On-line monitoring of multi-phase flow in a novel oscillatory screening reactor using fibre optical probes

Nuno Reis, J. A. Teixeira, A. A. Vicente[*]

Centro de Engenharia Biológica, Universidade do Minho, Campus de Gualtar, 4710-057 Braga, Portugal. Phone: +351.253.604400; Fax: +351.253.678986

Keywords: on-line monitoring; oscillatory flow; screening reactor; optical probes
Topic: Catalysis and Reaction Engineering.

Abstract

This paper demonstrates the effectiveness of using fibre optic micro-probes for the measurement of dispersion and mixing in continuous flow within a novel screening reactor operating under oscillatory flow conditions. The unsteady tracer injection technique was used at different oscillation conditions, with oscillation frequencies from 0 to 20 Hz and amplitudes from 0 to 3 mm (centre-to-peak). Application of optical micro-probes for on-line and real-time acquisition of experimental data allowed modelling and comparison with three different well-known non-ideal models (tanks-in-series, with no backflow; differential backmixing; stagewise backmixing) and with one two-parameter flow model (a plug flow and a stirred tank reactor in series). Model parameters were found by fitting the theoretical response with experimental data in both Laplace and time domains by different methods. An intermediate mixing behaviour (between plug flow and stirred tank reactor) was achieved in that range of oscillation frequencies and amplitudes. Dispersion was found to be dependent on the oscillation conditions (amplitude and frequencies) and related with the fluid backflow and with the breaking of flow symmetry. The discrete (stagewise) backmixing model was considered as the best model representing residence time behaviour in the small-scale tube.

1 Introduction

Reis et al. (2005) and Harvey et al. (2003) recently presented a novel screening reactor based on the oscillatory flow technology (Harvey et al., 2001) as a new technology for reaction engineering and particle suspension applications. Such reactor features enhanced performances at fluid micro-mixing and suspension of catalyst beads. Due to the small volume (about 4.5 ml), this novel miniature reactor is suitable for applications at specialist chemical manufacture and high throughput screening. Furthermore, a high control of environment conditions (e.g. mixing intensity, temperature) coupled with online monitoring would turn this reactor suitable for multi-phase applications at small-scale in the bioengineering field, such as fast parallel bio-processing tasks.

In this study on-line and real-time monitor and acquire information concerning reactor hydrodynamics, such as Residence Time Distributions (RTDs), is accomplished to this novel small-scale reactor by fibre optic micro-probes. The optical system can be used in the UV/VIS/NIR range allowing for applications such as colour, dissolved oxygen, biomass and bio-products concentration measurement.

2 Materials and Methods

The screening reactor unit consists of 4.4 mm internal diameter and 35 cm long jacketed glass tubes, with a unit volume of 4.5 ml and provided with smooth periodic constrictions (SPCs), with an average baffle spacing of 13 mm (Figure 1, E). This unit is able to operate under batch or continuous mode, simply by configuring the tubes in parallel or in series, according to the intended application. Mixing is achieved by oscillating the fluid at the bottom or the top of the reactor by means of a piston pump, using oscillation amplitudes and frequencies ranging from 0 to 3 mm centre-to-peak and 0 to 20 Hz, respectively. The coloured tracer used in the experiments was an aqueous solution of Indigo carmine

