$) however they were found unsuitable for scaled-up OBCs and MOBCs for a number of reasons as follows.

A possible strategy for scale-up of OBCs from single-orifice columns is based on increasing $d_c$ by keeping both $Re_o$ and $St$ constant. Following from Eq. (6) this would require $x_0$ to be increased in proportion to $d_c$, therefore $f$ being reduced by 1–2 orders of magnitude in order to keep $Re_o$ constant according to Eq. (5). This happens because currently $Re_o$ on its current form is only based on $d_c$ and not in $d_o$ or the equivalent diameter of the obstacle, ($d_{obs}$) as anticipated from a detailed understanding of the fluid mechanics behind flow separation around obstacles. An alternative and more elegant approach for scale-up of OBCs uses multi-orifice baffles. With that approach, $d_c$ is increased but both $d_o$ and $d_{obs}$ are kept constant. This is equivalent to consider multiple OBCs working effectively in parallel in the same column.

A number of variants to Eq. (5) has been proposed by several authors for multi-orifice baffles, see for example Ni and Gough,29 Smith and Mackley,21 yet the effect of $\alpha$ in the performance of MOBCs has not yet been considered. As this current study used baffles with a range of $d_o$ and $\alpha$ both $Re_o$ and $St$ were modified to accurately represent the state of mixing in the MOBC and support scale-up from single-orifice to multi-orifice OBCs.

Eddy formation in free flow problem around obstacles is controlled by the diameter of the obstacle, the properties of the fluid and the free mean liquid velocity. In that respect, the most important characteristic length in respect to vortices formation is $d_{obs}$, and in analogy it can be described for the MOBC as the “equivalent” diameter of the baffle area that surround each open orifice:
\[ d_{\text{obs}} = d_c \sqrt{\frac{1-\alpha}{n}} \]  \hspace{1cm} (7)

where \( n \) is the number of orifices in the baffle. For multi-orifice baffles \( d_{\text{obs}} \) (not \( d_o \) or \( d_c \) as it happens for single single-orifice OBCs) should be the main geometrical parameter governing flow separation and eddy formation in the column.

From the perspective of mass conservation, the flow of an uncompressible fluid through a multi-orifice baffle differs from free-boundary flow problem for the fact that the fluid has to accelerate when passing through the orifices. Neglecting the effect of the column walls (because of the large \( d_c \) value the pseudo-steady flow is turbulent in the inter-baffle spaces), the mean free stream velocity relevant for vortices formation from the surface of the obstacles is not just controlled by the imposed mean fluid velocity (or peak fluid velocity \( 2\pi x_0 f \) in case of unsteady flow) but also by \( \alpha \). Taking these simple concepts into account, a modified \( Re_o' \) for multi-orifice baffles could be written as follows:

\[ Re_o' = \left( \frac{2\pi x_0 \rho}{\mu} \right) \cdot d_{\text{obs}} \cdot \left( \frac{1}{\alpha} \right) \]  \hspace{1cm} (8)

Combining Eqs. (8) and (7) yields:

\[ Re_o' = \left( \frac{2\pi x_0 \rho}{\mu} \right) \cdot \left( \frac{d_c}{\sqrt{n}} \right) \cdot \sqrt{\frac{1-\alpha}{\alpha^2}} \]  \hspace{1cm} (9)

Mathematically Eq. (9) differs from the equation presented by Smith and Mackley\(^{21}\) for a multi-orifice OBC on the term \([ (1-\alpha)/\alpha^2 ] \) which measures the effect of the open area of the baffle. This yields significant differences in \( Re_o' \) values as can be seen in Table 3. For example, \( Re_o' \) calculated from Eq. (9) for baffle design 2 is about 8-fold lower than value of \( Re_o \) based on Eq. (5) because of the small value of \( d_o \) used.
Similarly, the Strouhal number $St$ in Eq. (6) was modified to represent the actual ratio of diameter of column to fluid amplitude in the region around each individual orifice on the baffles in a MOBC. That required determining the equivalent hydraulic diameter of a single-orifice column, $d_h$:

$$d_h = \frac{d_c}{\sqrt{n}}$$  \hspace{1cm} (10)

Replacing $d_c$ in Eq. (6) by $d_h$ from Eq. (10), a modified Strouhal number ($St'$) was obtained:

$$St' = \frac{d_c}{4\pi \alpha_0} \frac{1}{\sqrt{n}}$$  \hspace{1cm} (11)

