CO₂ Dissolution and Design Aspects of a Multiorifice Oscillatory Baffled Column

Filipa M. Pereira,‡ Diana Z. Sousa,§ M. Madalena Alves,‡ Malcolm R. Mackley,‡ and Nuno M. Reis†,‡,§

†IBB - Institute for Biotechnology and Bioengineering, Centre of Biological Engineering, Universidade do Minho, Campus de Gualtar, 4710-057 Braga, Portugal
‡Department of Chemical Engineering and Biodiversity, University of Cambridge, New Museum Site, Pembroke Street, CB2 3RA Cambridge, U.K.
§Department of Chemical Engineering, Loughborough University, Loughborough LE11 3TU, U.K.

ABSTRACT: Dissolution of CO₂ in water was studied for a batch vertical multiorifice baffled column (MOBC) with varying orifice diameters \(d_o\) 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 \(K\_t\_a\) 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 \((f = 0–10 \text{ Hz and } x_0 = 0–10 \text{ mm})\). Remarkably, baffles presenting large \(d_o\) of 30 mm and \(\alpha = 36\%\), therefore in the range typically found for single-orifice oscillatory baffled columns, were outperformed with respect to BSD control and CO₂ dissolution by the other baffle designs or the same aerated column operating without baffles or fluid oscillations. Flow visualization 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 \(K\_t\_a\) values up to \(65 \pm 12 \text{ h}^{-1}\) in the range of the \(U_g\) tested, representing up to 3-fold increase in the 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 multiorifice 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 medium.

1. INTRODUCTION

The sequestration of carbon dioxide (CO₂) 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 CO₂ to produce bulk chemicals and green fuels.¹⁻³ The intensification of dissolution of CO₂ and other gases requires generating fine bubbles and reducing the mass transfer resistances around the bubbles surface by means of strong mechanical mixing using, for example, a mechanical impeller, which is not always possible in biological processes involving living cells as the external energy input must also ensure cell integrity.⁴⁻⁵

Conventional gas–liquid contacting technology based on, for example, bubble columns (BCs), stirred tank reactors (STRs), and air-lift reactors (ALRs) are somewhat inefficient and present very modest performances⁶ with 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, the contacting times, therefore, being extremely short.

The overall volumetric mass transfer coefficient \(K\_t\_a\) for CO₂ has been experimentally measured only in a small number of studies.⁷⁻¹⁰ Calderbank and Lochiel¹⁰ investigated \(K\_t\_a\) the bubble’s velocity, and shape for CO₂ freely rising in distilled water and showed that \(K\_t\_a\) 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 coauthors⁸ showed that \(K\_t\_a\) for CO₂ can be predicted from the known \(K\_t\_a\) values measured for O₂, using the following relation: \(K\_t\_a\_CO₂ = 0.893 \times K\_t\_a\_O₂\) which has been derived from a diffusion coefficient correction factor. On the basis of that same relationship these authors have predicted a maximum possible \(K\_t\_a\) value for CO₂ in the order of 140 h\(^{-1}\), based on \(K\_t\_a\) values measured for O₂ at a \(Q_{gas} = 1 \text{ vvm}\) in a 2.45 L STR filled with distilled water and obtained \(K\_t\_a\) values of 20–120 h\(^{-1}\) using a 10% \(v/v\) CO₂, well below the typical \(K\_t\_a\) values measured for O₂–water mass transfer in well-mixed vessels. It is however unclear from that study what were the specific conditions that allowed Hill¹⁰ to achieve the highest \(K\_t\_a\) values reported. Nevertheless, this stresses the difficulty in predicting or comparing performance of different gas contacting systems in respect to CO₂ dissolution.

