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.

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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 Peershossaini, 2002) and experimentally (Acharya et al., 1992; Chaguy 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 theoretically 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 (Klohnmann 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 (Palazoglug 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. US00733783582 (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:

$$\frac{\partial C}{\partial t} = -\frac{1}{r} \frac{\partial}{\partial r} \left( r \frac{\partial C}{\partial r} \right) + D_{ax} \frac{\partial^2 C}{\partial z^2}$$

(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:

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

(2.2)

where \( \tau = tu/L \) and \( \xi = z/L \), \( L \) being the length of the vessel. \( \hat{C} \) 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, \( Pe_L = uL/D_{ax} \).

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:

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

(2.3)
where $\delta$ is the axial spread attained at the outlet of the vessel (that is, after a time equal to $L/u$). Equivalently, $N_t$ can be regarded as the ratio of the convection time to the dispersion time; thus, the larger the vessel dispersion number is, the broader the RTD is. $N_t$ is widely used to characterize fluid dynamic dispersion and to compare dispersion performances of vessels of different lengths or operated in different fluid dynamic conditions.

Often, a similar group is employed in the literature, defined by using the pipe diameter $d_t$ as characteristic length:

$$N_d = \frac{D_{ax}}{ud_t} \quad (2.4)$$

$N_d$ has been widely used to characterize dispersion in dimensionless flow regime maps as a function of the Reynolds number $Re$ (Levenspiel, 1999).

The axial dispersion model can be applied to both turbulent and laminar flows. However, while in the first case there are no limitations to its applicability, for laminar flows the model holds only under certain conditions. These are reported in Fig. 1, where the system is characterized in terms of the Bodenstein number:

$$Bo = Re \cdot Sc = \frac{\rho ud_t}{\mu} \frac{\mu}{\rho D_m} = \frac{ud_t}{D_m} \quad (2.5)$$

($D_m$ is the molecular diffusivity of the tracer) and of the ratio $L/d_t$. If the point identified by these two coordinates falls within the region of validity of the axial dispersion model, the latter is applicable. The figure also reports the analytical expressions (derived theoretically by Aris (1960) and Taylor (1953) for straight channels) needed to calculate the axial dispersion coefficient. In particular, it is:

$$Bo < 100 : \quad D_{ax} = D_m + \frac{u^2 d_t^2}{192 D_m}$$

(2.6)

and

$$Bo > 100 : \quad D_{ax} = \frac{u^2 d_t^2}{192 D_m}$$

(2.7)

These theoretical expressions can be verified experimentally, as described in the following section, by resorting to RTD experiments.

2.2. Calculation of the axial dispersion coefficient

The axial dispersion coefficient cannot be measured directly from experiments; however, it is possible to fit experimental data to the solution of the axial dispersion model (that is, of Eq. (2.1)). Two different methods of fitting experimental data exist based on the extent of axial dispersion in the system, small ($N_t < 0.01$) and large ($N_t > 0.01$).

For small extents of dispersion, the shape of the RTD curve varies little on passing through the outlet of the vessel (and in particular through the tracer detection system). This means that the resultant experimental RTD curve is close to a Gaussian distribution. In this case, the experimental RTD curve can be fitted with the analytical solution valid for open–open boundary conditions, as suggested by Levenspiel (1999):

$$N_t < 0.01 : \quad \epsilon_{t,ADM} = \sqrt{\frac{\pi}{4D_{ax}L}} \exp \left[ -\frac{(L-ut)^2}{4D_{ax}L} \right]$$

(2.8)

For large extents of dispersion, the RTD curve changes substantially on passing through the vessel outlet, becoming skewed and therefore asymmetrical. In this case, it is fitted to the following curve (Schaschke, 2014):

$$N_t > 0.01 : \quad \epsilon_{t,ADM} = \frac{u}{\sqrt{\pi D_{ax}L}} \exp \left[ -\frac{(L-ut)^2}{4D_{ax}L} \right]$$

(2.9)

The experimental RTD curves are fitted to Eq. (2.8) or Eq. (2.9) so that the only unknown parameter, $D_{ax}$, can be calculated.

This study focuses on CFIs and HCTs, not on straight pipes; therefore, there is no guarantee that the axial dispersion model (that is, Eqs. (2.8) and (2.9)) should hold and that an axial dispersion coefficient can be used to quantify the axial dispersion in these systems. This needs to be verified.

