



# Mass transfer enhancement as a function of oscillatory baffled reactor design

Safaa M.R. Ahmed<sup>a,b,\*</sup>, Anh N. Phan<sup>a</sup>, Adam P. Harvey<sup>a</sup>

<sup>a</sup> School of Engineering, Newcastle University, Merz Court, Claremont Road, Newcastle Upon Tyne NE1 7RU, UK

<sup>b</sup> Department of Chemical Engineering, College of Engineering, Tikrit University, Tikrit, Salah ad Din, Iraq



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## ABSTRACT

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

## 1. Introduction

Studies on mass transfer enhancement of gas-liquid and liquid-liquid systems have been conducted in many devices including bubble columns, internal and external loop columns, gas-sparged stirred tanks, and pulsed packed column [1–3]. The bubble column (BC) is a two-phase, gas-liquid, contactor in which a gas flows as bubbles in a liquid phase. It is commonly used for oxidation, chlorination, and hydrogenation, exploiting design advantages such as good heat and mass transfer, lack of moving parts, ease of operation and low operating and maintenance costs. However, complicated flow structures, the back-mixing of the phases, and unpredictable scale up are the key disadvantages of BC [4]. Deckwer suggested adding internals (baffles) to overcome the backmixing problem in the BC [5]. In addition, pulsation has been applied to various devices, packed columns for example, to enhance mass transfer [e.g. 3]. However, the pulsation incurs greater running costs, at low frequency in particular.

Depending on operating conditions (gas/liquid velocity), four flow patterns can be observed: homogeneous (“bubbly”) flow, heterogeneous (“churn” or “turbulent”) flow, slug flow, and annular flow [4–7]. Numerous studies have been conducted to identify the flow regimes in the BC using different methods or techniques such as visual observation, evolution of global hydrodynamic parameters, Particle

Image Velocimetry (PIV), electrical capacitance tomography, Laser Doppler Anemometry (LDA), and  $\gamma$ -ray Computed Tomography (CT), entropy (photon counts) and information entropy [8–12].

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

\* Corresponding author at: School of Engineering, Newcastle University, Merz Court, Claremont Road, Newcastle Upon Tyne NE1 7RU, UK.  
E-mail address: [s.m.r.ahmed@newcastle.ac.uk](mailto:s.m.r.ahmed@newcastle.ac.uk) (S.M.R. Ahmed).

**Nomenclature**

$D$	OBR diameter, m
$f$	oscillation frequency, Hz
$x_o$	oscillation amplitude, m
$l$	baffle spacing/helical pitch, m
$k_L a$	volumetric mass transfer coefficient, $s^{-1}$
$U_G$	superficial gas velocity, $m s^{-1}$
$U_L$	liquid velocity, $m s^{-1}$
$j_{GL}$	drift flux velocity, $m s^{-1}$
$H$	liquid height in the absence of gas, m
$H^*$	liquid height with the presence of gas, m
$C^*$	the saturated dissolved oxygen concentration, $g l^{-1}$
$C$	the dissolved oxygen concentration, $g l^{-1}$
$C_o$	initial oxygen concentration at time $t = 0$ s, $g l^{-1}$
$t$	time, sec
$N_b$	the number of baffles per unit length, $m^{-1}$
$P/V$	power density, $W m^{-3}$
$St$	Strouhal number, dimensionless
$Re_n$	net flow Reynolds number, dimensionless
$Re_o$	oscillatory flow Reynolds number, dimensionless
$C_D$	the orifice discharge coefficient, dimensionless
$g$	the gravitational constant, $m s^{-2}$
$d_o$	orifice diameter, m

$vvm$	aeration rate, the volumetric flowrate of air per volume of liquid per minute
$\mu$	viscosity, Pa s
$\rho$	density, $kg/m^3$
$\varepsilon_G$	gas hold up, dimensionless
$\alpha$	open cross-sectional area, dimensionless
$\omega$	angular frequency ( $2\pi f$ )
OBR	oscillatory baffled reactor
OR	oscillatory reactor (un-baffled)
OHBR	oscillatory helical baffled reactor
OIBR	oscillatory integral baffled reactor
OSBR	oscillatory single-orifice baffled reactor
OMBR	oscillatory multi-orifice baffled reactor
BC	bubble column
OBC	oscillatory baffled column
PIV	particle image velocimetry,
CT	$\gamma$ -ray Computed Tomography
LDA	Laser Doppler Anemometry
PBR	pulsed baffled reactor
ST	stirred tank
STF	stirred tank fermenter
CSTR	continuous stirred tank reactor
DO	dissolved oxygen

examine the effect of oscillation conditions on the mass transfer enhancement for the gas-liquid system [2]. Two different behaviours of gas hold-up were observed for the effect of  $f$  on  $\varepsilon_G$  due to the different flow patterns were observed such as slug flow which occurred as a result of the OBR's geometry and the size. Moreover, it was observed that the flow patterns of the gas-water mixture were dominated by the oscillatory flow. The same behaviour for the gas hold up ( $\varepsilon_G$ ) were observed in an OBC with 50 mm i.d [18], where the  $\varepsilon_G$  enhancement appeared beyond  $Re_o = 4000$ . In addition, the calculated  $k_L a$  values obtained from an integral baffled meso-OBR were higher than that obtained in conventional gas-liquid systems (BC), stirred tank (ST), and continuous stirred tank reactor (CSTR) [19,20]. A multi-orifice baffle column (MOBC) has been used to enhance the dissolution of  $CO_2$  in water [13]. In this study,  $k_L a$  increased 3-fold compared to the un-baffled column.

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

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

## 2. Experimental apparatus and procedure

### 2.1. Experimental setup and $k_L a$ determination

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

The setup consists a 10 mm inner diameter (i.d.) and 450 mm height tube and mounted vertically. An oscillator was connected at the end of the tube to provide a sinusoidal oscillation with a wide range of

oscillation frequencies ( $f$ ) (0.5–15 Hz) and centre-to-peak amplitudes ( $x_o$ ) (1–15 mm). Various designs of baffles were used in this study including oscillatory helical baffle reactor (OHBR), oscillatory integral baffled reactor, smooth periodic constriction (OIBR), oscillatory single-orifice baffled reactor (OSBR), and oscillatory multi-orifice baffled reactor (OMBR), as well as an oscillatory reactor (OR), a smooth tube. The baffle spacing ( $l$ ) for all designs was maintained at 1.5 times column diameter. An isometric view of the OBRs and their geometries are also shown in Fig. 1b, and the dimensional details of the baffles are given in Table 1. A Mettler Toledo polarographic oxygen probe (response time: 30 s and error:  $\pm 1\%$  at 25 °C) was located at 345 mm from the gas inlet and positioned diagonally to avoid air bubbles sticking to the membrane [11] to continuously measure dissolved oxygen (DO). The DO probe was connected to a transmitter and the data was transferred to a PC via a portable EL-USB-4 Data Logger. The collected data of DO in water were analysed using EasyLog USB Software, where the profile of DO concentration against time,  $t$ , is obtained. The OBR columns were operated in semi-batch mode system (continuous gas flow and no liquid throughput) at atmospheric pressure and 20 °C. The liquid (distilled water) height ( $H$ ) was fixed at 380 mm, with the free liquid surface, for all the experiments.

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

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

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

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

where  $C^*$  and  $C$  are, respectively, the oxygen solubility and oxygen concentration in the liquid ( $g l^{-1}$ ). Assuming the liquid phase is

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