Residence Time Distribution in Corning ®Advanced-Flow™ Reactors. Experiment and Modelling

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Corning® Advanced-Flow™ glass reactors are continuous flow reactors with hydraulic diameters in the millimetre range. These devices enable the switch of chemical reactions from batch mode to continuous processing through more efficient, more economical and safer processes. In addition, these reactors provide a platform for developing innovative chemistries that have never been considered industrially practical, either for hazard or yield reasons.

Corning proprietary apparatuses are compact, adaptable and scalable, optimizing overall production cost and quality of high-value specialty, fine, and pharmaceutical chemicals. Corning Advanced-Flow™ glass reactors are composed of multiple inter-connected devices having different designs, offering the advantages of process intensification.

This paper presents the comparison between experimental and CFD modelling results for the residence-time distribution of a family of glass devices. As the simulated residence-time distribution curves compared very well with experimental data, CFD was used to predict the flow behaviour of new, much smaller devices that are more difficult to characterize experimentally.

1. Introduction

Microreaction technology provides opportunities for improving process capability and control in chemical/biochemical synthesis, and can allow safer and more efficient chemical/biochemical kinetic investigations. Compared to normal scale reactors, microreactors provide an increase in surface to volume ratio due to their small dimensions, fast and efficient process development, lower environmental impact, and increased safety. In addition, these large surface-to-volume ratios improve heat transfer for exothermic reactions, preventing either thermal degradation or explosive evolution (Mae, 2007; Watts and Wiles, 2007; Hessel et al., 2008). The operating conditions, which offer more accurate control than for classical reactors, allow the obtaining of higher yields, which makes the separation process cost-effective.

Microreactor technology provides relatively simple and quick means towards the commercialization of a process, since the scale-up is avoided and a reasonable number of reactors can be connected in parallel for mass production (Lavric and Woehl, 2009).

Corning has developed proprietary continuous small scale reactor processing technologies that are compact, adaptable and scalable, thereby optimizing the overall quality of high-value specialty, fine, and pharmaceutical chemicals. The key component of the Corning® Advanced-Flow^{TM} Reactors is a specialty glass fluidic module which integrates good mixing and/or dwell time with heat transfer and has hydraulic diameters in the range of 0.3 up to a few millimetres. These devices exhibit internal volumes from 0.5 to about 100 ml and are devoted to both laboratory studies and production.

Residence time distribution (RTD) is a reasonable indicator of the type and extent of mixing in a flow system. Although the concept of RTD is not new when it comes to its application to conventional flow systems (Adeosun and Lawal, 2010), its use in the characterization of flow and mixing in micromixers is still novel (Lohse et al., 2008; Trachseletal., 2005; Salman et al., 2007; Cuantu-Perez et al., 2008; Boskovic and Loebbecke, 2008; Adeosun and Lawal, 2009; Adeosun and Lawal, 2010).

By performing so-called tracer or stimulus-response experiments, RTD data can be obtained that can be used for flow and mixing evaluation in continuous flow systems. Information from tracer experiments not only provides a quantitative measure for mixing, but likewise indicates qualitative features such as the presence of dead zones or channelling, by-passing, etc. With the present available technology, the challenges associated with setting up a suitable experimental technique become amplified as the devices to be characterized get smaller. Thus, developing a “miniaturised version” of the conventional RTD characterization technique for micromixers/reactors is one of the challenges associated with the successful evaluation and optimization of these devices.

In this work, RTD has been analyzed by a series of numerical experiments with a full 3D Computational Fluid Dynamic model using FLUENT software. The developed approach includes: (a) the steady-state calculations of the flow velocity pattern, and (b) transient calculations of RTD. In the transient calculation, the flow of massless tracer particles was superimposed, and their motion through the fluidic module was monitored using the previously computed velocity fields. Our prior study, carried out in order to characterize the velocity profiles that showed good agreement with the experimental data (Chivilikhin et al. 2010), was used as a foundation for RTD analysis.

2. Experimental analysis

The analyzed devices, largely described elsewhere (Chevalier et al., 2008; Lavric and Woehl, 2009), are composed of chains of identical cells having variable cross sections and internal elements (Figure 1). The device incorporates an injector, enabling feeding and mixing of two fluids, but can also be used as dwell only.