The analytical solution (either Eq. (2.8) or Eq. (2.9)) to use depends on the vessel dispersion number $N_t$, which is unknown before performing the fitting. As a consequence, the fitting was carried out using both solutions and then selecting the correct one. The fitting was based on the least squares method and the residual error $\epsilon$ of the solution was used to
judge the quality of the fitting. The error \( \epsilon \) is calculated as follows:

\[
e = \sum_i [E_{t,ADM}(D_{ax}, t_i) - E_{t,exp}(t_i)]^2
\]  

(2.10)

where \( E_{t,ADM}(D_{ax}, t_i) \) is the value of the analytical RTD curve at any time \( t_i \) and \( E_{t,exp}(t_i) \) is the corresponding value of the experimental RTD curve.

We used this parameter to propose a criterion for the applicability of the ADM in systems other than straight channels (see Section 4.2), for which flow maps, such as that reported in Fig. 1, are unavailable.

3. Experimental procedure

3.1. Experimental setup

The experimental setup is pictured in Fig. 2. 25 ml SGE glass syringes were filled with carrier fluid (deionized water) and tracer fluid (aqueous solution of Basic Blue 3, Dye content 25%, Sigma-Aldrich). With a syringe pump (Harvard Apparatus PHD 2000), the fluids were simultaneously pumped to a six-way injection HPLC valve that allows switching between the tracer and the carrier fluid flowing within the channel; one fluid was disposed of as waste, while the other was fed to the millichannel. Initially the carrier fluid flowed through the channel; at a time \( t_0 \), the feed was abruptly switched to tracer fluid, generating a step in tracer concentration. At the channel outlet, optical fibers, connected to a light source (Ocean Optics DH-2000-BAL), shone light through the channel (PTFE capillary), which passed through a flow cell. The light that passed through the fluid reached, via optical fibers, a spectrophotometer (Ocean Optics USB2000 + UV-vis-ES) whereby the light intensity was measured at small regular time intervals. As discussed in more detail in the following section, prior to RTD measurements, light absorption by the tube wall and pure water was measured and removed as background signal.

3.2. Effluent tracer and RTD measurement

The tracer concentration was measured at the channel outlet to determine the RTD. To do so, the Lambert–Beer law (Eq. (3.1)) was used. This is a linear equation, valid at low tracer concentrations, that relates concentration to light absorption. Tests were conducted at different tracer concentrations to check the validity of the law. PTFE capillaries with OD 1.6 mm and ID 0.5, 0.8 and 1.0 mm were used for calibration. The absorbance was measured in stagnant conditions and three readings were taken. A deviation from linearity was observed at tracer concentrations greater than 200 mg/l. We therefore set the tracer concentration at the channel inlet equal to 100 mg/l.

The Lambert–Beer law is valid only when light travels perpendicularly to the flow direction (Parnis and Oldham, 2013), and reads:

\[
A = -\log \left( \frac{I}{I_0} \right) = \varepsilon C
\]  

(3.1)

where \( I \) is the intensity of the light detected by the receiving sensor and \( I_0 \) is the intensity of the light emitted, \( \varepsilon \) is the extinction coefficient (a property of the medium), \( l \) is the path length of the beam of light through the material sample and \( C \) is the concentration of the tracer. The absorbance \( A \) was found as described below.

The flow cell is made of a rectangular aluminum block with four holes, one at the center of each lateral side. Two of them allow fitting the channel passing through the cell, while the other two accommodate the optical fibers. With this configuration the channel and the optical fibers are perpendicularly aligned to each other. The light beam leaving the channel via the optical fiber reached the spectrophotometer that measured its mean intensity. The light intensity depended entirely on the tracer concentration of the fluid present at the reactor outlet, because the carrier fluid and the tracer absorb light at different wavelengths. For this experiment the light intensity was averaged for wavelengths between 645 and 660 nm. The mean intensity measurements in the spectrophotometer were converted into absorbance values by using Eq. (3.1). For \( t < t_0 \), carrier fluid occupied the channel; this intensity value was taken and used as reference value. In Eq. (3.1), this value is \( I_0 \). For \( t > t_0 \), the light intensity \( I \) was measured. If the tracer concentration in the fluid that passes through the reactor outlet were zero, the intensity measurement would be approximately equal to \( I_0 \), the absorbance vanishing. For nonzero tracer concentrations, the light intensity decreases due to
absorption at the 645–660 nm wavelengths, thus increasing the absorbance. Intensity measurements were taken at very small time intervals, thus enabling the absorbance to be plotted against time. Measurements were recorded using the Ocean Optics Spectra Suite software.