The oscillatory baffled column (OBC)¹¹ is a new mixing technology that has been successfully applied to the
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>Qgas [vvm]</th>
<th>UG [mm s⁻¹]</th>
<th>dₕ [mm]</th>
<th>α [%]</th>
<th>Kᵦ [h⁻¹]</th>
<th>ref</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°</td>
<td>16</td>
<td>~90–450</td>
<td>Ni et al.²²</td>
</tr>
<tr>
<td>batch reciprocating plate baffled column</td>
<td>air–water (self-aerating)</td>
<td>190</td>
<td>n/a</td>
<td>10–50</td>
<td>7–31</td>
<td>~0–23</td>
<td></td>
<td>Mackley et al.¹⁴</td>
</tr>
<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></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>~20–720</td>
<td>Gagnon et al.⁶</td>
</tr>
<tr>
<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>~0–144</td>
<td>Oliveira and Ni²³–²⁴</td>
</tr>
<tr>
<td>continuous dual-reciprocating plate</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>~72–432</td>
<td>Gomaa et al.³³</td>
</tr>
<tr>
<td>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>~7–54</td>
<td>Rama Rao and Baird¹⁹</td>
</tr>
<tr>
<td>continuous single-orifice OBC</td>
<td>pure CO₂–water</td>
<td>94</td>
<td>1.3–3.6</td>
<td>26–72</td>
<td>50</td>
<td>28</td>
<td>~8–100</td>
<td>Tasilim and Takrifi²⁶</td>
</tr>
<tr>
<td>continuous, single-orifice meso-OBC</td>
<td>air–water</td>
<td>4.4</td>
<td>0.064</td>
<td>0.37</td>
<td>1.6</td>
<td>14</td>
<td>~0–576</td>
<td>Reis et al.³⁰</td>
</tr>
</tbody>
</table>

*Authors reported a baffles width/diameter of 30 mm, so it was assumed an open orifice diameter of 20 mm in the calculations; (n/d) not disclosed by the authors; (n/a) not applicable/available.

2. EXPERIMENTAL METHODS AND PROCEDURES

2.1. Multiorifice 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 (Vₑ) 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 CO₂ 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 CO₂ absorption and to minimize the effect of response time of the dissolved CO₂ 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. The gas flow rate was controlled by a needle valve and measured with a calibrated inline gas flow meter. The range of Qgas values herein tested was 0.01–0.1 vvm, corresponding to a range of Uₑ of 0.12–0.81 mm s⁻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 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. The moving base piston was capable of delivering fluid oscillation frequency (f) and center-to-peak amplitude (xₑ) in the ranges of 0–10 Hz and 0–10 mm, respectively. Because of the nature of design of the Sauter mean diameter (Dₜ₂) 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 used for characterizing the oscillatory flow mixing intensity were revisited, which should establish the principles for the design of MOBC and scale-up from single-orifice to multiorifice OBCs.
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 multiorifice 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 baffle 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. 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. 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 is moved by the action of a piston, following the OBC scale-up rule established by Smith and Mackley. Thus, for baffle design 3 the value of $L$ was adjusted to 40 mm on the basis of the optimization studies reported in literature, 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 Visualization and BSDs.

For optical imaging of gas bubbles and particle tracing experiments in the MOBC, a Perspex-optical box was fitted at midheight 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.

A fluorescent lamp attached to a light diffuser provided the necessary illumination for the tracking of bubble size using a low-speed (60 fps) or high-speed (1000 fps) CCD camera. For