---

[*] Corresponding author. Tel + 351-253-604419. E-mail:avicente@deb.uminho.pt

obtained from Merck (Darmstadt, Germany). This substance was selected as it did not adsorb to installation pipes or to SPC tube walls. It has a maximum optic absorption between 610 and 612 nm.
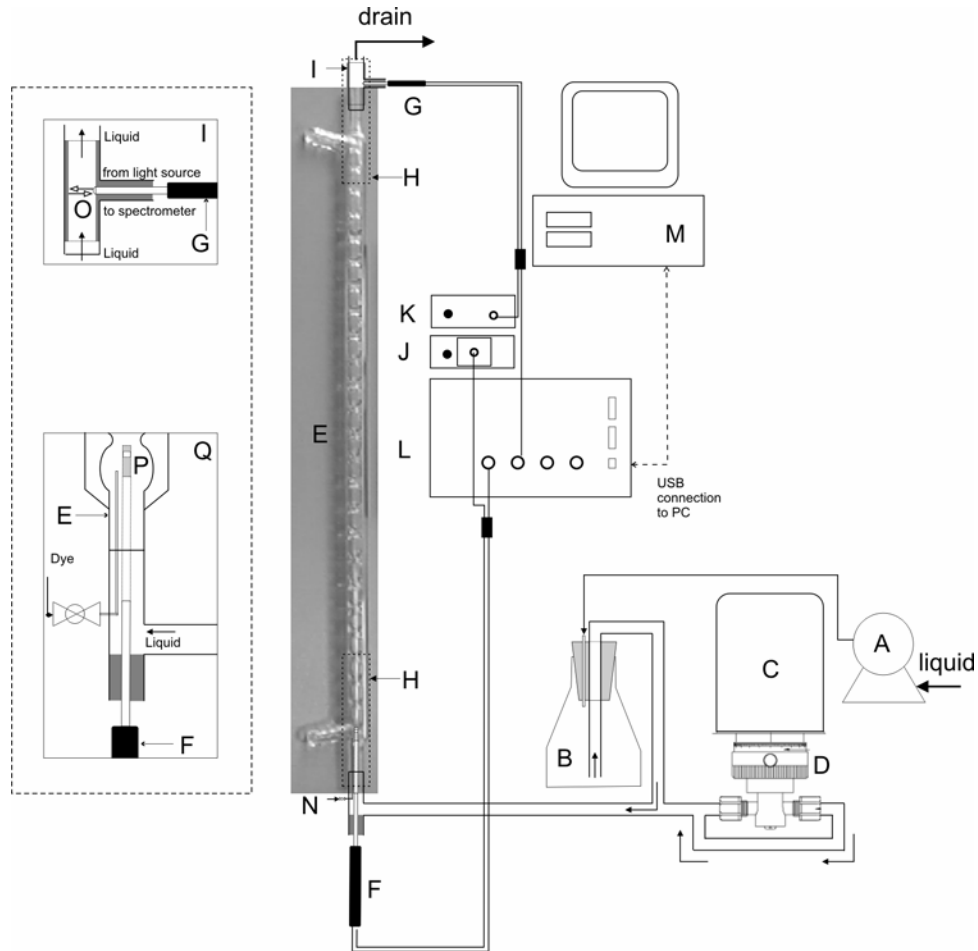
Figure 1. Experimental setup. A: Peristaltic pump; B: Reservoir; C: Electric motor; D: Piston pump; E: SPC tube; F: Micro transmission dip optical probe; G: Reflection optical probe; H: Aluminium foil; I: In-line cell; J: Tungsten halogen light source; K: 475 nm LED light source; L: Multi-channel fibre optic spectrometer; M: Personal computer; N: Tracer injection; O: Optical path of reflection probe; P: Optical path of transmission probe (2 mm); Q: section of dye injection.

2.1 Fibre optical system

On-line and real-time tracer concentration was measured by means of optical micro-probes connected to a multi-channel optic spectrometer system (Avantes, Eerbeek, The Netherlands). Due to the geometry of the SPC tube (small scale and closed system), two different probes were used. A micro transmission dip optical probe (FDP-UV-micro-1, Figure 2a), with 2 mm optical length, was used to monitor the (local) tracer concentration inside the first cavity of the SPC tube at an axial distance of 15 mm from the inlet. At the outlet (axial distance of 358 mm from the inlet point), a reflection probe (FCR-7UV200-1,5x100-2, Figure 2b) with a small tip (1.5 mm) was installed perpendicularly to the flow direction, into a 5 mm internal diameter in-line flow cell with white walls. Reading of the light coming from both probes was made on-line and simultaneously using slave1 and slave2 channels of a 4-channel optical spectrometer AvaLights-2048. The CCD detector was connected to an electronic board with 14 bit AD converter and USB/RS-232 interface. Data transfer between the optic spectrometer and a personal computer was controlled by AvaSoft full software. The system response was highly linear at tracer concentrations up to 0.3 kg m^{-3} for the transmission probe and up to 1 kg m^{-3} for the reflection probe.
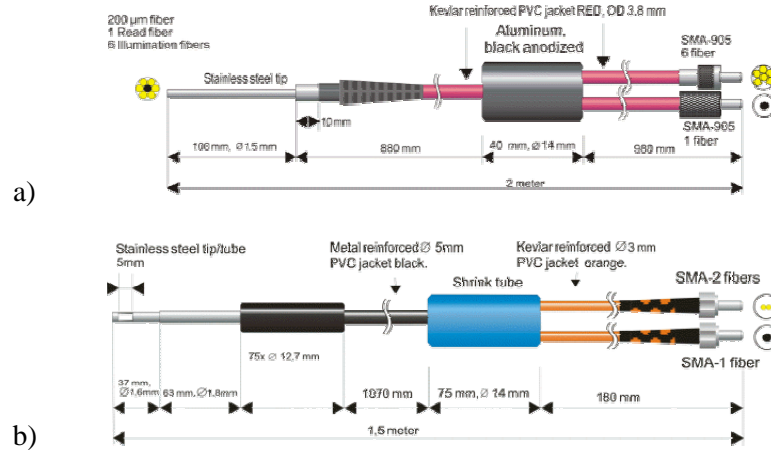
Figure 2. Optical micro-probes used in RTD study. a) transmission dip probe. b) Reflection probe.