A MATLAB script was used to smooth the collected tracer concentration data \( C(t) \) and reduce the noise arising from the measurements. The transformation of the experimental concentration curve \( C(t) \) to the cumulative distribution function \( F(t) \) was then achieved by normalizing the tracer concentration over the time:

\[
F(t) = \frac{C(t)}{C_{\text{max}}} \tag{3.2}
\]

Another MATLAB script was developed to differentiate the data from the first script. The RTD curve was thus produced. Differentiating Eq. (3.2) gives the experimental RTD curve:

\[
E(t) = \frac{dF(t)}{dt} \tag{3.3}
\]

3.3. **Design of HCTs and CFIs**

CFIs and HCTs were constructed using PVC tubes as supporting frame. For HCTs the support was a straight section of PVC pipe, while for CFIs the PVC pipe was cut in small sections with 45-degree edges and glued together forming a tridimensional frame. PTFE capillaries with different internal diameters were coiled on the PVC frame. A schematic representation of a 90-degree bend in a CFI is shown in Fig. 3.

The capillary was firmly secured on the supporting frame using cable ties. The axial pitch was minimized by having successive turns (tube coils or tube rings) contiguous so that the distance between successive turns equals the external diameter of the capillary.

Before presenting the design of the HCTs and CFIs employed in this work, the following dimensionless numbers need to be introduced.

- **Coil-to-tube diameter ratio**, \( \lambda \). This number is related to the effect of centrifugal force on the flow and is defined as follows:

\[
\lambda = \frac{D_s}{d_t} \tag{3.4}
\]

where \( D_s \) is the diameter of the cylindrical support and \( d_t \) is the internal diameter of the coiled tube. The inverse of the radius of a circle represents the curvature of the latter; thus, \( \lambda \) represents the ratio between the curvature of the coiled tube and that of the cylindrical support. Decreasing the value of \( \lambda \) results in a stronger centrifugal force; this is consequence of the increased curvature of the cylindrical support in relation to that of the coiled tube. The coil-to-tube diameter ratio is a crucial design parameter; however, its effect on the fluid dynamic behavior of CFIs has not been extensively investigated in the literature.

- **Dean number** \( (De = Re\lambda^{-0.5}) \). This number relates to the balance between inertial, centrifugal and viscous forces. As \( De \) becomes small (i.e., less than unity), viscous forces become dominant and the effect of the centrifugal force vanishes. There exists a critical value of \( De \) (or of \( Re \) for any fixed value of \( \lambda \)) above which the effect of secondary flow enhances the reduction of axial dispersion. In the literature, it is suggested that the minimum value of \( De \) required to significantly flatten the velocity profile of a fluid is 3 (Klutz et al., 2015).

Our focus in this work was on the effect of the parameter \( \lambda \) rather than of the number of flow inversions, which has already been investigated in the literature. Moreover, data in the literature show that the number of bends \( n_b \) strongly affects axial dispersion (Saxena and Negam, 1984). However, for \( n_b < 7 \) the difference with the RTD of a straight helix is minor. To effectively reduce axial dispersion Saxena and Negam (1984) found that coiled flow inverters should be constructed keeping constant the number of turns on each arm, \( n_b \). To investigate the effect of the curvature ratio alone, the number of bends \( n_b \) and the total number of turns \( n \) were kept constant for all CFIs constructed; in particular, we set \( n_b = 19 \) and \( n = 160 \). The value of \( n \) led to very long capillaries ranging between 9 and 19 m, depending on the diameter \( D_s \) of the particular CFI frame (Table 3). It was found by Saxena and Negam (1984) \( n_b = n/(n_b + 1) = 8 \) to be the optimal number of turns on each arm of the CFIs (in their article, Saxena and Negam used

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**Fig. 3 – Schematic of a 90-degree bend in a CFI. In each helical coil section \( n_b = 8 \).**
the parameter $R_a = 1/(n_b + 1)$, which is different from $n_b$ but related to it, being $R_a = n_b/n$. This value allowed the secondary flow to fully develop. To have sharp flow inversions, the free space on each arm of the CFI had to be minimized.

By changing both $D_s$ and $d_{s}$, we could construct CFIs and HCTs with $\lambda$ ranging between 17 and 64. The CFIs and HCTs constructed and investigated are shown in Fig. 4.

