The fluid mechanics relating to a novel oscillatory flow micro reactor

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1. Introduction

A novel continuous reactor based on the oscillatory flow technology [1] composed of 4.4 mm diameter tubes with smooth periodic constrictions is presented as a new technology for reaction engineering applications, particularly those involving screening solid catalysts. Experimental studies using Particle Image Velocimetry (PIV) technique [2] showed that this micro reactor improves fluid mixing and is also able to keep polymer bead-supported catalysts (with a wide range of sedimentation velocities) completely suspended. Results from 2-D and 3-D numerical simulations [3] performed on “Fluent” exhibit a semi-quantitative agreement with the experimental data. Further experiments allowed to conclude that gas bubbles retention can be prevented by configuring the tubes at appropriate angles. A single reactor consisting of a series of tubes can be configured to perform sequential reactions by keeping different particles suspended in different parts of the reactor. Potential application areas for this novel reactor are specialist chemical manufacture and high-throughput screening.

2. Materials and Methods

The micro reactor consists of a 35 cm long and 4.4 mm internal diameter glass jacketed tubes provided with smooth periodic cavities (SPC), with an averaged baffle spacing of 13 mm and a baffle thickness of 6 mm. The diameter on the constricted zone (baffle internal diameter) is 1.6 mm, leading to a baffle free area of 13 %.

The fluid was oscillated using a piston moved by an electromagnetic oscillator. Oscillation amplitudes and frequencies were ranging from 0 to 20 mm and 0 to 25 Hz, respectively. All the experiments were performed on batch mode and all the amplitudes expressed as centre-to-peak.

The fluid mechanics of the reactor was observed by PIV under several oscillation conditions (oscillatory Reynolds number - Re_o - values from 12 to 1335) and different angle positions of the SPC tube: 90°, 45° and 10°.

Numerical simulations using the CFD software Fluent 5.5 (Fluent Co., U.S.A.) were carried out in order to match the experimental observations from the PIV. The simulations started with 2-D continuous flow and then with unsteady laminar conditions. A planar and an axisymmetrical mesh were used to test the differences between these 2-D laminar sub-models. Laminar 3-D numerical simulations and 3-D Large Eddy Simulations (LES) were subsequently used. Different cells spacing between 0.3 and 0.05 mm were used to test the mesh independency at steady and unsteady flow conditions. Mesh independency was tested at continuous and oscillatory conditions.

To test the performance of the micro-OFR on suspension of particles, some particles were injected through the top of the tube and oscillated until a steady concentration distribution was achieved. Three different kinds of particles used in catalysis field were tested, namely, *a)* silica resin, *b)* polyamine resin and *c)* ionic exchange resin with maximum sedimentation velocities of 23, 1.5 and 2.5 $mm.s^{-1}$, respectively. Different particle concentrations were used.

Further experiments were performed to test the behaviour of the micro-OFR in the presence of a gas phase. Air bubbles were injected in the bottom of the tube while the oscillation conditions were kept constant.

3. Results and Discussion

PIV visualisations showed that the micro-OFR displays different flow patterns for the studied range of Re_o. In particular, the formation of axisymmetric vortex rings was observed at a Re_o of 12 and axisymmetry was broken for values of Re_o above 100. At these values of Re_o, interactions between the eddy structures are very intense and those occupy all the cavity of the reactor, thus minimizing the existence of dead zones and improving the mixture. Further PIV experiments at different tube angles showed that fluid mechanics is not affected by gravity and that for particle suspension very intensive eddy structures are required in all phase angles, which is guaranteed by operation at high oscillation frequencies.

Numerical simulations using a 2-D laminar axisymmetric model showed a broad agreement with the PIV observations for all the range of studied Re_o, namely in terms of the position and number of vortex rings. However, due to the imposed axisymmetry condition, other model had to be considered in order to match the flow patterns where axisymmetry is broken (Re_o above 100). A 2-D planar laminar model was also unable to break the axisymmetry and consequently the use of 3-D models was considered. The 3-D laminar model used showed to be unsuitable to match the visual observations and this was only achieved when a 3-D Large Eddy Simulation (LES) model was employed. The comparison of the PIV observations with the results of the 2-D laminar axisymmetric model and the 3-D LES model is made in Fig. 1.

The micro-OFR was capable of keeping a maximum of 40 % (v/v) of silica particles in a well mixed suspension, when oscillated at 12.1 Hz and 4 mm, being the tube placed in the vertical position. No visible particle concentration gradient was detected throughout the tube. For angles approaching the horizontal position the suspension was even easier, being 12.1 Hz and 3 mm sufficient to keep the same amount of particles suspended.