2.2 Intermediate mixing flow models

Three non-ideal flow model and one two-parameter model were considered for RTD characterization in a SPC tube: a) tanks-in-series model; b) differential backmixing model; c) stagewise backmixing model ($N_{sw} = 26$) and d) a two-parameter model, the parameters being V_p and V_m. A schematic representation of the SPC tube according to each hydrodynamic model is presented in Figure 3.
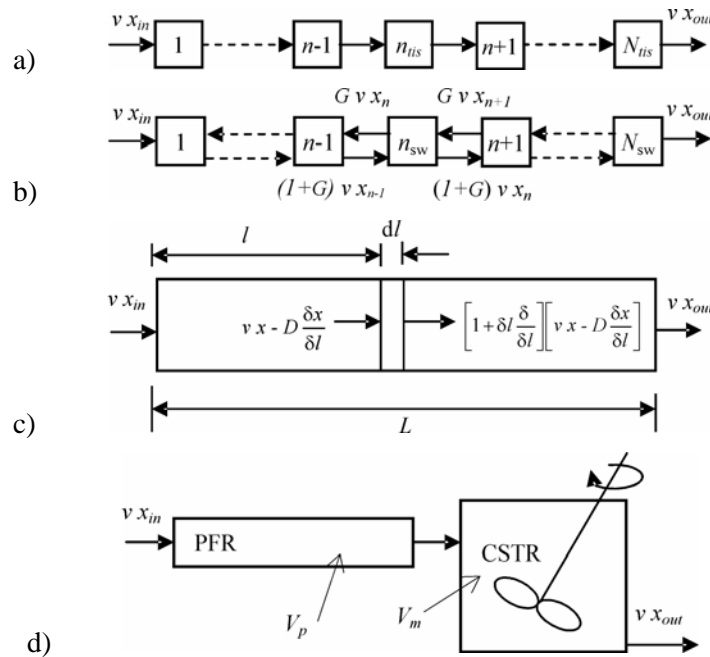

Figure 3. Schematic representation of hydrodynamic models. a) tanks-in-series. b) differential reactor with backmixing (Mecklenburgh and Hartland, 1976). c) stagewise backmixing model –mass balance only presented for stage *n* (Mecklenburgh and Hartland, 1976). d) two-parameter model, composed by a single plug (with volume V_p) and a single well-mixed (with volume V_m) region connected in series.

2.3 Estimation of model dispersion parameters

The actual parameter estimation was done by four different techniques: 1) direct nonlinear regression in time domain; 2) moments of experimental system-response curves; 3) comparison of the numerical Laplace transform of the model, $g(T)$ with that of the pulse response around a point of T that suppresses the effect of the exponential term (tail); 4) comparison of the numerical Laplace transform $g(T)$ with the pulse response found at two different points (e.g. the outlet and one internal position).