**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>Number of baffles in the column</td>
<td>10</td>
<td>9</td>
<td>9</td>
</tr>
<tr>
<td>Average number of orifices per baffle</td>
<td>9</td>
<td>210</td>
<td>31</td>
</tr>
<tr>
<td>Orifice diameter $d_o$, mm</td>
<td>30.0</td>
<td>6.4</td>
<td>10.5</td>
</tr>
<tr>
<td>Equivalent diameter of obstacle $d_{ob}$, mm (Eq. 7)</td>
<td>40.0</td>
<td>7.9</td>
<td>24.8</td>
</tr>
<tr>
<td>Equivalent hydraulic diameter for single-orifice column $d_h$, mm (Eq. 10)</td>
<td>50.0</td>
<td>10.4</td>
<td>26.9</td>
</tr>
<tr>
<td>Baffle spacing $L$, mm</td>
<td>50</td>
<td>50</td>
<td>40</td>
</tr>
<tr>
<td>Baffle thickness</td>
<td>2.0</td>
<td>3.4</td>
<td>3.0</td>
</tr>
<tr>
<td>Open area $\alpha$, %</td>
<td>36</td>
<td>42</td>
<td>15</td>
</tr>
<tr>
<td>Construction material for baffle</td>
<td>Stainless steel</td>
<td>Polypropylene sandwiched between 2 thin stainless steel layers</td>
<td>Acrylic</td>
</tr>
</tbody>
</table>
liquid flow visualization, polyamide particles having a mean size of 20 μm were dispersed in the liquid phase and illuminated at 90 deg to the camera by a mercury vapor lamp to give an 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 1000 fps. A sequence of at least 600 image snapshots was taken at different combinations of x₀ and f, which provided more than 2000 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 analyzed (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 mm² 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 (dₜ) and the Sauter mean diameter (D₃,2). The size of each individual bubble was quantified from dₜ, which was calculated from the projected area (Aₚroj) according to eq 1:

\[ dₜ = \sqrt{\frac{4\cdot Aₚroj}{\pi}} \]  
(1)

In this equation it is assumed that all bubbles have spherical shape. This might have 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 D₃,2 provides a good approximation and reduced error propagation from eq 1.

Given the restrictions in the flow visualization and postprocessing of the imaged bubbles, the minimum value of dₜ that could be resolved was 0.16 mm. As CO₂ dissolution involved mass transfer through an interfacing area, D₃,2 was used and calculated using eq 2:

\[ D₃,2 = \frac{\sum dₜ^3}{\sum dₜ^2} \]  
(2)

2.3. Measurement of Kₐ for CO₂ Dissolution. The dissolved CO₂ concentration in water was continuously monitored for each set of experiments using a dissolved CO₂ probe (InPro5000, Mettler Toledo) installed at a fixed position at the center of the MOBC, with the tip located at half-column height. Because of the large oscillatory Reynolds numbers used in this study, the estimate mixing times were in the range of few seconds²⁶ which is insignificant compared to the response time of the probe (150–180 s) and the long aeration times with 5% v/v CO₂ 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, N₂ to 5% CO₂ mixture was used to estimate Kₐ values for CO₂ 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 min to promote degassing of the liquid and to set the reference 0% CO₂ saturation while starting data acquisition. The gas phase was then switched to a 5% v/v CO₂ mixture, and the gas flow rate was adjusted using a calibrated rotometer. 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 the dissolved CO₂ probe response was detected which was associated by other authors¹⁰ 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 − t₀), set as a constant for each gas flow rate used, was defined during data analysis, in which the time delay (t₀) 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 t₀ determined for each Q_gas was within ±10% of the gas residence time that can be calculated on the basis of the gas flow rate, headspace volume, and gas holdups in the column.

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 (Cₐ*) were considered during the best-fitting procedure. According to Oliveira and Ni²⁷ a first order model and a step change in concentration technique can be used to evaluate the 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:

\[ \frac{Cₐ(t) - Cₐ}{Cₐ - Cₐ₀} = \exp(-K_p(t)) \]  
(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 the value of K_p was determined, it was then used to determine the volumetric CO₂–water mass transfer coefficient, Kₐ from the CO₂ dissolution plots, assuming a steady-state behavior for the gas dynamics (i.e., no significant decrease in partial pressure of CO₂ in the gas phase) and a 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

\[ Cₐ(t) = Cₐ₀ \frac{Cₐ₀ - Cₐ}{K_p - Kₐ} \left[K_p \exp[-K_p(t - t₀)] - Kₐ \exp[-Kₐ(t - t₀)]\right] \]  
(4)

Equation 4 was then used to determine the Kₐ values for each experiment by best-fitting the experimental CO₂ dissolution profiles data to the model using Excel Solver, with the objective function being the minimum root-square difference between the two curves in the range of CO₂ saturation levels of 10–95% of Cₐ*.