The inlet of the CFIs or HCTs was directly connected to the HPLC valve, so as to achieve a sharp step input directly at the inlet boundary. Their outlet was directly connected to the flow through cell (Fig. 2).

### 4. Results and discussion

#### 4.1. Determination of the molecular diffusion coefficient of the tracer

In this section, we report how we determined the molecular diffusion coefficient $D_m$ of the tracer in the carrier fluid employed. The exact value of $D_m$ for Basic Blue 3 is not clearly known, as only a range of values for $D_m$ is available in the literature (McKay et al., 1981).

As discussed in Section 2.1, when $Bo$ is greater than 100, the molecular diffusion coefficient $D_m$ in Eq. (2.6) can be neglected and so Eq. (2.6) reduces to Eq. (2.7), which can be rearranged as:

$$D_m = \frac{\nu^2 d_{s}^2}{192D_{ax}} \quad (4.1)$$

This equation can be used to determine the value of the molecular diffusion coefficient from RTD experiments in straight tubes according to the following procedure:

- RTDs were measured in a straight capillary with aspect ratio $L/d_s = 5000$ under different values of the Reynolds number ($Re = \rho u d_s / \mu$) by varying the fluid flow rate from 0.025 to 1 ml/min. According to Eq. (2.5), $Bo = Re Sc$; therefore, when changing $Re$, the point representing the fluid dynamic condition of our system in the flow map regime of Fig. 1 follows a vertical line.
- The RTD curves measured experimentally were fitted with the analytical solutions of the ADM (Eqs. (2.8) and (2.9)). $D_{ax}$ was the fitting parameter; thus $D_{ax}$ was calculated from the experimental results. Fig. 5 shows sample fitted curves.
- The values of the molecular diffusion $D_m$ were calculated using Eq. (4.1) and plotted against $Re$. The results are reported in Fig. 6.

We observed that when $Re$ was smaller than about 2 the calculated molecular diffusivity $D_m$ was constant (Fig. 6), being equal to $6.4 \times 10^{-10}$ m$^2$/s, while for larger values of $Re$ it increased with the latter. The increase does not have any physical basis: being an intrinsic transport property of the water-tracer system, $D_m$ is not a function of $Re$. We concluded that the correct value of $D_m$ was $6.4 \times 10^{-10}$ m$^2$/s. This value lies within the measured range of values ($3.78 \times 10^{-10}–6.63 \times 10^{-10}$ m$^2$/s) of the molecular diffusivity of Basic Blue 3 in water available in the literature (McKay et al., 1981).

Once the correct value of the coefficient of molecular diffusion was established, we could calculate the Bodenstein numbers for the experiments carried out and plot the oper-

![Fig. 4 - CFIs and HCTs constructed.](image)
Fig. 5 – Sample E-curve fitting results for (a) \( \text{Re} = 21; \ D_m = 1.80 \times 10^{-9} \text{m}^2/\text{s}; \ N_L = 0.0123 \). (b) \( \text{Re} = 1.58; \ D_m = 6.40 \times 10^{-10} \text{m}^2/\text{s}; \ N_L = 0.0026 \).

Fig. 6 – Calculated diffusion coefficient of Basic Blue 3 in water as a function of Re. Each experiment was performed three times and then the error bars shown were determined. For Re = 0.5, the error is \( \pm 1.7 \times 10^{-11} \text{m}^2/\text{s} \); the error bar is not visible.

At points on the flow regime map (refer to Fig. 7), it can be observed that the incorrect values of the operating point obtained at the lowest flow rate of 0.025 ml/min (Re = 0.5) is positioned in the middle of the region of full applicability of the ADM. Here \( B_0 = 8.29 \times 10^2 > 100 \) and so Eq. (4.1) is valid. When the flow rate is increased, the operating point moves on the vertical line \( L/d_t = 5000 \) to higher values of the Bodenstein number. At flow rates larger than 0.1 ml/min, which corresponds to \( \text{Re} = 2 \) and \( B_0 = 3.32 \times 10^3 \), the coefficient of molecular diffusion is no longer constant and becomes a function of the Reynolds number. This value of Re also identifies the threshold in Fig. 6 after which the coefficient of molecular diffusion is no longer constant. The incorrect values of \( D_m \) obtained for Reynolds numbers above this threshold value arise because the operating point is close to the line separating the intermediate region from the zone of applicability of the ADM, so that Eq. (4.1) no longer holds.