3. RESULTS AND DISCUSSION

3.1 The impact of $Q_{gas}$ and fluid oscillations on bubble size in the MOBC and comparison with a bubble column

The $D_{3,2}$ and BSD are recognised to play a major role in controlling $K_La$ in gas-liquid and multiphase systems in single-orifice OBCs and other gas-liquid contacting systems, therefore the first part of this study aimed testing the effect of $Q_{gas}$ and fluid oscillations on the mean bubble size in the MOBC for selected multi-orifice baffle designs. This was done using very low values of $U_G$ of 0.12–0.81 mm s$^{-1}$ which is desirable to attain very high efficiencies of dissolution. Figure 2 shows the optical visualisation of bubbles rising in the MOBC equipped with different sets of multi-orifice baffles in the absence and presence of fluid oscillations. The mean bubble size was found strongly dependent on the baffle design, in particular the small orifice diameter in design 2 ($d_o = 6.4$ mm, $\alpha = 42 \%$) resulted in nearly 50% reduction in bubble size when compared to design 1 ($d_o = 30$ mm and $\alpha = 36 \%$). Nevertheless, no trend could be observed in respect to the effect of intensity of fluid oscillations on the mean bubble size, as increasing $Re_o'$ and $St'$ for a given baffle design returned similar values for $D_{3,2}$ of $\sim 5$ or $\sim 3$ mm for baffle designs 1 and 2, respectively. With baffle designs 1 and 2 it was generally observed that the presence of the baffles per se had a stronger impact on
bubble size than the intensity of the fluid oscillations on its own, as can be concluded by comparing the $D_{3,2}$ for each data set with the steady column baffled MOBC conditions (i.e., $f = 0$ Hz and $x_0 = 0$ mm) in Table 3. Baffle design 3 ($d_o = 10.5$ mm, $\alpha = 15\%$) with the smaller value of $\alpha$ produced an extremely large fraction of microbubbles, which is desirable for enhancement of gas-liquid mass transfer processes. Nevertheless, this presented a barrier for optical visualisation of individual bubbles in the MOBC which is essential for calculating mean bubble sizes and BSDs even at such low values of $U_G$, for that reason it was not possible to systematically collect data about bubble size in Table 3. The three baffle geometries developed in this study aimed at covering the spectrum of orifice diameters and open areas previously used in single-orifice OBCs and its impact on BSD is presented in more detail in Figures 3 and 4 for varying $Q_{\text{gas}}$ in a realistic number of experiments.

The operation of the MOBC with baffle design 1 revealed a bimodal bubble population in the column (Figures 3a–b), with the first population having $d_e < 1$ mm, and the second bubble population an average $d_e$ around 4 mm. This bimodal population is typical in gas-liquid systems and results from the simultaneous bubbles coalescence and breakage phenomena occurring in the column. At the higher $Q_{\text{gas}}$ of 0.1 vvm (Figure 3a) a number of fine bubbles in the range of few hundreds of micrometres could be detected in the column, however there was no significant difference between the MOBC and sparging the baffled column in the absence of fluid oscillations. This is illustrated in Figure 3a for two different combinations of fluid oscillations ($f = 3$ Hz, $x_0 = 1$ mm, $St' = 4.0$; and $f = 3$ Hz, $x_0 = 2.5$ mm, $St' = 1.6$). At a lower $Q_{\text{gas}} = 0.01$ vvm (Figure 3b), the effect of fluid oscillations remained unnoticed. The large $d_o$ value used in baffle design 1 (i.e. 30 mm) was clearly ineffective in promoting radial mixing and bubble breakage in gas-liquid flow, consequently $Q_{\text{gas}}$ was the main effect in respect to control of overall BSDs. This result was to some extent unexpected, as several studies using oscillatory flow mixing have previously shown enhanced bubble breakage for experiments performed with similar $Q_{\text{gas}}$ but different single orifice OBC designs.30,31 This suggested
that a correct length scale of $d_o$ and $d_{obs}$ combined with an even distribution of the orifices across the baffle are essential to promote effective eddy formation and achieve a desirable reduction in bubble sizes.

The BSDs obtained using baffle design 2 with $d_o = 6.4$ mm is shown in Figures 3c and 3d. Again, a bimodal distribution was observed for all experiments in the baffled vertical column in the absence of fluid oscillations at the gas flow rates tested, with a main population of larger bubbles with $d_e$ in the range of 1.5–3 mm, and a second population composed of small bubbles having $d_e < 1$ mm. In the presence of fluid oscillations unimodal BSDs were produced for all values of $Q_{gas}$ tested. In fact, in the presence of fluid oscillations mainly sub-millimetre size bubbles were observed in the MOBC. A detailed optical observation of the CO$_2$ bubbles using high-speed image recording showed that in certain phases of the oscillation cycle the fine bubbles moved in the opposite direction of the liquid flow, revealing strong secondary mixing and consequently bubble being trapped within each inter-baffle cavity for a fraction of the period of oscillation. This is expected to enhance contacting times and its overall impact in respect to $K_La$ is discussed in detail in section 3.3.

In the presence of baffle design 3 (with $d_o$ slightly larger but smaller $\alpha$ than baffle design 2) an unimodal BSDs was observed in the presence of fluid oscillations, with virtually no bubbles larger than 1 mm to be observed in the column (Figure 4). For the range of $Re_o'$ and $St'$ tested it was not possible to accurately determine $D_{3,2}$ because virtually at all combinations of $f$ and $x_0$ tested with this baffle design an extremely large number of microbubbles was generated even at the lowest value of $Q_{gas}$. At the highest values of $Re_o'$ the liquid in the column turned opaque as a result of the extremely high number of microbubbles in the gas-liquid solution, which suggests enhanced gas-liquid contacting.

Figure 5 shows photographic images of bubbles at increasing $Re_o'$ and a constant gas flow rate of $Q_{gas} = 0.01$ vvm when the MOBC was equipped with baffle design 3. A 68 % reduction in $D_{3,2}$ was
observed with fluid oscillations, at $Re_0' = 16170$ and $St' = 1.1$ (Figure 5b) and $Re_0' = 24260$ and $St' = 0.7$ (Figure 5c), compared with the un-baffled steady column. This significant reduction in $D_{3,2}$ at high values of $Re_0'$, resulted in increased interfacial area for mass transfer, which is an effective mean of enhancing mass transfer rates in gas-liquid systems. The combination of a small $d_o$ (as used by Reis et al.\textsuperscript{30}) with high $Re_0$ values (as used by Oliveira and Ni\textsuperscript{31}) was apparently the central point for achieving reduced mean size of bubbles in the MOBC. This can be briefly explained by recalling the physics behind drop generation in constricted flows as follows in section 3.2.