2.4. Modified Oscillatory Flow Dimensionless Numbers. In OBCs the oscillatory motion is complex²⁸ and traditionally the mixing intensity and mass transfer rates in the
interbaffle regions of small diameter single-orifice OBCs is assumed as governed by two dimensionless numbers, the oscillatory Reynolds number \( \text{Re}_0 \) and the Strouhal number \( \text{St} \):

\[
\text{Re}_0 = \frac{2\pi f x_0 d_i}{\mu} \quad (5)
\]

\[
\text{St} = \frac{d_i}{4x_0} \quad (6)
\]

where \( d_i \) 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 center-to-peak fluid oscillation amplitude (m).

The \( \text{Re}_0 \) 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 through the piston full stroke. The \( \text{St} \) and \( \text{Re}_0 \) dimensionless numbers in eqs 5 and 6 are routinely used in studies involving single-orifice OBCs in which there is a direct link between \( d_i \) 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 the \( d_i \) by keeping both \( \text{Re}_0 \) and \( \text{St} \) constant. Following from eq 6 this would require \( x_0 \) to be increased in proportion to \( d_i \) and therefore for \( f \) to be reduced by 1–2 orders of magnitude in order to keep \( \text{Re}_0 \) constant according to eq 5. This happens because currently \( \text{Re}_0 \) on its current form is only based on \( d_i \) and not in \( d_o \) or the equivalent diameter of the obstacle \( (d_{\text{obs}}) \), 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 multiorifice baffles. With that approach, $d_o$ is increased but both $d_o$ and $d_{obs}$ are kept constant. This is equivalent to multiple OBCs working effectively in parallel in the same column.

A number of variants to eq 5 was proposed by several authors for multiorifice baffles (see, for example, Ni and Gough,29 Smith and Mackley21), 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 multiorifice OBCs.

Eddy formation in the free flow problem around obstacles is controlled by the diameter of the obstacle, the properties of the fluid, and the free mean liquid velocity. Therefore, the most important characteristic length in respect to vortices formation $OBCs$.

MOBC and support scale-up from single-orifice to multiorifice OBCs.

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 as follows. Baffle design 1: $f = 3$ Hz, $x_o = 2.5$ mm, $Re'_o = 5070$, $St' = 1.6$, and $Q_{gas} = 0.1$ L min$^{-1}$ (0.01 vvm). Baffle design 2: $f = 2$ Hz, $x_o = 10$ mm, $Re'_o = 2310$, $St' = 0.1$, and $Q_{gas} = 0.4$ L min$^{-1}$ (0.04 vvm). Baffle design 3: $f = 2$ Hz, $x_o = 10$ mm, $Re'_o = 20220$, $St' = 0.2$, and $Q_{gas} = 0.1$ L min$^{-1}$ (0.01 vvm). Scale bar corresponds to 10 mm.

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 $D_{0.5}$ and BSD are recognized as playing 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 at testing the effect of $Q_{gas}$ and fluid oscillations on the mean bubble size in the MOBC for selected multiorifice baffle designs. This was done using very low values of $U_{c0}$ of $0.12–0.81$ mm s$^{-1}$ which

\[ Re'_o = \frac{2\pi f x_o \mu}{\rho} d_{obs} \left( \frac{1}{\alpha} \right) \]  

\[ Re'_o = \frac{2\pi f x_o \mu}{\rho} \left( \frac{d_a}{\sqrt{n}} \right) \left( 1 - \frac{\alpha}{\alpha^2} \right) \]  

Mathematically eq 9 differs from the equation presented by Smith and Mackley21 for a multiorifice OBC on the term \( [(1 - \alpha)/(\alpha^3)]^{1/2} \) 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, $d_h$:

\[ d_h = \frac{d_i}{\sqrt{n}} \]  