This novel micro-OFR also presents extra performance in terms of bubble suspension. The capacity of this reactor to keep gas bubbles suspended inside each cavity means a several-fold increase of the mean residence time of the bubbles, leading to an increase in mass transfer rate which is useful in many industrial processes. An auto-cleaning capacity of the micro-OFR was observed at angles above 45°. At lower angles (near horizontal position) the smallest bubbles are retained inside each cavity. Furthermore, the presence of solid particles, coupled with intensive mixing, brakes the bubbles aggravating this problem. The optimum angle for design of the novel micro-OFR was found to be 10°. At this position all the bubbles can be cleaned from the system in less than 1 min if a net flow of 14 $ml.min^{-1}$ is coupled with the oscillatory flow.

The envisaged reactor unit will be composed of a series of SPC tubes, the oscillation being provided by a single oscillator at the bottom of the first tube. Different particles at different environment conditions can be supported in each tube. Several reactor units can be stacked in parallel, allowing for uses such as high throughput screening. This configuration allows for on-line measurements and a good control of operation conditions.

4. References

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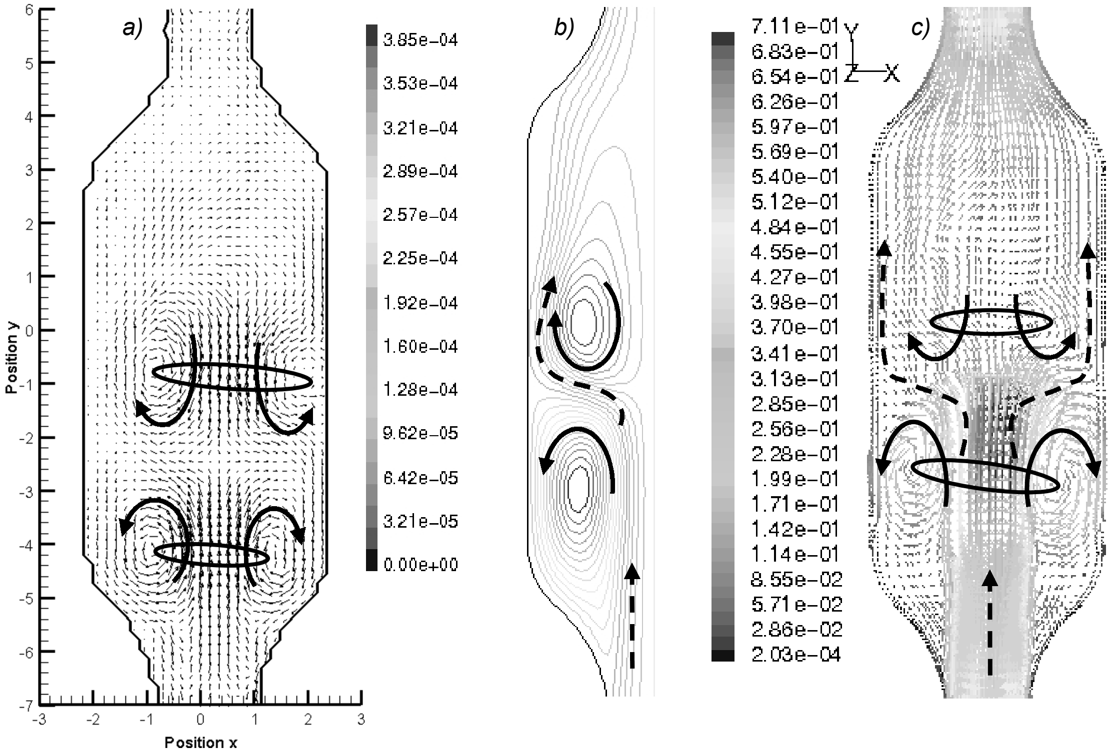

Fig. 1 Results from PIV visualisations and numerical simulations at 1/5th way through cycle, i.e. before flow reversing: *a)* phase-averaged velocity vector map measured by PIV at Re_o = 348, x_0 = 1.1 mm and f = 11.1 Hz; *b)* contours of stream function (Kg/s) of the simulated flow patterns for Re_o = 450, x_0 = 0.8 mm and f = 20 Hz, using a 2-D axisymmetric laminar model, after 2 simulation cycles; *c)* simulated velocity vectors graded by velocity magnitude (m/s) for Re_o = 350, x_0 = 1.2 mm and f = 10 Hz, using a 3-D LES model, after 7 simulation cycles. No net flow, i.e. Re_n = 0. Fluid sinusoidally oscillated at the bottom of the grid. Dash arrows illustrate the flow direction and solid arrows illustrate the vortex rings position.