3.2 The effect of open orifice diameter and simple shear on bubble breakage

The breakup of liquid drops or gas bubbles can occur in constricted flows by the action of interfacial forces or inertial forces. Resulting from the very low viscosity of the liquid phase, the maximum capillary number calculated from the peak fluid velocities through the orifices in the three baffle designs tested was $Ca = 0.012$ (calculated for $f = 7$ Hz and $x_0 = 3$ mm), which usually indicates the interfacial forces should dominate the shear stresses. Nevertheless, the high Reynolds numbers of the fluid being forced through the orifices means the dynamics of fluid flow should be actually dominated by inertial effects. As mentioned in section 3.1 the presence of baffles per si was sufficient for reducing the mean size of bubbles, which suggested bubble breakup mechanism is mediated by inertial effects as the liquid and bubbles were pushed through the orifices. On such conditions, the bubble breakup can be connected to the simple shear, $\dot{\gamma}_{SS}$ through an orifice with diameter $d_o$, which can be estimated from:

$$\dot{\gamma}_{SS} = \frac{V_{mean}}{d_o}$$ \hspace{1cm} (12a)

where $V_{mean}$ is the peak fluid velocity through the orifice during the fluid oscillation, which can directly calculated from the input $f$ and $x_0$:

$$V_{mean} = \frac{4f x_0}{\pi d_o^2}$$
\[ V_{\text{mean}} = 2\pi f x_0 \cdot \frac{1}{\alpha} \]  

(12b)

Combining Eqs. (12a) and (12b) yields:

\[ \dot{\gamma}_{SS} = \frac{a}{\alpha d_o} \]  

(13)

where \( a = 2\pi f x_0 \) and depends only on the fluid oscillation conditions selected. Equation (13) returned \( \dot{\gamma}_{SS} = 93*a \), \( \dot{\gamma}_{SS} = 372*a \) and \( \dot{\gamma}_{SS} = 634*a \) for baffle designs 1, 2 and 3, respectively. Comparatively, this represents a 4-fold increase in simple shear by replacing the baffle design 1 with baffle design 2 (with smaller orifice size) and a 6.9-fold increase in \( \dot{\gamma}_{SS} \) by replacing baffle design 1 with baffle design 3 which highlights the relevance of \( \alpha \) and \( d_o \) on BSD. This also showed that \( D_{3,2} \) is inversely proportional to the \( \dot{\gamma}_{SS} \) agreeing with the traditional models for energy dissipation. Similar conclusions were also reported in other studies available in literature.\(^{6,19}\)

### 3.3 Flow visualisation of liquid and spatial tracking of bubbles in the MOBC

A further set of experiments used a high-speed camera for tracking the liquid flow and CO\(_2\) bubbles in the MOBC equipped with baffle designs 2 or 3; design 1 was discarded as it underperformed in respect to BSD control as mentioned in section 3.1. Firstly, the liquid phase was traced with polyamide particles, and an image sequence recorded at 1000 fps. Photographic sequences taken in the MOBC equipped with baffled design 2 in three different positions of the oscillation cycle using \( f = 4 \text{ Hz} \) and \( x_0 = 5 \text{ mm} \), can be found in Supporting Information (Figure S1). The area viewed corresponded to an entire inter-baffle cavity (the position of the two baffles can be seen in the top and bottom of the figures). Although a range of values of \( Re_o' \) and \( St' \) was tested, baffle design 2 showed little evidence of strong eddy formation. The very large ratio \( L/d_h = 4.8 \) and the large number of orifices used in that particular baffle design presumably means the eddies were unable to reach the centre of cavity and the energy dissipation was limited to the edges of the
orifices. The particle tracing experiments showed poor secondary eddy mixing through the oscillation cycle as fluid appeared to move only in straight lines in the direction of the piston stroke (Figure S1, in Supporting Information). Although this baffle configuration delivered smaller bubbles sizes than design 1 it was also found inappropriate for the intensification of gas-liquid flows for presenting limited gas-liquid contacting ($K_LA$ values presented in section 3.4 were in the basis of this final conclusion).

Optical flow visualisation in the MOBC equipped with baffle design 3 showed a very distinct liquid flow patterns. A photographic sequence of the liquid flow patterns in the inter-baffle region (a pair of baffled can be seen on the top and bottom of the figures) with increasing $Re_o$ but approximately constant $St'$ can be found in Supporting Information (Figure S2). Strong eddies were observed at different phases of the oscillation cycle and the intensity and size of eddies increased with increasing $Re_o$ as expected. At the highest value of $Re_o$ tested ($Re_o = 24260, f = 8$ Hz, $x_0 = 3$ mm), the flow patterns revealed a mix of chaotic flow with well-defined toroidal vortices resulting in strong radial movement of the fluid, which is desirable for enhancing gas-liquid contacting and ultimately extend the contacting times in the column.