Fig. 5a shows the dimensionless RTD curves for \( \text{Re} = 21.1 \). By definition, \( E(t) = tE(t); \ u = u/\tau \) and \( \tau = L/u \), where \( L \) is the length of the tube and \( u \) is the mean fluid velocity (Levenspiel, 1999).

For \( \text{Re} = 21.1 \), the system operates at the limit of applicability of ADM (see Fig. 7) and the highest error between experiments and model is observed. According to Levenspiel (1999), the width and the height of the RTD curve are strong functions of the vessel dispersion number. In Fig. 5a, the width and the height of the fitted and experimental curves are close, and the shapes of the curves are very similar, the fit is therefore good, and we conclude that Eqs. (2.8) and (2.9) can be deemed to be applicable. Fig. 5b shows that for a lower value of the Reynolds number (Re = 1.58), the ADM fits the experimental data very closely; thus, as expected, the ADM (that is, Eqs. (2.8) and (2.9)) applies.

4.2. Criterion for applicability of the axial dispersion model in non-straight tubes

In this section we introduce a general criterion for judging whether the ADM is applicable or not in geometries different than straight pipes.

As discussed in the previous section, to use Eq. (4.1), one must perform experiments under full applicability of the dispersion model, the validity of which depends on the value of \( B_0 \). The ADM developed by Taylor and Aris is derived for straight pipes, and therefore it might be invalid for other channel geometries. In this regard, a procedure to gather information on the applicability of the axial dispersion model is described below.
Fig. 7 – Operating points on the flow regime map. Range of applicability of the ADM.

Fig. 8 – Residual error of the fitting of RTD experiments in a straight capillary with solutions of the ADM as a function of Re. Analytical solution of the ADM used: Eq. (2.8) for \( N_\lambda < 0.01; \) Eq. (2.9) for \( N_\lambda > 0.01. \) The fitting is based on the least squares method and is carried out using both equations and then, based on the value of the solutions, selecting the correct one.

We calculated the fitting errors \( \varepsilon \) at the different flow rates employed (0.025–1 ml/min) and we reported the values of \( \varepsilon \) as a function of the Reynolds number investigated (Fig. 8). From the plot, one can clearly observe that the residual error increases with Re. This is because an increase in Re leads to an increase in Bo (see Fig. 7). At high values of Bo, the system operates near the limit of applicability of ADM. As a result, the error \( \varepsilon \) increases while the system approaches this limit.

The experiments close to the boundary with the intermediate regime (Re = 21.1) generate the greatest error, while the residual error, and in turn the quality of the fitting, improves as Re decreases. For this reason, we identified \( \varepsilon_{cr} = 10^{-3} \) as the critical value of the residual error that one should use in judging the applicability of the model. If residual error is beyond that limit, axial dispersion model is deemed to be invalid.

4.3. Axial dispersion in HCTs and CFIs

The adoption of the dispersion number \( N_d \) is convenient because it allows comparing the extent of dispersion measured in CFIs and HCTs with that in straight tubes predicted by the Taylor–Aris equation for dispersion, Eq. (2.6). Written in terms of dispersion number, the equation becomes:

\[
N_{dTA} = \frac{1}{ReSc} + \frac{ReSc}{192}
\]  

(4.2)

The first term on the right hand side of Eq. (4.2) can be neglected, because it is significant only for Bo < 100 and in this work the Bodenstein number ranged between 10^4 and 10^5. Also, the Schmidt number was kept constant as deionized water and aqueous solutions of Basic Blue 3 were used as carrier fluid and tracer, respectively. The properties of the fluid were assumed to be those of water at room temperature (\( \rho = 997 \text{ kg/m}^3, \mu = 1.002 \times 10^{-3} \text{ Pa s} \)) and the diffusion coefficient of the tracer in water, measured experimentally in a straight capillary (see Section 4.1) was \( 6.40 \times 10^{-10} \text{ m}^2/\text{s} \), leading to Sc = 1677. Eq. (4.2) can then be written as follows:

\[
N_{dTA} = \frac{(Sc/192) Re = 8.7Re}{ReSc}
\]  

(4.3)

Eq. (4.3) expresses the relationship between dispersion and Reynolds numbers for straight pipes. \( N_{dTA} \) is an increasing linear function of Re for any given value of Sc.