3 Results

About one hundred different experiments were run at different fluid oscillation conditions (oscillation amplitudes and frequencies). All experiments were performed at a constant net flow rate of 1.94 ml min^{-1}. Averaged mean residence times of the tracer (from optical micro-probes response) within the SPC tube at different oscillation conditions are shown in Figure 4.
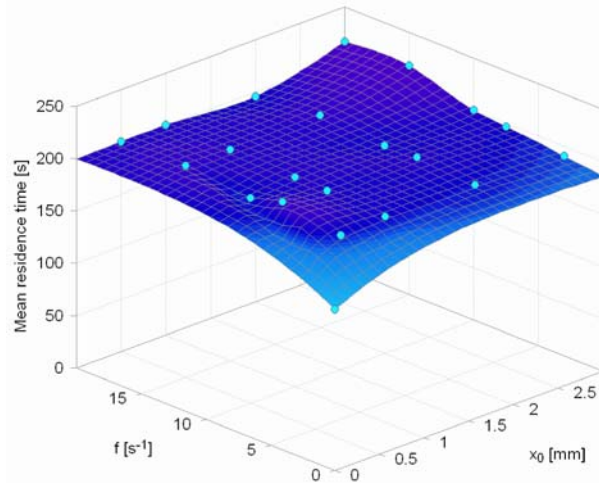

Figure 4. Average mean residence times (s) of the tracer at the outlet of SPC tube as a function of fluid oscillation conditions (oscillation frequency and amplitude). Net flow rate is 1.94 ml min^{-1}.

Time was then turned dimensionless ($\theta = t/\bar{t}$) and the quantity of the tracer represented in cumulative F-diagrams. Two typical tracer response curves at the outlet of the SPC tube are presented in Figure 5a and Figure 5b.
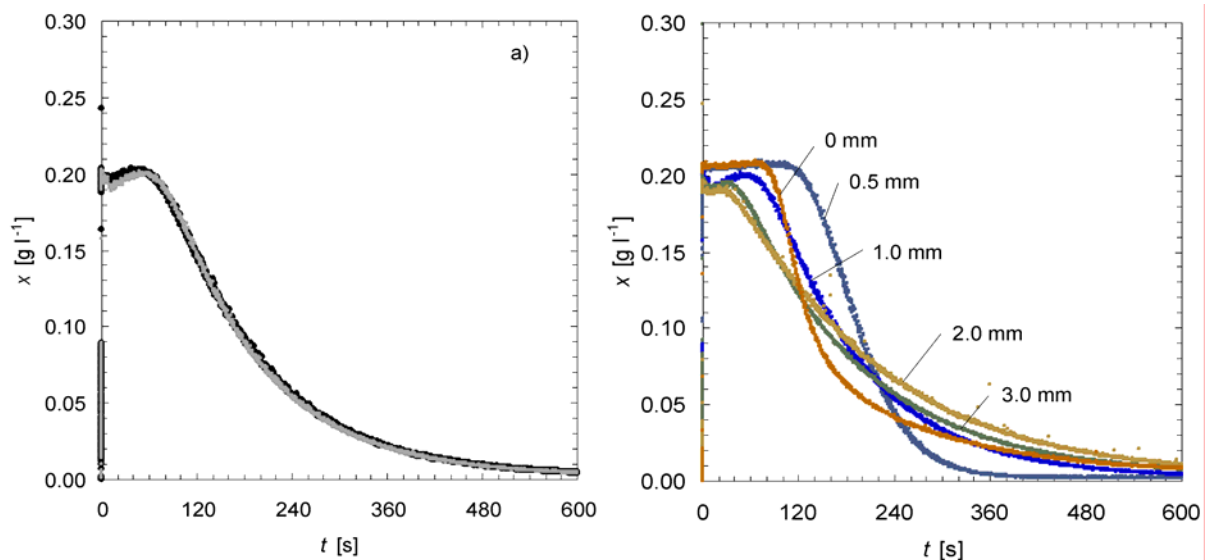

Figure 5. Tracer response curves at the outlet of the SPC tube. a) Repeatability of two different experiments (oscillation amplitude of 1 mm); b) Experimental data for fluid oscillation amplitudes of 0, 0.5, 1.0, 2.0 and 3.0 mm. Fluid oscillation frequency of 20 Hz and a net flow of 1.94 ml min-1.

