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Experimental characterization of axial dispersion in coiled flow inverters

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ABSTRACT

Narrow residence time distributions (RTDs) are desirable in many chemical engineering processes. However, when a system operates in the laminar flow regime, significant fluid dynamic dispersion takes place. This problem is often encountered in micro and millifluidic devices. Exploiting the beneficial effects of secondary flow and chaotic advection, so-called coiled flow inverters (CFIs) are a promising solution for the reduction of fluid dynamic dispersion. These devices, however, have not been extensively used due to the lack of experimental data and of correlations relating the design parameters and operating conditions to the amount of axial dispersion. In this work, we investigated RTDs in micro and millifluidic devices using step input injection and UV–vis inline spectroscopy for the detection of the concentration of a tracer. Experiments were performed for different operating conditions and geometries. Helically coiled tubes (HCTs) were similarly characterized. Dispersion data were expressed in terms of an axial dispersion coefficient and an empirical correlation was derived. The experimental results show that lower axial dispersion is achieved in CFIs as compared to HCTs and straight tubes.

1. Introduction

In micro and millifluidic devices, the flow behavior is dictated by viscous forces rather than inertial forces, as a result of low operating Reynolds numbers (Squires and Quake, 2005). In devices with simple geometry (such as straight channels), the absence of turbulence makes diffusion the only transport mechanism in the radial direction and, despite the short length scales characterizing the flow, in general this is a rather slow process. A whole branch of microfluidics has been dedicated to the development of complex microstructures capable of achieving faster and effective mixing. Parallel and serial lamination, split and recombination, and flow focusing are examples of passive micromixers designed to shorten mixing times (Hessel et al., 2005; Nguyen and Wu, 2005). These are effective tools in applications in which fast mixing between different streams is required. However, in many continuous processes what is essential is attaining low fluid dynamic axial dispersion. Approaching plug-flow behavior is required for a wide range of chemical reactions as well as for the synthesis of nano and microparticles (Marre and Jensen, 2010).

A solution proposed for approaching plug-flow behavior resorts to multiphase segmented flow (Taylor, 1961). Slugs or droplets behave as small batch reactors traveling through the system, effectively reducing residence time dispersion. However, forming stable slugs or droplets, particularly for cases in which long residence times are needed, can be quite challenging. Furthermore, the introduction of an additional phase requires downstream separation, which may be undesirable in industrial applications.

In this work, we focus on a class of simple structures that have been shown to act positively on the fluid dynamics of tubular systems operating under laminar flow conditions. Exploiting the action of the centrifugal force, one can form a secondary flow that induces recirculation in the radial direction and which alters the radial velocity profile, rendering it more uniform. This flow can be achieved in helically coiled tubes, constructed with capillaries wrapped on cylindrical supports. By

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changing the direction of action of the centrifugal force, the radial vortices, generated by the latter, rotate around the axis of the tube; this results in further mitigation of the radial velocity gradients. This idea was firstly proposed by Saxena and Nigam (1984) in their pioneering work on coiled flow inverters (CFIs) and, since then, several works have been published in this field.

Enhanced mixing owing to secondary flow has been exploited primarily in heat transfer operations. Radial mixing induces forced convection, reducing the thickness of boundary layers and improving overall heat exchange. This, combined with the compactness of CFIs and their low manufacturing cost, has motivated several research groups to investigate the extent by which heat transfer is enhanced in these devices. Correlations between Nusselt number and design parameters of CFIs (and other systems with coiled geometries) have been widely explored theoretically (Acharya et al., 2001, 1992; Kumar and Nigam, 2005; Lemenand and Peerhossaini, 2002) and experimentally (Acharya et al., 1992; Chagny et al., 2000; Mokrani et al., 1997; Singh et al., 2014). Recently, it has become common to use the heat transfer correlation of Mandal et al. (2010), derived for CFIs. Furthermore, heat transfer in coiled configurations without flow inversion has also been widely investigated; the summary of the finding can be found in the papers of Kumar and Nigam (2007), for laminar flow, and Mridha and Nigam (2008), for turbulent flow.

In terms of industrial implementation, a review by Vashisth et al. (2008) has outlined the potential application of curved tubes; this remains the most comprehensive review on the topic. However, at the point of writing the review, only limited research had been done on topics other than heat transfer. In the last decade, the focus has started to shift to other applications. CFIs have been used as a platform for polymerization (Parida et al., 2014a,b; Mandal et al., 2011), in which case it was shown, both numerically and experimentally, that monomer conversion in coiled flow reactors was higher than that achieved in equivalent straight channel reactors. In another application, CFI membrane modules were shown to be beneficial in oxygenation and carbonation of water, with significant mass transfer enhancement being observed (Singh et al., 2016). In biotechnology, CFIs have been utilized for protein and antibody synthesis (Sharma et al., 2016; Kateja et al., 2016; Klutz et al., 2016) owing to their ability to enhance mixing and narrow down RTD widths.