The results of experiments on both CFIs and HCTs with different values of \( \lambda \) are reported in Figs. 9–11. The plots show the value of \( N_d \) as a function of Re. The different values of the Reynolds number are obtained by changing the flow rate between 0.05 ml/min and 5 ml/min. The corresponding values of the Dean number are calculated and displayed on a secondary axis. As a reference, the curve for straight tubes is...
Fig. 10 – Axial dispersion number as a function of Re (primary axis) and De (secondary axis) for a CFI (squares) and a HCT (circles) for $\lambda = 25$. The straight line refers to Eq. (4.3). Maximum standard deviation of $N_d$ is ±4.39. Not all error bars are visible.

Fig. 11 – Axial dispersion number as a function of Re (primary axis) and De (secondary axis) for a CFI (squares) and a HCT (circles) for $\lambda = 64$. The straight line refers to Eq. (4.3). Maximum standard deviation of $N_d$ is ±3.21. Not all error bars are visible.

also reported (Eq. (4.3)). The values of Nd were averaged over three experiments for each set of parameters. The standard deviation is shown as error bars on the data points.

For any value of the coil-to-tube diameter ratio, both HCTs and CFIs show less dispersion than straight tubes, confirming that the secondary flow plays a major role in defining the dispersion behavior. In CFIs the reduction of axial dispersion is as large as 10-fold for the higher values of the Reynolds number investigated. The dispersion number increases with the Reynolds number, but at a rate smaller than that of straight tubes. The rate appears to depend on Re, $\lambda$ and the type of configuration. The curves for CFIs increase at a smaller rate than those for HCTs, suggesting that flow inversion has a strong effect on the dispersion behavior. It can also be observed that the rate decreases with Re, revealing a different trend as compared with straight tubes, for which the correlation is linear.

As Re tends to zero, the dispersion curves for HCTs and CFIs approach the line for straight pipes. This is expected because at low values of Re the intensity of secondary flow, quantified by De, is modest and the Dean roll-cells, which are primarily responsible for the reduction in dispersion, disappear. When De decreases the dispersion behavior of HCTs and CFIs meets the one obtained for straight tubes. This point can be roughly estimated by analyzing the curves reported: it appears that all data point with De < 0.5 approach closely the curve for straight tubes. It was suggested in the literature that when De < 0.25 one can use the Taylor–Aris dispersion equation to quantify the dispersion in HCTs (Erdogan and Chatwin, 1967; Nigam and Vasudeva, 1976). This agrees with our findings.

For any value of $\lambda$, CFIs are characterized by lower dispersion compared to HCTs; this confirms the observations of Saxena and Nigam (1984). They reported that significant reduction in axial dispersion was achieved compared to HCTs. Moreover, they observed that the vessel dispersion number (NL = Dax/ul) did not depend on De, for CFIs with nb = 15, 30, 57 in the range 2 < De < 60. Our results are qualitatively different because Nd increases with De, meaning that as the secondary flow intensifies, so does the reduction on the dispersion behavior. Note that Nd and NL are qualitatively equivalent, as NL = Nd (dt/L). The importance of flow inversion in creating further radial mixing and in narrowing down the RTD lies in the change of direction of the centrifugal force, which makes the position of the Dean roll-cells vary. In the absence of secondary flow (and so of Dean roll-cells) the effect of flow inversion is expected to vanish. For De less than about 2 the curves for HCTs and CFIs overlap before approaching the curve of the straight tubes. This suggests the existence of a narrow region (0.5 < De < 2) in which the centrifugal force still reduces axial dispersion, but the Dean roll-cells are not strong enough to induce chaotic mixing at the bends. As a result, when operating in this region, HCTs and CFIs behave identically in terms of axial dispersion.

4.4. Effect of coil-to-tube diameter ratio

The effect of the coil-to-tube diameter ratio was investigated in CFIs and is shown in Fig. 12. A significant dependence of Nd on $\lambda$ can be observed in the range investigated. The dependence is stronger at larger values of Re. As the mean velocity increases (and so Re), the centrifugal force, also dependent on $\lambda$, that acts on the fluid intensifies. This results into stronger recirculation owing to Dean roll-cells and more effective flow inversion. Conversely, the coil-to-tube diameter ratio has a lower impact on the magnitude of axial dispersion in HCTs (Fig. 13). This can be explained considering that in CFIs two mechanisms contribute to the reduction of axial dispersion: Dean roll-cells in the sections of straight helical coils and change of orientation of Dean roll-cells at the bends. Consequently, for a given value of Re, when $\lambda$ decreases, the stronger secondary flow manifests both in the straight helical coils and at the bends. The second mechanism is absent in HTCs; this explains the weaker dependence of the axial dispersion number on $\lambda$. These observations suggest that $\lambda$ can be used more effectively in CFIs as a design parameter for narrowing down the RTD for a given value of Re.
where the values of $k_1(\lambda)$ and $k_2(\lambda)$ are found with a least squares method. The fitted curves are reported as dashed lines in Figs. 12 and 13. We assumed that $k_1$ and $k_2$ only depend on $\lambda$, as any other parameter was kept constant.