The experiments were carried out for 1X devices (Figure 1, left) having an internal volume of about 8 mL and a recommended operating flow rate in the range of 30 to 120 g/min for water-like fluids.

The RTD can be obtained by performing tracer or stimulus–response experiments, in which a tracer is injected instantaneously (a pulse input) or at a constant rate (a step input) at the inlet of a flow system, and its concentration, C(t), is measured at the exit as a function of time. RTD analysis is generally applicable to a flow system with one-inlet stream where a tracer is injected (Adeosun and Lawal, 2010).

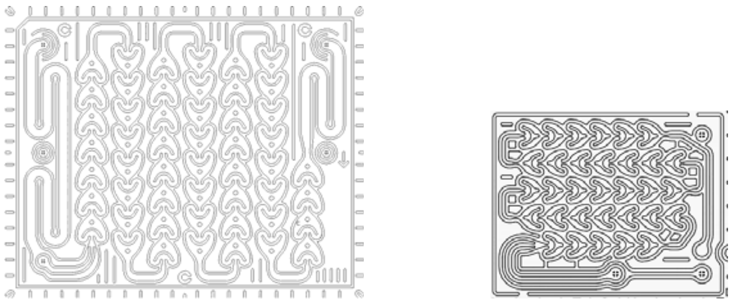

Figure 1: Mixing and/or dwell time fluidic module for multi-phase applications. Left side: 1X. Right side: low-flow fluidic module

This limitation is mitigated in the numerical simulation and in the experiment for our two-inlet, one-outlet flow system by introducing a carrier fluid (water) with a flow ratio of 1:100. This reduces dead volumes within the devices and effectively makes the flow system a one-inlet, one-outlet system as desired, with practically no flow disturbance. The exit concentration of the tracer, a solution of dye, was measured using a spectrometer.

Although the pulse input method directly provides the RTD curve, it is hard to obtain a reasonable pulse of the tracer because of the short residence time characteristic to the fluidic modules (less than 10 s). Therefore, the step input was used.

Different flow rates were investigated, with at least two replicates for each flow rate.

3. Numerical analysis

For this study, RTD was analyzed by a series of numerical experiments with a full 3D Computational Fluid Dynamic model using FLUENT software. The used approach relies on: (a) the steady-state calculations of the flow velocity pattern, and (b) transient calculations of RTD. In the latter, the motion through the fluidic module of superimposed massless tracer particles was monitored using previously computed velocity fields.

GAMBIT pre-processor was used for extracting the fluid flow domains of the geometries studied and for meshing.

For the 1X device, the whole mesh size for a velocity field computation consists of 25 million cells, and it takes about 30 h to achieve convergence. The results of the hydrodynamic simulation showed very good agreement with flow fields measured with μPIV and with the experimentally measured pressure drop (Chivilikhin et al. 2010).

The mesh size of a low-flow fluidic module is composed of 3.4 million cells and convergence was reached in 5 h.

The FLUENT segregated pressure-based solver was used for the solution of 3D fluid flow and species transport problem, in which the respective equations were solved in a sequential manner using appropriate boundary conditions and numerical algorithms. The steady state solution to the flow equations was obtained by specifying as boundary conditions: the mass flow rate at the inlet feeds, no-slip condition at the walls, and gauge pressure of zero at the outlet of the configurations. Using the converged solution of the steady state fluid flow equations, the tracer species equation was solved as an unsteady simulation, by specifying zero diffusive flux as boundary condition at the walls; its solution was then used for RTD analysis. Species mass fractions of one (for time t=0 at step injection of tracer into water) were specified at the inlet zone and maintained for subsequent time steps. Using FLUENT's integrated postprocessor, tracer concentration data is acquired at the outlet from the time-dependent nodal values of the mass fraction of the tracer.

The cumulative, F, was computed as in experimental studies, using the predicted tracer concentration at the fluidic module outlet. The length of the tubing connecting the injection point to the flow/mixing system and its outlet to the spectrometric cell was reduced as much as possible, so as to minimize the axial dispersion of the tracer before reaching the device and respectively the detection zone. Despite this, its volume together with the cell still represents about 40 % of the internal volume of the fluidic module. Therefore the contribution of connectors, tubing and spectrometric cell was taken into account in the modelling.