Replacing $d_i$ in eq 6 by $d_h$ from eq 10, gives a modified Strouhal number ($St'$):

\[ St' = \frac{d_h}{4\pi x_o \sqrt{n}} \]
are desirable to attain very high efficiencies of dissolution. Figure 2 shows the optical visualization of bubbles rising in the MOBC equipped with different sets of multiorifice 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 that of design 1 ($d_o = 30$ mm and $\alpha = 36\%$). Nevertheless, no trend could be observed with 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 ~5 or ~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 visualization 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 collect systematically quantitative data about bubble size for 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 their impact on BSD is presented in more detail in Figures 3 and 4 for varying $Q_{gas}$ values in a realistic number of experiments.

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.
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 having 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 \( Q_{gas} \) of 0.1 vvm (Figure 3a) a number of fine bubbles in the range of few hundreds of micrometers 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 \( (f = 3 \text{ Hz}, x_0 = 1 \text{ mm}, St' = 4.0; \text{ and } f = 3 \text{ Hz}, x_0 = 2.5 \text{ mm}, St' = 1.6) \). At a lower \( Q_{gas} = 0.01 \text{ vvm} \) (Figure 3b), the effect of fluid oscillations remained unnoticed. The large \( d_e \) 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 \( Q_{gas} \) was the main effect with 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 \( Q_{gas} \) but different single-orifice OBC designs.\(^{30,31}\) This suggested that a correct length scale of \( d_e \) 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 \( d_e = 6.4 \) mm is shown in Figures 3c,d. 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 submillimeter size bubbles were observed in the MOBC. A detailed optical observation of the CO\(_2\) bubbles using a 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 bubbles being trapped within each interbaffle cavity for a fraction of the period of oscillation. This is expected to enhance contact time, and its overall impact with respect to \( K_LA \) is discussed in detail in section 3.3.

In the presence of baffle design 3 (with \( d_e \) slightly larger but smaller \( \alpha \) than baffle design 2) unimodal BSDs were 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 \( D_{1.2} \) 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 \( Q_{gas} = 0.01 \text{ vvm} \) when the MOBC was equipped with baffle design 3. A 68% reduction in \( D_{1.2} \) was observed with fluid oscillations, at \( Re'_o = 16170 \) and \( St' = 1.1 \) (Figure 5b) and \( Re'_o = 24260 \) and \( St' = 0.7 \) (Figure 5c), compared with the un baffled steady state. This significant reduction in \( D_{1.2} \) at high values of \( Re'_o \) resulted in an increased interfacial area for mass transfer, which is an effective mean of enhancing mass transfer rates in the gas–liquid systems. The combination of a small \( d_e \) (as used by Reis et al.\(^{30}\)) with high \( Re' \) values (as used by Oliveira and Ni\(^{31}\)) was apparently the central point for achieving a reduced mean size of bubbles in the MOBC. This can be briefly explained by recalling the physics behind droplet 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 \( f = 7 \text{ Hz} \) and \( x_0 = 3 \text{ mm} \)), 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 se was sufficient for reducing the mean size of bubbles, which suggested that the 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

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

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

$$V_{\text{mean}} = \frac{2\pi fx_0}{\alpha}$$  \hspace{1cm} (12b)

Combining eqs 12a and 12b yields

$$\dot{\gamma}_{SS} = \frac{a}{\alpha d_o}$$  \hspace{1cm} (13)

where $a = 2\pi fx_0$ and depends only on the fluid oscillation conditions selected. Equation 13 returned $\dot{\gamma}_{SS} = 93a$, $\dot{\gamma}_{SS} = 372a$ and $\dot{\gamma}_{SS} = 634a$ for baffle designs 1, 2, and 3, respectively. Comparatively, this represents a 4-fold increase in simple shear by replacing 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 the literature.\textsuperscript{6,19}