A second set of optical observations consisted in real-time tracking of bubbles in the MOBC. This was carried out only for baffle designs 2 and 3, and aimed establishing a qualitative link between gas-phase movement and the mass transfer performance. Figures 6 and 7 show on the left hand side a tracking of $(x, y)$ position for a set of 4 bubbles randomly selected that could be observed rising through one inter-baffle space, and on the right hand side the instantaneous axial (vertical) velocity for each bubble corresponding to $V_y = \Delta y/\Delta t$ (mm s$^{-1}$). As a reference, the instantaneous mean fluid velocity imposed by the piston given by $V_y = 2 \pi f x_0 \sin(2\pi ft)$ was also shown on the plots in Figures 6d, 7b and 7d. The arrows in Figures 8a, 8c, 9a and 9c represented the direction and starting position of bubbles at the beginning of the tracking process. Using baffle design 2 and in the absence of fluid
oscillations (Figure 6a–b), bubbles ascended the column with a mean instantaneous velocity of 300–350 mm s\(^{-1}\) which agrees well with the value for terminal velocity of bubbles estimated from Stokes law in a bubble column.\(^{32}\) In the presence of fluid oscillations (Figure 6c–d) there was some noticeable lateral displacement of the bubbles in the column which was an indicator of secondary or non-axisymmetric flow being generated in the column. Analysis to \(V_y\) during an entire oscillation cycle (Figure 6d) has revealed two important facts. First, the rising velocity of bubbles varied throughout the oscillation cycle, just like the liquid velocity did, independently of the size of bubble selected. Secondly, \(V_y\) corresponded approximately to the net difference between the rising velocity in free flow (i.e. with no fluid oscillations – Figure 6b) and the instantaneous liquid flow velocity through the oscillation cycle. The \(V_y\) values were always positive, showing bubbles were delayed when the oscillating piston was moving downwards but accelerated as the piston moved upwards. This resulted in a net increasing in the residence time of the bubbles, therefore increased contacting times in the column.

In respect to baffle design 3 the bubble tracking revealed something substantially different. Two combinations of frequency and amplitude for the same \(Re_0' = 20220\) were presented in the Figure 7 (\(f = 2\) Hz, \(x_0 = 10\) mm, \(St' = 0.2; f = 10\) Hz, \(x_0 = 2\) mm, \(St' = 1.1\)). The bubble tracking showed reduced vertical and increased lateral (radial) bubbles displacement in the inter-baffle regions. This was associated with the strong radial mixing produced in the column by the formation of strong periodic eddies that are capable of trapping bubbles and overtake the natural buoyancy. Figure 7b and 7d showed bubbles effectively following the liquid flow in respect to space and time. At higher frequency, \(f = 10\) Hz bubbles could be seen trapped in the inter-baffle regions for at least two full oscillation cycles (Figure 7d). This was due to the small open area of the baffles, which allowed effective generation of strong eddies throughout the oscillation cycle. In addition to a major
reduction in $D_{3.2}$ reported in section 3.1 the contacting time for mass transfer of CO$_2$ from gas phase to the liquid phase in the column was also increased, which suggests larger mass transfer rates.

### 3.4 Effect of fluid oscillations on $K_{La}$

Table 3 summarises $K_{La}$ values obtained with the three different baffle configurations. The initial CO$_2$ dissolution trials using baffle designs 1 and 2 showed a marginal increase on $K_{La}$ when fluid oscillation conditions were used when compared to the steady column. This was associated with the large mean bubble sizes (design 1) and poor eddy mixing (design 2) observed in the MOBC. For that reason, only CO$_2$ dissolution using baffle design 3 is discussed in detail in this section. Before any comparison is made with $K_{La}$ values available in literature, it is important to highlight that the present study aimed high CO$_2$ dissolution efficiencies, which involved using very low superficial gas velocities; therefore, the obtained $K_{La}$ values are somewhat smaller than the maximum $K_{La}$ values reported by some authors for CO$_2$ and other gases.\textsuperscript{6,19,33} Nevertheless, when comparing the $K_{La}$ values reported in the other few studies that match the same range of mean gas velocities used in our study ($U_G = 0.12–0.81$ mm s$^{-1}$), some improvements can be observed. For example, in Hewgill \textit{et al.}\textsuperscript{13} study, using a O$_2$-water system, a range $k_{La}$ values of 7–13 h$^{-1}$ can be estimated from the $K_{La}$ vs $U_G$ correlation reported (for $U_G = 0.42–0.81$ mm s$^{-1}$), which is 2 to 4 times lower than what we have herein reported (14–57 h$^{-1}$).

Figure 8a summarises the impact of baffles and fluid oscillations on CO$_2$ dissolution profile in the MOBC using baffle design 3. The required sparging time for 90 % CO$_2$ saturation in the un-baffled column was observed as 14.5 min, and reduced to 12.8 min in the baffled (i.e. no fluid oscillations) column, whilst the use of “mild” (5 Hz, 2 mm) or “strong” (7 Hz, 3 mm) fluid oscillations reduced it further to 10.0 min and 8.2 min, respectively. This represents up to 43 % savings on CO$_2$-air mixture injected into the column in order to reach same CO$_2$ saturation level. Despite fluid oscillations requires external energy input that represents an additional cost to be considered, this type of mixing
is energetically efficient as shown by power input studies in OBCs; typical power inputs are in the range of 0.5–0.6 kW m$^{-3}$ – see for example Baird et al.$^{12}$

