Chemical Engineering Science 62 (2007) 7454–7462

Chemical Engineering Science

www.elsevier.com/locate/ces

# The intensification of gas–liquid flows with a periodic, constricted oscillatory-meso tube

N. Reis[a,*], P.C. Mena[b], A.A. Vicente[a], J.A. Teixeira[a], F.A. Rocha[b]

[a] *IBB—Institute for Biotechnology and Bioengineering, Centro de Engenharia Biológica, Universidade do Minho, Campus de Gualtar, 4710-057 Braga, Portugal*

[b] *ISR-P—Instituto de Sistemas e Robótica do Porto, Faculdade de Engenharia da Universidade do Porto, Rua Dr. Roberto Frias, 4200-465 Porto, Portugal*

Received 21 April 2007; received in revised form 24 August 2007; accepted 10 September 2007

Available online 20 September 2007

**Abstract**

The experimental study of gas dispersion in a vertical periodically, constricted, oscillatory meso-tube (OMT) is herein presented. Water was continuously pumped through the OMT in the laminar flow regime along with an oscillatory flow component superimposed into the net flow in a range of fluid oscillation frequency ($f$) and centre-to-peak amplitude ($x_0$) of 0–20 $s^{-1}$ and 0–3 mm, respectively, in the presence of a very low superficial gas velocity (0.37 mm $min^{-1}$). Bubble images were recorded with a CCD camera and analysed with Visilog® software. A bimodal distribution of bubble size was in general observed but the bubble size was found strongly dependent on the oscillatory flow mixing conditions imposed into the fluid. A number fraction of micro-bubbles (with an equivalent diameter, $D_{eq}$, equal or bellow 0.2 mm) up to 60% was generated with increasing values of $x_0$ (i.e. 3 mm) and values of $f$ in the range 10–15 $s^{-1}$. Furthermore, it is demonstrated that the Sauter mean diameter, $D_{32}$, and the specific interfacial area, $a$, can be fined tune by setting both $f$ and $x_0$ in this studied range. The high number fraction of micro-bubbles was concluded to have a positive impact in enhancing the liquid-side mass transfer coefficient, $k_L$. Globally, the differences in bubbles sizes were found to play a marginal effect in the global enhancement of the $k_L a$ in the meso-tube in comparison with the intensive contact experimented by the bubbles rising in the oscillatory flow. The higher order of magnitude of the $k_L$ values found in this work (up to 0.0021 m $s^{-1}$) is promising for running numerous industrial gas–liquid flows processes through smaller and better, while aeration of biotransformations can be run more efficiently, as supported by our recent proof-of-concept studies carried out in the platform.


*Keywords:* Constricted tube; Oscillatory flow; Aeration; Bubble; Mass transfer; Laminar flow

## 1. Introduction

In the last few years chemical and biochemical engineers have started acknowledging the advantages of carrying out reactions at small scale. Although plant design is traditionally based in scaled-out equipment, recent developments in scale-down platforms are powering the process intensification of industrial processes because of the important key advantages presented by these technologies.

The intensification of gas–liquid processes is particularly challenging because one must assure an efficient contacting for promoting the mass transfer between a gaseous and a liquid phases with very distinct densities. Limitations of mass and heat transfer as well as hydrodynamics (fluid dynamics) should be substantially reduced in comparison with the intrinsic kinetics of the phenomena involved. In particular, the enhancement of the volumetric mass transfer coefficient ($k_L a$) in gas–liquid systems requires the ability to engineer enhanced mass transfer coefficients, $k_L$, and maximum specific interfacial area, $a$. This involves the controlling of dependencies between a large set of hydrodynamic and physical parameters, including agitation intensity, sparger configuration, bubble size, bubble velocity and residence time, gas hold-up and superficial gas-velocity, etc. This is highly demanding especially for scale-down systems because of reduction in process residence times.

Process intensification (PI) describes the strategy of making dramatic reductions in plant volume in order to meet a certain production objective. PI offers the possibility to develop and carry out chemical, pharmaceutical and biochemical reactions

* Corresponding author.
*E-mail address:* nunoreis@deb.uminho.pt (N. Reis).

0009-2509/$ - see front matter © 2007 Elsevier Ltd. All rights reserved.
doi:10.1016/j.ces.2007.09.018

in a sustainable way and with higher selectivity. The size of the equipment is adapted to the reaction, following the motivation of "doing better with less" (Ramshaw, 1999; Stankiewicz and Moulijn, 2000). Accordingly, microprocess engineering is changing the classical philosophy in plant engineering by developing "green technologies", where the number of side products is reduced and the selectivity and efficiency increased. Consequently, plant volume is shrunk and hazardous inventory reduced. Plant capital and operational costs (especially those related with cleaning) are also significantly reduced. In fact, "smaller" is a sign of "better" as demonstrated by several works and reviewed in detail (Hessel and Lowe, 2003a–c). PI is intimately related with the miniaturisation and the generation of enhanced force fields and is presently the major driving force for the development of "meso-technologies" (Pieters et al., 2006, 2007).