F-diagrams in Figure 6a and 6b show the evolution of cumulative (dimensionless) concentration of tracer both in the first cavity (transmission dip probe) and at the exit (reflection probe) of the SPC

tube. The presented values are the best-fitted. At high-intensity oscillation conditions (mainly high oscillation amplitude) the fluid approaches a completely mixed state: the concentrations near the inlet and at the outlet become very similar and the flow within the SPC tube approaches that of a single STR.
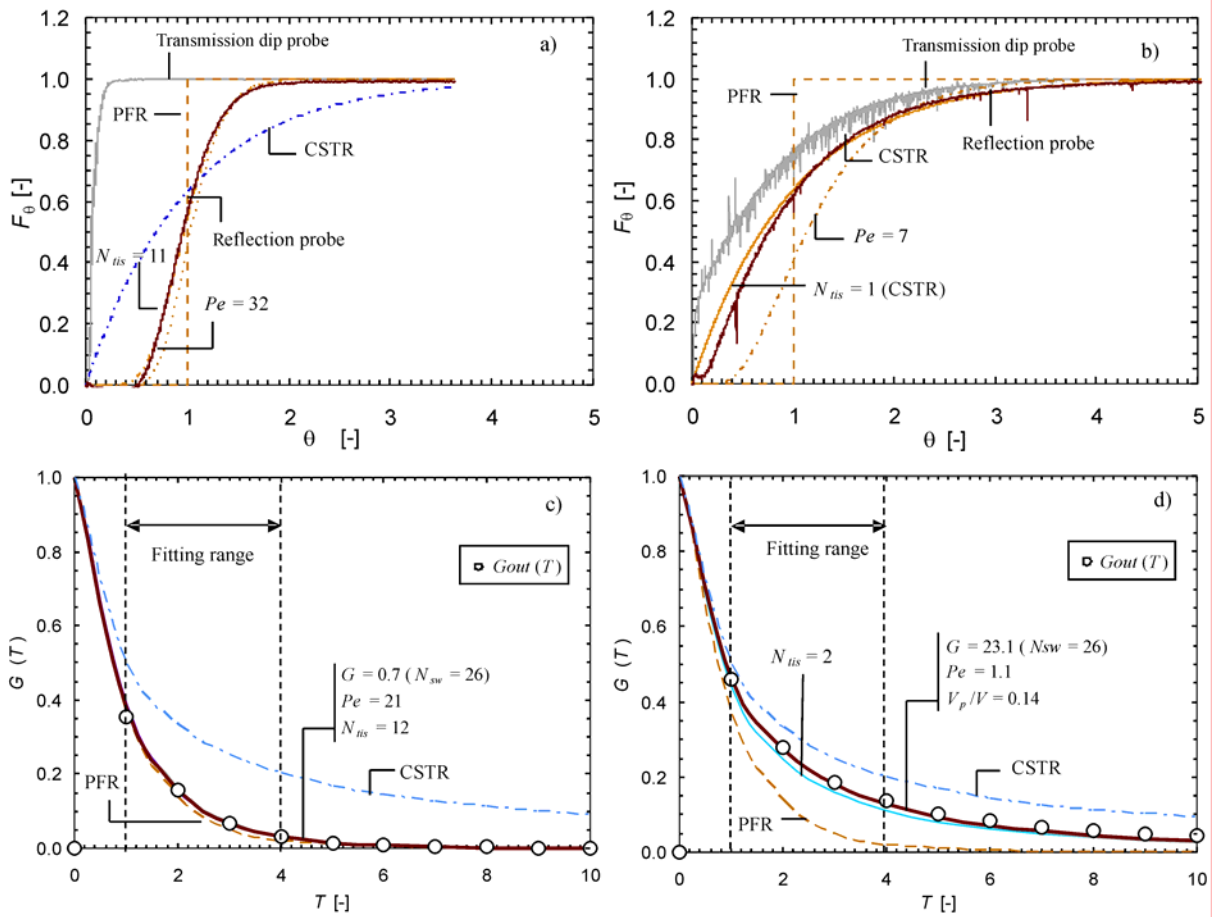

Figure 6. Best fitted parameters in (dimensionless) time – graphs a) and b) – and Laplace– graphs c) and d) – domains when fluid is oscillated at: a) and c) 3 Hz and 0.3 mm; b) and d) 20 Hz and 3 mm. Only fitting of reflection probe response signal (at the exit of the SPC tube) is presented since a perfect step change of concentration was considered at the inlet. In a) and b) typical F-diagrams of the response of the transmission (inside the first cavity) and reflection probes are shown. Net flow rate of 1.94 ml min^{-1}.