In respect to $K_{La}$ values, baffle design 3 revealed a major improvement in mass transfer rates when compared to the other baffle designs initially explored. The $K_{La}$ increased with increasing of both $Re_{o'}$ and $U_G$ as shown in Figures 8b and 8c. This agrees well with previous gas-liquid mass transfer studies using single-orifice OBCs$^{13,20,27}$ and multi-perforated reciprocating plate column$^{19}$. A maximum value for $K_{La}$ of 65 ± 12 h$^{-1}$ was obtained at $f = 2$ Hz and $x_0 = 10$ mm, which corresponded to a 3.3 and 2.7-fold increase in $K_{La}$ in comparison with steady ‘baffled’ ($K_{La} = 20$ h$^{-1}$) and ‘un-baffled’ ($K_{La} = 24$ h$^{-1}$) column, respectively. The $K_{La}$ values herein obtained were similar to those achieved by Taslim and Takriff$^{9}$ for a pure CO$_2$-water system however with a 13 to 36-fold reduction in $Q_{gas}$.

It could also be observed in Figure 8b that the use of "gentle" fluid oscillations, at low values of $f$ and $x_0$ (e.g. up to $f = 2$ Hz and $x_0 = 2$ mm in this study) were in general detrimental to CO$_2$-water mass transfer process, as the values of $K_{La}$ obtained at such conditions were slightly lower than the $K_{La}$ values obtained with the steady un-baffled column (dashed horizontal line in Figure 8b). This could be explained by the fact that "gentle" fluid oscillations generate very weak eddy vortices and a net acceleration of the bubbles during the piston stroke upward, as explained for bubble tracking experiments in section 3.3. The axial sinusoidal movement of the fluid leads to a net increase on the rising velocity of bubbles and consequently to reduced residence time of the bubbles in the column followed by a net drop on $K_{La}$. From Figure 8c it can be estimated a minimum value of $Re_{o'} = 3000–4000$ to produce an effective increase in $K_{La}$. It was however not possible to confirm experimentally that the increase in $K_{La}$ in the MOBC resulted from an enhancement in the gas-liquid contacting with increasing $Re_{o'}$ value (i.e. mixing intensity) or from the change in the total interfacial area, as the cloudiness of the CO$_2$-water dispersions at higher $Re_{o'}$ obstructed the direct optical measurement of
individual bubble sizes. Nevertheless, the images sequences as presented in Figure 5 suggested that the increase in $Re_o'$ resulted in no additional decrease in bubble size, but rather only on an increase in the number of bubbles in the inter-baffle regions. This suggested the enhanced liquid mixing and higher velocity fluctuations on the gas-liquid interface reduced the boundary layer on the bubble's surface, as previously shown in similar studies.\textsuperscript{27,31}

In this study, it was found that $K_{La}$ seems to vary linearly with $Q_{gas}$ and $U_G$ (Figure 8c). Other studies carried out in single-orifice OBCs of Oliveira and Ni,\textsuperscript{27} Hewgill \textit{et al.},\textsuperscript{13} and Taslim and Takriff\textsuperscript{8} have shown a power law relationship between $K_{La}$ and $U_G$, of the type obtained for bubble columns that could not be observed with baffle design 3. Al-Abduly \textit{et al.}\textsuperscript{34} and Hewgill \textit{et al.}\textsuperscript{13} obtained a relationship very close to the linearity. Gomaa \textit{et al.}\textsuperscript{33} compiled a set of 8 correlations commonly used or $K_{La}$ estimation of the type $K_{La} \propto U_G^b$, where $b$ has a value in the range of 0.14–1.55. For one of those correlations $b$ is close to unity, as it happens with the MOBC.

The high $K_{La}$ values obtained for dissolution of CO$_2$ in water become relevant when considering the very low gas flow rates used (i.e. $Q_{gas} \leq 0.1$ vvm). For example, Hill\textsuperscript{10} used a stirred tank reactor and $Q_{gas}$ in the range of 0.08–0.80 vvm (i.e. up to 8 times higher aeration rates than the current study) and achieved $K_{La}$ values in the range of 20–120 h\textsuperscript{-1} (despite the conditions at which the highest $K_{La}$ values have been obtained could not be determined from their work). That same study mentioned the best-fitted $K_{La}$ value was obtained at 27.5 °C, 0.45 vvm and 375 rpm and was equal to 41.4 h\textsuperscript{-1}. Taslim and Takriff\textsuperscript{8} performed similar CO$_2$ mass transfer studies in an single orifice OBC and reported similar values for $K_{La}$, although working with very large $Q_{gas}$ in the range of 1.3–3.6 vvm using pure CO$_2$. The high $K_{La}$ values herein reported highlights the successful scale-up and high efficiency of CO$_2$ dissolution upon a proper baffled design in the MOBC. The fine gas-liquid dispersion with enhanced gas-liquid contacting times and improved $K_{La}$ obtained in the MOBC equipped with baffle design 3 is unique in respect to efficiency of CO$_2$ dissolution.
4. CONCLUSIONS