Gas–liquid flow is widely used in chemical and biochemical engineering, e.g. in gas–liquid catalytic reactions, fermentations and photosynthesis by micro-organisms. Extensive research has been carried out for decades in platforms conventionally used in large-scale, such as air-lift reactors, bubbles columns and stirred tanks. As most of gas–liquid processes are mass-transfer limited, a high demand still exists for innovation in continuous gas–liquid reactor designs as seen from the number of patents assigned (e.g., Schutte and Eickhoff, 2002).

Investigations of micro-technologies for gas–liquid contacting have been mainly performed in technically accepted reactor concepts, namely micro-packed bed reactor, falling-film micro-reactor and micro bubble column (Hessel and Lowe, 2003b). The high specific interfaces created in these micro-technologies are the key design for their improved performance in gas–liquid reactions. In particular, the micro-bubble column (Haverkamp et al., 2006) resembles the flow regimes in conventional bubble columns (a major established gas–liquid contacting technology) only for very low gas and liquid velocities. However, the small residence times (in the order of few seconds, as often the micro-channels are limited to few centimetre) require very high specific gas–liquid interfaces in order to speed up reactions. The bubbly mode is difficult to reproduce experimentally due to dosing problems at very small flow rates, thus increases of $a$ are obtained through operation at high gas and high liquid velocities. Consequently, it becomes difficult to set up the flow patterns maximising the specific gas–liquid interface (i.e., annular flow).

The oscillatory motion of a fluid through a set of periodic constrictions has demonstrated effective contacting and improvement of mass transfer rates in multiphase systems since Van Dick's (1935) work. However different designs were developed based on such technology. The oscillatory flow reactor (OFR) (Mackley, 1991) has proved to be the most popular design, consisting basically in a tube provided with periodic constrictions (with variable geometric configurations) where fluid pulsations (oscillations) are induced by a piston(s) installed in the tube inlet or at both ends moving sinusoidally along time. The strong radial mixing generated by the vortex rings (detaching from the narrow constrictions during flow reversal) enhances the contact between immiscible phases and improves the residence time distribution in the tube essentially for laminar net flows (Mackley and Ni, 1991). Thus, the OFR naturally presents strategic advantages in PI (e.g., Harvey et al., 2003). Additionally, studies in small internal-diameter tubes (few millimetres diameter) have shown dispersed droplets and bubbles flowing through narrow, periodic-constrictions in a particular way, namely, with reduced mean rising velocities and considerable deformations when passing through the constrictions (Graham and Higdon, 2000; Hemmat and Borhan, 1996; Muradoglu and Gokaltun, 2004; Tsai and Miksis, 1994). This contributes to an enhancement of the mean residence time and specific interfacial area for dispersed phases.

This work characterises a recently presented (Reis et al., 2005) meso-technology as a gas–liquid contactor for the intensification of mass transfer rates from a dispersed gas at a very low gas flow velocity. A small diameter tube provided with narrow, periodic constrictions and operated in oscillatory flow conditions (here mentioned as the oscillatory meso-tube; OMT) was operated with a continuous liquid flow rate and a gas phase (air) sparged at a constant, small flow rate. The bubble size and the bubble size distribution (BSD) were determined using image analysis and here in presented for different combinations of fluid oscillation frequency and centre-to-peak amplitude. A significant impact of the oscillatory mixing intensity on the fine control of BSD, specific bubble area, residence time of bubbles (i.e. gas hold-up) and liquid-side mass transfer coefficient is demonstrated, highlighting potential applications of the OMT in processes with low, efficient requirements of a dissolved gas, as found in the aeration of cell cultures.

## 2. Materials and methods

### 2.1. Experimental setup

The configuration of the OMT was previously presented (Reis et al., 2005) and is illustrated in Fig. 1. It consists in a 4.4 mm internal diameter ($d$) tube provided with narrow, smooth-periodic constrictions. The average inter-constrictions spacing ($L$) is $\sim$ 13 mm and the mean constrictions diameter ($d_c$) is $\sim$ 1.6 mm.

Gas–liquid flow experiments were run as follows. A 350-mm length OMT (with and approximate internal volume of 4.5 ml) was positioned vertically and connected to a hard tubing in the bottom through a $T$-piece, allowing the continuous pumping of a constant flow rate ($Q_L = 1.58\,\text{ml min}^{-1}$) of distilled water through the bottom of OMT with a peristaltic pump. This corresponds to a mean superficial liquid velocity, $U_L = 2.1\,\text{mm s}^{-1}$, based on the mean cross section tube diameter of 4.0 mm. The distilled water was passed through a reservoir thus (eliminating the fluid pulsations from the peristaltic pump) and a valveless, rotating piston pump (CKCRH0, Fluid Metering Inc., New York) working in a closed-loop. The ceramic piston was driven by a stepper motor and the charge–discharge motion through a single revolution reproduced very well a sinusoidal oscillation in the fluid. The fluid oscillation frequency ($f$) was controlled by setting the speed in the motor driver while the centre-to-peak amplitude ($x_0$) was controlled by setting the