Starting from a non-oscillating state, the introduction of oscillations at low amplitudes and low frequencies led to an increase of Peclet number. This was related with the increase of intensity of the vortex rings generated inside the cavities of the reactor, leading to high radial mixing rates, decreasing the overall backflow.

It was also detected that an increase of the oscillation frequency does not affect the dispersion as significantly as an increase of the oscillation amplitude does. In general, the effect of oscillation frequency over the axial dispersion was found to be negligible and the oscillation amplitude appears to be the main factor. Mackley and Ni (1991) reported similar conclusions in the study of a conventional OFR. In the case of the SPC tube, the main exception was for the experiments performed at Re_o's below 180-200. For these conditions, the Peclet number increased (i.e. axial dispersion decreases) with the presence of fluid oscillations, from a value of about 6 in the absence of oscillations to a value of about 20 (i.e. about 30 % of decrease of axial dispersion) achieved at a Re_o of ca. 180 ($f = 7.5$ Hz and $x_0 = 1$ mm). This was believed to be related with the break of flow symmetry as reported by previous

studies (Reis et al., 2004) in the SPC tube. Ni and Pereira (2000) reported a 25 % lower dispersion for a fluid oscillated inside a conventional OFR when compared with the flow in a plain pipe. The differential backmixing model (Mecklenburgh and Hartland, 1076) successfully fitted RTDs of a single SPC tube at all the tested oscillation conditions in Laplace domain, whereas the stagewise backmixing model was the non-ideal flow model envisaged to represent RTD in a SPC tube due to its natural analogy with the geometry of the tube (number of stages equal to the number of cavities, i.e. 26) and the nature of the oscillatory flow.

The best oscillation conditions for a near plug flow behaviour were found to be oscillation amplitudes from 0.5 to 1 mm and frequencies from 7.5 to 10 Hz. The near completely mixed state can be accomplished at high oscillation amplitudes (> 3 mm) and frequencies (> 20 Hz), due to high backflow rates.

4 Conclusions

Fibre optics have been successfully applied in the determination of residence time distributions in a small-scale tube provided with smooth periodic constrictions which is the base unit of a novel oscillatory flow screening reactor envisaged for two and three-phase flows. An intermediate mixing behaviour was obtained at oscillation conditions up to 20 Hz and 3 mm centre-to-peak. This was related to the vortex rings and eddy structures formed in the reactor, which produce convective mixing in the direction of the flow, according to previously reported studies on the screening reactor.

It has been demonstrated that a RTD analysis in the Laplace domain generates reproducible results and allows fitting theoretical models that would otherwise be very difficult to test in the time domain. Considering the presence of discrete cavities and the observed good local mixing within each cavity as previously reported by Reis et al. (2005), the discrete stagewise mixing model appears to give a better physical description of flow within the small-scale tube than that of a continuous plug flow with axial dispersion.

References

Harvey, A.P., Mackley, M.R.,Stonestreet, P., (2001). Operation and optimization of an oscillatory flow continuous reactor. *Industrial & Engineering Chemistry Research*, 40(23), 5371-5377

Harvey, A.P., Mackley, M.R., Reis, N., Vicente, A.A.,Teixeira, J.A., (2003). The fluid mechanics relating to a novel oscillatory flow micro reactor. *4th European Congress of Chemical Engineering*, Granada, Spain

Mackley, M.R.,Ni, X., (1991). Mixing and Dispersion in a Baffled Tube for Steady Laminar and Pulsatile Flow. *Chemical Engineering Science*, 46(12), 3139-3151

Mecklenburgh, J.C.,Hartland, S., (1976). *The Theory of Backmixing* John Wiley & Sons, Inc., New York, pp.

Ni, X.W.,Pereira, N.E., (2000). Parameters affecting fluid dispersion in a continuous oscillatory baffled tube. *Aiche Journal*, 46(1), 37-45

Reis, N., Vicente, A.A., Teixeira, J.A.,Mackley, M.R., (2004). Residence times and mixing of a novel continuous oscillatory flow screening reactor. *Chemical Engineering Science*, 59(22-23), 4967-4974

Reis, N., Harvey, A.P., Vicente, A.A., Teixeira, J.A.,Mackley, M.R., (2005). Fluid Mechanics and Design Aspects of a Novel Oscillatory Flow Meso-Reactor. *Chemical Engineering Research & Design*, 83(A4), 357-371