Major improvements in $K_La$ for CO$_2$ dissolution in water were reported for a MOBC working under oscillatory flow mixing and stagnant conditions. The $K_La$ values reported of up to $65 \pm 12$ h$^{-1}$ for very small of superficial gas velocities below 1 mm s$^{-1}$ are in the range of $K_La$ values reported for other gas-liquid contacting systems operating at gas flow rates 10 to 40-fold higher. Baffle design showed a major impact in the performance of the gas-liquid contacting system in respect to $D_{3,2}$, BSD and $K_La$ control. The scale-up of baffle configurations from single-orifice OBCs required even distribution of small diameter orifices and small aperture areas in order to generate a high degree of secondary mixing in the column, therefore the main dimensionless numbers that govern oscillatory flow mixing have been redefined. The shear caused by the oscillatory flow in the highly constricted baffles resulted in the formation of monodispersed microbubbles. For the first time, it was visually shown microbubble trapping by the strong toroidal vortices in the inter-baffle regions. The increased residence times and gas hold-ups caused by the retention of fine bubbles in the column combined with intensive oscillatory gas-liquid contacting were the main parameters responsible for major increase obtained in $K_La$ for CO$_2$. As significant $K_La$ values were obtained with low $U_G$, the MOBC is an advantageous system for large-scale use in gas-liquid reactions and multiphase biotransformations. The results presented in this work are of general relevance to gas-liquid mass transfer in sparged systems and of particular relevance to bioreactor design.
Figure 1. Configuration of the multi-orifice oscillatory baffled column (MOBC) used on CO\textsubscript{2} mass transfer studies. 1 – Dissolved CO\textsubscript{2} probe; 2 – CCD camera; 3 – CPU; 4 – Gas flow controller (rotameter); 5 – Servo-hydraulic unit; 6 – Piston; 7 – Gas sparger; 8 – Display; 9 – Interbaffle cavity; 10 – Optical box (filled with glycerol). Dimensions were: liquid height in column, \( h_L = 450 \) mm; inter-baffle spacing, \( L = \) variable (specific of the baffle design tested – see Table 2 for more details); diameter of piston, \( d_P = 125 \) mm; maximum internal diameter of column, \( d_c = 150 \) mm.
Figure 2. Optical observation of air bubbles rising in an interbaffle cavity in the vertical MOBC. (a) Stagnant fluid; (b) Oscillated fluid. The gas aeration rates, $Q_{gas}$ and fluid oscillation conditions used were: baffle design 1 - $f = 3$ Hz, $x_0 = 2.5$ mm, $Re_o' = 5070$, $St' = 1.6$, and $Q_{gas} = 0.1$ L min$^{-1}$ (0.01 vvm); baffle design 2 - $f = 2$ Hz, $x_0 = 10$ mm, $Re_o' = 2310$, $St' = 0.1$ and $Q_{gas} = 0.4$ L min$^{-1}$ (0.04 vvm); baffle design 3 - $f = 2$ Hz, $x_0 = 10$ mm, $Re_o' = 20220$, $St' = 0.2$ and $Q_{gas} = 0.1$ L min$^{-1}$ (0.01 vvm). Scale bar corresponds to 10 mm.
Figure 3. Bubble size distributions in the MOBC fitted with (a–b) baffle design 1, or (c–d) baffle design 2.
Figure 4. Bubble size distribution in the MOBC fitted with baffle design 3; comparison with unbaffled column.
Figure 5. Impact of fluid oscillation conditions on bubble sizes in the MOBC configured with baffle design 3. (a) $Re' = 0$ (no fluid oscillations); (b) $Re' = 16170$, $St' = 1.1$, $f = 8$ Hz and $x_0 = 2$ mm; (c) $Re' = 24260$, $St' = 0.7$, $f = 8$ Hz and $x_0 = 3$ mm. $Q_{gas}$ was kept constant at 0.1 L min$^{-1}$ (0.01 vvm). The scale bar corresponds to 10 mm (the full image sequences are shown in film files supplied as supplementary data).
Figure 6. Time-tracking of \((x,y)\) position and instantaneous vertical velocity \((V_y)\) for 4 bubbles randomly selected in the inter-baffle region in the MOBC configured with baffle design 2. The aeration rate was kept constant at 0.04 vvm. (a) and (b) stagnant column (i.e. \(Re_o' = 0\)); (c) and (d) fluid oscillated at \(f = 2\) Hz, \(x_o = 10\) mm, \(Re_o' = 2310\), \(St' = 0.1\). Arrows in (a) and (c) show initial position and direction of the bubbles tracked.
Figure 7. Time-tracking of \((x, y)\) position and instantaneous vertical velocity \(\left( V_y \right)\) for 4 bubbles randomly selected in the inter-baffle region in the MOBC configured with baffle design 3. The aeration rate was kept constant at 0.01 vvm. (a) and (b) obtained at \(f = 2 \text{ Hz}, x_0 = 10 \text{ mm}, Re_o' = 20220, St' = 0.2\); (c) and (d) obtained at \(f = 10 \text{ Hz}, x_0 = 2 \text{ mm}, Re_o' = 20220, St' = 1.1\). Arrows in (a) and (c) show initial position and direction of the bubbles tracked.
\( Q_{\text{gas}} = 1.0 \text{ L min}^{-1} \) (0.1 vvm); \( U_G = 0.81 \text{ mm s}^{-1} \)

Un-baffled column (no oscillations)

\( \text{k}_a, \text{a} [\text{h}^{-1}] \)

\( \text{Re} = 16170 \) 
\( (f = 8 \text{ Hz}, x_0 = 2 \text{ mm}) \)