3.3. Flow Visualization 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 not analyzed as it underperformed with respect to BSD control as mentioned in section 3.1. First, the liquid phase was traced with polyamide particles, and an image sequence was recorded at 1000 fps. Photographic sequences taken in the MOBC equipped with baffle design 2 in three different positions of the oscillation cycle using $f = 4$ Hz and $x_0 = 5$ mm can be found in the Supporting Information (Figure S1). The area viewed corresponded to an entire interbaffle 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 $St'_o$ 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 center of the 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 the 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 ($K_{L}a$ values presented in section 3.4 were part of the basis of this final conclusion).

Optical flow visualization in the MOBC equipped with baffle design 3 showed very distinct liquid flow patterns. A photographic sequence of the liquid flow patterns in the interbaffle region (a pair of baffles 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 to establish 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 the $(x,y)$ position for a set of four bubbles randomly selected that could be observed rising through one interbaffle space, and on the right-hand side the instantaneous $\Delta y/\Delta t$ (mm s$^{-1}$). As a reference, the instantaneous mean fluid velocity imposed by the piston given by $V_y = 2\pi x_0 \sin(2\pi ft)$ is also shown on the plots in Figures 6d, 7b, and 7d. The arrows in Figures 6a,c and 7a,c represent 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 the 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 nonaxisymmetric flow being generated in the column. Analysis of $V_y$ during an entire oscillation cycle (Figure 6d) has revealed two important facts. First, the rising velocity of bubbles varied throughout the oscillation cycle in a similar way to the liquid velocity and independently of the size of bubble selected. Second, $V_y$ 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 that bubbles were delayed when the oscillating piston was moving downward but accelerated as

---

**Figure 7.** Time-tracking of $(x,y)$ position and instantaneous vertical velocity $(V_y)$ for four bubbles randomly selected in the interbaffle region in the MOBC configured with baffle design 3. The aeration rate was kept constant at 0.01 vvm. (a and b) obtained at $f = 2$ Hz, $x_0 = 10$ mm, $Re'_e = 20220$, $St' = 0.2$; (c and d) obtained at $f = 10$ Hz, $x_0 = 2$ mm, $Re'_e = 20220$, $St' = 1.1$. Arrows in panels a and c show initial position and direction of the bubbles tracked.
the piston moved upward. This resulted in a net reduction in the residence time of the bubbles, therefore reducing 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 \( R_e' \) were presented in Figure 7 (\( f = 2 \) Hz, \( x_0 = 10 \) mm, \( S' = 0.2; f = 10 \) Hz, \( x_0 = 2 \) mm, \( S' = 1.1 \)). The bubble tracking showed reduced vertical and increased lateral (radial) bubbles displacement in the interbaffle 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 7bd 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 interbaffle 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 \( D_{12} \) reported in section 3.1 the contact time for mass transfer of CO\(_2\) from the 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 summarizes \( K_La \) 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 \( K_La \) in the presence of fluid oscillations when compared to the un baffled bubble 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 the literature, it is important to highlight that the present study aimed at high CO\(_2\) dissolution efficiencies, which involved using very low \( U_G \) values. At such values of \( U_G \) the mass transfer enhancement is usually challenging because of the very limiting interfacial area in a gas–liquid or multiphase system. Consequently, the \( K_La \) values obtained were somewhat smaller than the maximum \( K_La \) values reported by some authors for CO\(_2\) and other gases\(^{6,10,13}\) at much higher \( U_G \) values. Compared to the very few studies carried out at a similar range of \( U_G \) mean gas velocities used in this study (\( U_G = 0.12–0.81 \) mm s\(^{-1} \)) significant improvements could be observed in respect to \( K_La \) values. For example, in the study of Hewgill et al.\(^{13}\) using a O\(_2\)-water system, a range of \( K_La \) values of 7–13 h\(^{-1} \) can be estimated from the \( K_La \) versus \( U_G \) correlation reported (for \( U_G = 0.42–0.81 \) mm s\(^{-1} \)), which is 2 to 4 times lower than the \( K_La \) values obtained with the MOBC (14–57 h\(^{-1} \)).