The outlet concentration was used to compute the exit age distribution, *E* curve, using the time step Δt_i:

$$E(t) = \frac{C_{out}(t)}{\int\limits_0^\infty C_{out}(t)dt} \cong \frac{C_{out}(t_i)}{\sum\limits_{i=0}^{\infty} C_{out}(t_i) \cdot \Delta t_i} \tag{1}$$

4. Results and discussion

Experimental and numerical data was obtained at different volumetric flow rates in the range of 50 to 250 g/min.

For the species transport simulation, the spatial average concentration data, which reasonably represents the measurements, was obtained at the outlet of the device, in order to closely mimic the experimental concentration data acquisition.

The functions of repartition, F (as a function of time), data obtained at 50, 100 and 250 g/min from experiment and CFD simulation are plotted and shown in Figure 2 left. The plots show good agreement between the simulation and the experimental results. The normalized density of the distribution functions, E(θ), obtained from simulation at 50 g/min, for the fluidic module including connecting system and spectrometric cell, and respectively only for the fluidic module, are shown in Figure 2 (right). This reveals that the unavoidable connectors and flow tubing lengths introduce an important axial dispersion. Figure 2 (right) also shows that, even at flow rates in the lower part of their operating range, these fluidic modules have plug-flow-like behaviour.

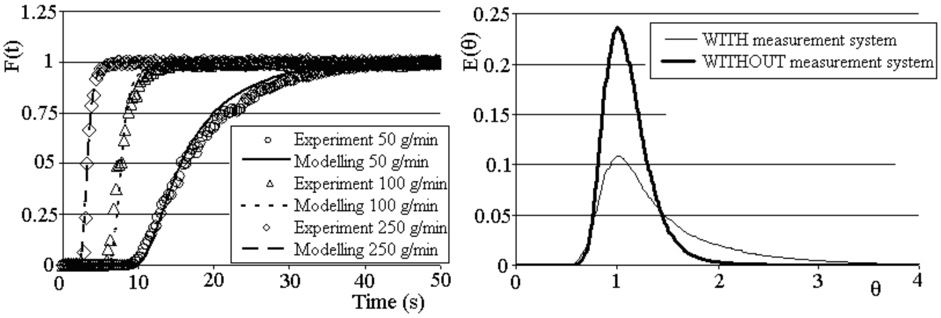

Figure 2: Residence time distribution of 1X device. Left side: Experimental and modelled response step signals. Right side: simulated RTD of the whole set-up (fluidic module and measurement cell - slab line) and of the fluidic module (gross line)

Numerical simulation having been validated against experimental data, it was then used to characterize the RTD of much smaller devices with an internal volume of 0.5 mL (Figure 1 right). The function of repartition, F (as a function of time), as well as the normalized RTD (as a function of dimensionless time), obtained at the nominal flow rate of 5 g/min are shown in Figure 3. In essence, the design generates flow patterns that give a narrow RTD.

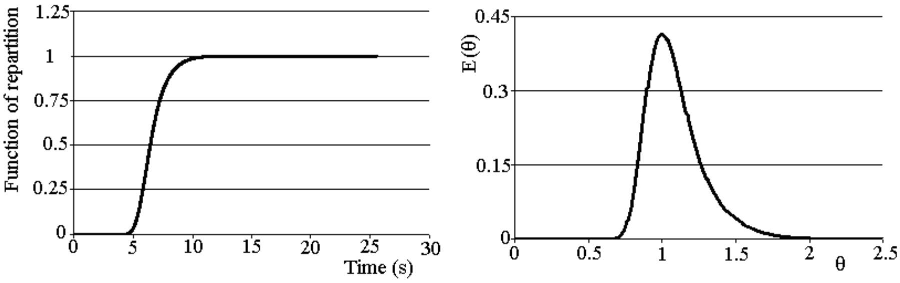

Figure 3: Function of repartition (left) and density of distribution for low-flow fluidic modules

Conclusions

The axial mixing induced by the flow through a family of fluidic modules used in Corning® Advanced-Flow™ Reactors was investigated experimentally as well as numerically. The work involved the analysis of the response of the device to a step-tracer input, the evaluation of residence-time distribution and the use of CFD simulations as a vital design, optimization and characterization tool. The RTD predictions have been found to be consistent with the experimental results.

The validated CFD model provided RTD data for much smaller fluidic modules whose experimental characterization is much more difficult.

The RTD exhibited by the analyzed devices is narrow, showing plug-flow behaviour.

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