\( \text{Re}'_a = 21230 \) 
\( (7 \text{ Hz}, 3 \text{ mm}) \)

\( \text{Mild Re} = 10110 \) 
\( (5 \text{ Hz}, 2 \text{ mm}) \)

(a)

(b)

(c)
Figure 8. Effect of $Re_o'$ and aeration rate, $U_G$ on the overall volumetric mass transfer coefficient, $K_{La}$ for the un-baffled and baffled multi-orifice column using baffles design 3 (see Table 2 for more details). (a) Example of CO$_2$ dissolution profiles at a constant aeration rate $Q_{gas} = 1.0$ L min$^{-1}$ (0.1 vvm) for different configurations and fluid oscillation conditions in the column; (b) Variation of $K_{La}$ with the modified oscillatory flow Reynolds number ($Re_o'$), at a constant flow rate $Q_{gas} = 1.0$ L min$^{-1}$ (i.e. 0.1 vvm); (c) Variation of $K_{La}$ with mean superficial gas velocity ($U_G$) at a constant $Re_o' = 16170$, $St' = 1.1$, $f$=8 Hz, $x_0$ = 2 mm. Error bars represent two standard deviations from experimental replicas.
### Table 1. Gas-liquid mass transfer studies in oscillatory baffled columns (OBCs)

<table>
<thead>
<tr>
<th>OBC</th>
<th>Gas-liquid system</th>
<th>i.d. [mm]</th>
<th>$Q_{gas}$ [vvm]</th>
<th>$U_G$ [mm s$^{-1}$]</th>
<th>$d_o$ [mm]</th>
<th>$\alpha$ [%]</th>
<th>$K_L a$ [h$^{-1}$]</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Batch single-orifice OBC</td>
<td>Air-fermentation media</td>
<td>50</td>
<td>0.5</td>
<td>3.2</td>
<td>20$^4$</td>
<td>16</td>
<td>$\sim$90–450</td>
<td>Ni et al.$^{22}$</td>
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<tr>
<td>Batch reciprocating plate baffled column</td>
<td>Air-water (self-aerating)</td>
<td>190</td>
<td>n/a</td>
<td>n/a</td>
<td>10–50</td>
<td>7–31</td>
<td>$\sim$0–23</td>
<td>Mackley et al.$^{14}$</td>
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<tr>
<td>Batch reciprocating plate baffled column</td>
<td>Air-water</td>
<td>150</td>
<td>n/d</td>
<td>0.32–1.14</td>
<td>70–90</td>
<td>22–36</td>
<td>n/a</td>
<td>Baird et al.$^{12}$</td>
</tr>
<tr>
<td>Batch reciprocating plate baffled column</td>
<td>Air-water</td>
<td>16.6</td>
<td>n/d</td>
<td>5–15</td>
<td>7.8</td>
<td>46.6</td>
<td>180–2880</td>
<td>Vasic et al.$^{24}$</td>
</tr>
<tr>
<td>Batch reciprocating plate baffled column</td>
<td>Air-water</td>
<td>228</td>
<td>n/d</td>
<td>1.2–11.8</td>
<td>6.4–19.1</td>
<td>31.2–35.7</td>
<td>$\sim$20–720</td>
<td>Gagnon et al.$^{6}$</td>
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<td>Batch single-orifice OBC</td>
<td>Air-water</td>
<td>50</td>
<td>0.05–0.2</td>
<td>1.1–4.3</td>
<td>24</td>
<td>23</td>
<td>$\sim$0–144</td>
<td>Oliveira and Ni$^{27,31}$</td>
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<tr>
<td>Batch single-orifice OBC</td>
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<td>26</td>
<td>n/d</td>
<td>0.4–2.4</td>
<td>15</td>
<td>33</td>
<td>$\sim$0–133</td>
<td>Hewgill et al.$^{13}$</td>
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<td>Ozone-water</td>
<td>25</td>
<td>n/d</td>
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<td>12.5</td>
<td>25</td>
<td>36–252</td>
<td>Al-Abduly et al.$^{34}$</td>
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<tr>
<td>Continuous dual-reciprocating plate baffled column</td>
<td>Air-water</td>
<td>100</td>
<td>n/d</td>
<td>0–1700</td>
<td>1.6–3.2</td>
<td>38</td>
<td>$\sim$72–432</td>
<td>Gomaa et al.$^{33}$</td>
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<tr>
<td>Continuous reciprocating plate baffled column</td>
<td>Air-water</td>
<td>150</td>
<td>n/d</td>
<td>6.3–17.7</td>
<td>6.4–90</td>
<td>23.5–54</td>
<td>$\sim$7–54</td>
<td>Rama Rao and Baird$^{19}$</td>
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<td>Continuous single-orifice OBC</td>
<td>Pure CO$_2$-water</td>
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<td>26–72</td>
<td>50</td>
<td>28</td>
<td>$\sim$8–100</td>
<td>Taslim and</td>
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<tr>
<td>Continuous, single-orifice meso-OBC</td>
<td>Air-water</td>
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<td>0.064</td>
<td>0.37</td>
<td>1.6</td>
<td>14</td>
<td>~0–576</td>
<td>Reis et al.</td>
</tr>
</tbody>
</table>

"Authors reported a baffles width/diameter of 30mm, so it was assumed an open diameter orifice of 20mm in the calculations; (n/d) not disclosed by the authors; (n/a) not applicable/available."
Table 2. Configuration of the 3 internal baffle designs used in the MOBC