Figure 8a summarizes 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 to be 14.5 min, and reduced to 12.8 min in the baffled (i.e., no fluid oscillations) column, while the use of “mild” (5 Hz, 2 mm) or “strong” (7 Hz, 3 mm) fluid oscillations reduced it further to 10.0 and 8.2 min, respectively. This represents up to 43% savings on CO\(_2\)-air mixture injected into the column in order to reach the same CO\(_2\) saturation level. Despite fluid oscillations requiring 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 $K_{a,t}$ increased with an increase of both $Re_c$ and $U_c$ as shown in Figure 8b,c. This agrees well with previous gas–liquid mass transfer studies using single-orifice OBCs\textsuperscript{13,20,21} and a multiperforated reciprocating plate column.\textsuperscript{19} A maximum value for $K_{a,t}$ of $65 \pm 12 \ h^{-1}$ was obtained at $f = 2 \ Hz$ and $x_0 = 10 \ mm$, which corresponded to a 3.3- and 2.7-fold increase in $K_{a,t}$ in comparison with steady “baffled” ($K_{a,t} = 20 \ h^{-1}$) column and “unbaffled” ($K_{a,t} = 24 \ h^{-1}$) column, respectively. The $K_{a,t}$ values herein obtained were similar to those achieved by Taslim and Takriff\textsuperscript{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 the $CO_2$–water mass transfer process, as the values of $K_{a,t}$ obtained at such conditions were slightly lower than the $K_{a,t}$ values obtained with the unbaffled bubble 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 $K_{a,t}$. From Figure 8b a minimum value of $Re_c = 3000$–$4000$ can be estimated to produce an effective increase in $K_{a,t}$. It was however not possible to confirm experimentally that the increase in $K_{a,t}$ in the MOBC resulted from an enhancement in the gas–liquid contacting with increasing $Re_c$ 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 $Re_c$ obstructed the direct optical measurement of individual bubble sizes. Nevertheless, the image sequences as presented in Figure 5 suggested that the increase in $Re_c$ resulted in no additional decrease in bubble size, instead increasing the number of bubbles in the interbaffle regions. This enhanced liquid mixing and yielded higher velocity fluctuations on the gas–liquid interface reducing the boundary layer on the bubble’s surface, as previously shown in similar studies.\textsuperscript{27,31}

In this study, it was found that $K_{a,t}$ seems to vary linearly with $Q_{gas}$ and $U_c$ (Figure 8c). Other studies carried out in single-orifice OBCs of Oliveira and Ni,\textsuperscript{27} Hewgill et al.,\textsuperscript{12} and Taslim and Takriff\textsuperscript{9} have shown a power law relationship between $K_{a,t}$ and $U_c$ of the type obtained for bubble columns that could not be observed with baffle design 3. Al-Abduly et al.,\textsuperscript{34} and Hewgill et al.,\textsuperscript{13} obtained a relationship very close to the linearity. Gomaa et al.,\textsuperscript{33} compiled a set of eight correlations commonly used for $K_{a,t}$ estimation of the type $K_{a,t} \alpha U_c^b$, 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 $K_{a,t}$ 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 in the current study) and achieved $K_{a,t}$ values in the range of 20–120 $h^{-1}$ (despite the conditions at which the highest $K_{a,t}$ values have been obtained could not be determined from their work). That same study mentioned the best-fitted $K_{a,t}$ value was obtained at 27.5 °C, 0.45 vvm and 375 rpm and was equal to 41.4 $h^{-1}$. Taslim and Takriff\textsuperscript{9} performed similar $CO_2$ mass transfer studies in a single-orifice OBC and reported similar values for $K_{a,t}$, although working with very large $Q_{gas}$ in the range of 1.3–3.6 vvm using pure $CO_2$. The high $K_{a,t}$ values herein reported highlight 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 $K_{a,t}$ obtained in the MOBC equipped with baffle design 3 is unique with respect to efficiency of $CO_2$ dissolution.