The choice of a power law is consistent with the curve trends. The values of $k_1$ and $k_2$ and the analytical expressions of the functions $k_1(\lambda)$ and $k_2(\lambda)$ are reported for both CFIs and HCTs in Table 2. It can be observed that the vessel dispersion number has a weaker dependence on the Reynolds number in CFIs compared to HCTs.

The proposed equation matches very well the data in the range investigated. It should be stressed that the values found are only valid for $n_b = 19$ and $n = 160$.

### 4.5. Applicability of the axial dispersion model in CFIs and HCTs

In this section, we investigated the applicability of the axial dispersion model for CFIs and HCTs. Fig. 14 shows the residual error of the ADM curves used to fit the experimental results as a function of the Reynolds number, for both structures. As discussed in Section 4.2 for straight tubes, the residual error $\epsilon$ increases with the Reynolds number, because the operating conditions of the system progressively move away from the region of full validity of the ADM (Fig. 7). A similar behavior is observed in Fig. 14. The criterion that we have proposed to judge the applicability of the ADM requires that the residual error should be lower than $\epsilon_{\text{p}} = 10^{-3}$. We considered only experiments fulfilling this criterion, as Fig. 14 confirms.

### 5. Conclusions

The main goal of this work was to investigate the dispersion characteristics of coiled flow inverters by conducting RTD experimental studies. A setup was developed to measure reliably RTDs in CFIs and, more in general, in different milli and microfluidic devices. Experiments in straight capillaries were also conducted to measure the molecular diffusion coefficient of the tracer in water and to generate a criterion for the applicability of the axial dispersion model in systems different than straight tubes.

Understanding the dependence of the principal geometrical parameters on the dispersion characteristic is crucial for the optimal design of CFIs. In this work, the effect of the coil-to-tube diameter ratio and of the Reynolds number was
The above flow investigated in HCTs and CFIs constructed using capillaries (with ID up to 1.0 mm) and PVC pipes as supporting structures. The coil-to-tube diameter ratio was found to influence the intensity of dispersion more significantly in CFIs than in HCTs. This demonstrates that flow inversion is extremely effective at narrowing down the RTD. However, it was also observed that flow inversion is active only for values of the Dean number above two. The operating value of the Dean number can be tuned by varying the value of the Reynolds number and of the coil-to-tube diameter ratio. The value of the former is often dictated by the operating conditions (flow rate and fluid properties) of the process. For a given Reynolds number, the value of the coil-to-tube diameter ratio must be as small as possible.

For CFIs to be adopted on a larger scale, it would be useful to obtain correlations relating the dispersion number to design and operating parameters. In this work, it was found that the dispersion number correlates with the Reynolds number according to a power law. This was obtained for fixed values of the Schmidt number and of numbers of bends and turns.

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Appendix A. Supplementary data

Supplementary data associated with this article can be found, in the online version, at http://dx.doi.org/10.1016/j.chemer.2017.02.011.

References


| Table 2 – Fixed values of \( k_1 \) and \( k_2 \) for both HCT and CFI. |
|-----------------------------|-----------------|-----------------|-----------------|-----------------|
| Coil-to-tube diameter ratio | \( k_1 \) | \( k_2 \) | \( k_1 \) | \( k_2 \) |
| 17                          | 0.466 | 0.740 | 1.142 | 0.281 |
| 25                          | 0.515 | 0.754 | 1.159 | 0.308 |
| 64                          | 0.551 | 0.763 | 1.438 | 0.327 |

\[
\begin{align*}
\text{for } \lambda < 1, & \quad k_1 = 1.53 \times 10^{-3} \lambda + 0.46 \\
\text{for } \lambda > 1, & \quad k_1 = 4.07 \times 10^{-3} \lambda + 0.74
\end{align*}
\]