<table>
<thead>
<tr>
<th></th>
<th>Baffles design 1</th>
<th>Baffles design 2</th>
<th>Baffles design 3</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Number of baffles in the column</strong></td>
<td>10</td>
<td>9</td>
<td>9</td>
</tr>
<tr>
<td><strong>Average number of orifices per baffle</strong></td>
<td>9</td>
<td>210</td>
<td>31</td>
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<tr>
<td><strong>Orifice diameter $d_o$, mm</strong></td>
<td>30.0</td>
<td>6.4</td>
<td>10.5</td>
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<tr>
<td><strong>Equivalent diameter of obstacle $d_{obs}$, mm (Eq. 7)</strong></td>
<td>40.0</td>
<td>7.9</td>
<td>24.8</td>
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<td><strong>Equivalent hydraulic diameter for single-orifice column $d_h$, mm (Eq. 10)</strong></td>
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<td><strong>Baffle thickness</strong></td>
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<td><strong>Open area $\alpha$, %</strong></td>
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<td><strong>Construction material for baffle</strong></td>
<td>Stainless steel</td>
<td>Polypropylene sandwiched between 2 thin stainless steel layers</td>
<td>Acrylic</td>
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Table 3. Averaged bubble Sauter mean diameter ($D_{3,2}$) and overall CO$_2$ mass transfer coefficient ($K_La$) values obtained in the different baffle designs

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<thead>
<tr>
<th>Baffle design</th>
<th>$Q_{gas}$ [vvm]</th>
<th>$U_G$ [mm s$^{-1}$]</th>
<th>$f$ [Hz]</th>
<th>$x_0$ [mm]</th>
<th>$Re_o$ [-]</th>
<th>$Re_o'$ [-]</th>
<th>$St$ [-]</th>
<th>$St'$ [-]</th>
<th>$D_{3,2}$ [mm]</th>
<th>$K_La$ [h$^{-1}$]</th>
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<td>0.43</td>
<td>0</td>
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<td>0</td>
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<td>*</td>
<td>5.51</td>
<td>9 (± 1)</td>
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</table>

(-) not measured; (*) Strouhal number not applicable for steady flow; (+) insufficient number of individual bubbles available for image analysis.
ASSOCIATED CONTENT

Supporting Information. Flow visualisation films recorded with a high speed camera (relevant for baffle designs 2 and 3), supporting data shown in Figure 5. A text document is also provided with supplementary Figures S1 and S2, containing results from fluid particle tracking. This material is available free of charge via the Internet at http://pubs.acs.org

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NOMENCLATURE

Abbreviations

fps frames per second
rpm rotations per minute
vvm volume of gas per volume of liquid per minute
ss simple shear
Symbols

\( a \)  
mass transfer interfacial area (m\(^2\))

\( A_{\text{proj}} \)  
projected area of the bubble (mm\(^2\))

\( C_{L,0} \)  
initial dissolved concentration (mg L\(^{-1}\))

\( C_L \)  
dissolved CO\(_2\) concentration (mg L\(^{-1}\))

\( C_L^* \)  
concentration of saturation (mg L\(^{-1}\))

\( D_{3.2} \)  
Sauter mean diameter (mm)

\( d_c \)  
internal diameter of the column (mm)

\( d_e \)  
equivalent spherical diameter of bubble (mm)

\( d_h \)  
equivalent hydraulic diameter for single-orifice column (mm)

\( d_o \)  
orifice diameter (mm)

\( d_{obs} \)  
equivalent diameter of the obstacle (mm)

\( d_p \)  
diameter of piston (mm)

\( f \)  
frequency of the oscillation (Hz)

\( h \)  
height of the column (mm)

\( h_L \)  
liquid height in the column (mm)

\( K_{L,a} \)  
overall gas-liquid mass transfer coefficient (h\(^{-1}\))

\( K_P \)  
constant of the probe (h\(^{-1}\))

\( L \)  
spacing between baffles (mm)

\( n \)  
number of orifices in the baffle (dimensionless)

\( Q_{\text{gas}} \)  
gas aeration rate (vvm or L min\(^{-1}\))

\( Re_o \)  
oscillatory Reynolds number (dimensionless)

\( Re_{o'} \)  
modified oscillatory Reynolds number (dimensionless)
$St$  Strouhal number (dimensionless)

$St'$  Modified Strouhal number (dimensionless)

t  aeration time (s)

$t_0$  time delay for the measuring of dissolved CO$_2$ concentration (s)

$U_G$  mean superficial gas velocity (mm s$^{-1}$)

$V_L$  working liquid volume (L)

$V_y$  instantaneous axial (vertical) bubble or liquid velocity (mm s$^{-1}$)

$x_0$  centre-to-peak amplitude of fluid oscillation (mm)

$x_{CO2}$  CO$_2$ molar composition inlet gas (mol/mol)

Greek letters

$\alpha$  fraction of open area of the baffle (dimensionless)

$\Delta t$  time interval (s)

$\Delta y$  vertical displacement (mm)

$\mu$  kinematic fluid viscosity (kg m$^{-1}$ s$^{-1}$)

$\rho$  specific mass of fluid (kg m$^{-3}$)

$\dot{\gamma}$  shear rate (s$^{-1}$)
REFERENCES