4. CONCLUSIONS

Major improvements in $K_{a,t}$ for $CO_2$ dissolution in water were reported for a MOBC working under oscillatory flow mixing and stagnant conditions. The $K_{a,t}$ values reported of up to $65 \pm 12 \ h^{-1}$ for very small superficial gas velocities below 1 mm s$^{-1}$ were in the range of $K_{a,t}$ 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 with respect to $D_{B,SD}$, and $K_{a,t}$ control. The scale-up of baffle configurations from single-orifice OBCs required the 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 microbubble trapping by the strong toroidal vortices in the interbaffle regions was visually shown. 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 the major increase obtained in $K_{a,t}$ for $CO_2$. As significant $K_{a,t}$ values were obtained with low $U_c$, 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 for fermentation of $C_1$ compounds that will be the subject of future publications.

ASSOCIATED CONTENT

Supporting Information
Flow visualization 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

AUTHOR INFORMATION

Corresponding Author
Tel. +44(0)1509 222 505. Fax +44 (0)1509 223 923. E-mail: n.m.reis@lboro.ac.uk.

Notes
The authors declare no competing financial interest.

ACKNOWLEDGMENTS

The authors acknowledge financial support of Fundação para a Ciência e Tecnologia (FCT) and the European Social Fund (ESF) through Ph.D. Grant SFRH/BD/62273/2009 awarded to F. M. Pereira. N. M. Reis is also grateful to the European...
Commission through the Marie Curie programme and to BBSRC, UK, for financial support.

**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 (m²)
- \( A_{proj} \) = projected area of the bubble (mm²)
- \( C_{L,0} \) = initial dissolved concentration (mg L⁻¹)
- \( C_\ell \) = dissolved CO₂ concentration (mg L⁻¹)
- \( C_P \) = concentration of saturation (mg L⁻¹)
- \( D_{SL} \) = Sauter mean diameter (mm)
- \( d_i \) = internal diameter of the column (mm)
- \( d_b \) = equivalent spherical diameter of bubble (mm)
- \( d_h \) = equivalent hydraulic diameter for single-orifice column (mm)
- \( d_o \) = orifice diameter (mm)
- \( d_{e,b} \) = equivalent diameter of the obstacle (mm)
- \( d_e \) = diameter of piston (mm)
- \( f \) = frequency of the oscillation (Hz)
- \( h \) = height of the column (mm)
- \( h_l \) = liquid height in the column (mm)
- \( K_o a \) = overall gas–liquid mass transfer coefficient (h⁻¹)
- \( K_o \) = constant of the probe (h⁻¹)
- \( L \) = spacing between baffles (mm)
- \( n \) = number of orifices in the baffle (dimensionless)
- \( Q_{gas} \) = gas aeration rate (vvm or L min⁻¹)
- \( R_e \) = oscillatory Reynolds number (dimensionless)
- \( R_{e,o} \) = modified oscillatory Reynolds number (dimensionless)
- \( S_t \) = Strouhal number (dimensionless)
- \( S_{t'} \) = Modified Strouhal number (dimensionless)
- \( t \) = aeration time (s)
- \( t_0 \) = time delay for the measuring of dissolved CO₂ concentration (s)
- \( U_{el} \) = mean superficial gas velocity (mm s⁻¹)
- \( V_l \) = working liquid volume (L)
- \( V_c \) = instantaneous axial (vertical) bubble or liquid velocity (mm s⁻¹)
- \( x_0 \) = center-to-peak amplitude of fluid oscillation (mm)
- \( x_{CO_2} \) = CO₂ 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)
- \( \gamma \) = shear rate (s⁻¹)

**REFERENCES**