Two-phase flow in CFIs deserves a separate mention due to abundance of research in this area. Fundamental studies into pressure drop (Vashisth and Nigam, 2007), gas–liquid mixing (Vashisth and Nigam, 2008a) and RTDs of a single phase in a two-phase flow (Vashisth and Nigam, 2008b) have been performed. Furthermore, slug flow in CFIs was used in continuous flow liquid–liquid extraction processes (Kurt et al., 2016; Vural Gürsel et al., 2016; Zhang et al., 2017). In this application, up to 20% increase in extraction efficiency was reported and this was explained by enhancement of mass transfer induced by the formation of Dean vortices (Kurt et al., 2016). Also, a cooling crystallization application of the CFI concept was recently reported (Hohmann et al., 2016) where narrow RTDs and smooth axial temperature profiles were outlined as the main benefits.

A general guideline for design of CFIs has being outlined by Klutz et al. (2015). In this publication, the key design parameters and the effect of each parameter on the device performance are outlined. Furthermore, the "best performance" design space of a CFI reactor is identified for the configuration and operating conditions considered. However, no general correlation for the relationship between the design variables and axial dispersion coefficients was derived. Other studies investigated the effect of number of bends (Castelain et al., 1997) and tube curvature ratio (Palazoglu and Sandeep, 2004) on RTDs, but no correlation which can be used to predict RTDs was suggested. A recent paper by Sharma et al. (2017) has proposed an empirical relation for the Peclet number as a function of the Dean number and of the number of bends. The proposed relation is only valid for a small number of bends (up to 3), which is suitable exclusively for previous design by the same authors, US. Pat. US007337835B2 (Nigam, 2008). The design consists of several 3-bend CFIs in series which can be stacked to achieve the required residence time, internal volume, heat exchange area etc. The correlation, as one would expect, cannot be used with other system configurations.

There is currently a "barrier of entry" into research on applications on CFIs. Lack of design correlations significantly limits the scope of design in several cases. The aim of this paper is to characterize experimentally the axial dispersion in coiled flow inverters considering in particular the effect of the Dean number and of the coil-to-tube diameter ratio. The latter is a crucial design parameter; however, in the literature few studies have been devoted to its effect on axial dispersion. The paper also aims to propose a correlation which can be later on utilized as a short-cut method for the design of CFIs of similar configuration to the one investigated. To this end, we resorted to a reliable and flexible experimental procedure to perform RTD experiments on different micro and millifluidic devices. The adoption of a suitable flow model was necessary to analyze and quantify fluid dynamic dispersion. The axial dispersion model (ADM) (Aris, 1960; Danckwerts, 1953; Levenspiel and Smith, 1957; Taylor, 1953) is the most adopted model; nevertheless, analytical correlations are only available for a limited class of systems, which does not include coiled flow inverters. The adoption of this model to CFIs, therefore, is also assessed.

This paper is organized as follows. In Section 2, we discuss the axial dispersion model and how to use RTDs to derive information on axial dispersion. In Section 3, we present our experimental system for performing RTD studies. In Section 4, we report the experimental results of our study on CFIs and helically coiled tubes (HCTs) and propose a correlation relating the dispersion number to the coil-to-tube diameter ratio and the Reynolds number for a fixed number of bends.

2. Theory

2.1. *Axial dispersion model*

The ADM is an extension of the PFR model that accounts for the longitudinal dispersion of a tracer flowing in the axial direction in straight tubes. The tracer concentration is assumed to be radially uniform (Schaschke, 2014). The model is based on the following equation:

$$\frac{\partial C}{\partial t} = -u\frac{\partial C}{\partial z} + D_{ax}\frac{\partial^2 C}{\partial z^2} \tag{2.1}$$

where C is the tracer concentration, dependent on the time t and on the axial coordinate z, u is the mean axial fluid velocity and D_{ax} is the axial dispersion coefficient, assumed to be constant. The axial dispersion coefficient is assumed to be independent of both the axial position and the tracer concentration. It relates to the axial dispersion rate in the vessel. To characterize the axial dispersion process under different conditions, one usually adopts dimensionless numbers. In dimensionless form, Eq. (2.1) reads:

$$\frac{\partial \bar{C}}{\partial \tau} = -\frac{\partial \bar{C}}{\partial \xi} + \frac{1}{Pe_L}\frac{\partial^2 \bar{C}}{\partial \xi^2} \tag{2.2}$$

where $\tau \equiv tu/L$ and $\xi \equiv z/L$, L being the length of the vessel. $\bar{C}$ denotes the dimensionless tracer concentration (the scaling factor can be chosen arbitrarily, as the equation is linear in the concentration; usually it is chosen to be equal to the inlet concentration). The model features a single parameter known as the Peclet number, $Pe_L \equiv uL/D_{ax}$.

The inverse of Pe_L is the vessel dispersion number N_L. This is a measure of the spread of tracer in the whole vessel; more precisely:

$$\frac{\delta}{L} \sim \sqrt{N_L} \tag{2.3}$$