EFFECTIVE RADIAL DIFFUSIVITIES TO MODEL ENHANCED MASS AND HEAT TRANSFER IN LAMINAR FLOW REACTORS

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ABSTRACT – Main features of a laminar flow reactor (LFR) are streamline flow and parabolic velocity profile. Consequently, heating and cooling are difficult and there is a large spread in the residence time distribution (RTD). Enhanced heat and mass transfer can be found in coils or corrugated tubes or in the presence of wall roughness, curves, pipe fittings or mechanical vibration. Modeling these cases can be complex because of the induced secondary flow. Objective of this work is to introduce this enhanced transport in the LFR model by means of effective radial diffusivities of heat and mass. Model validation experiments were conducted in a LFR with high wall relative roughness and curves. RTD experiments were compared with model simulations of tracer dispersion to obtain the mass diffusivity. Heat transfer experiments provided the effective thermal diffusivity. Tested fluids were a glycerin/water mixture (Newtonian) and a CMC solution (pseudoplastic). Model was successfully adjusted and parameters were correlated with Reynolds number for flow.

1. INTRODUCTION

Non-isothermal laminar flow in tubes is a recurrent problem in many chemical and food processing applications, such as laminar flow reactors (LFR), tube sterilizers or tubular heat exchangers. A phenomenological model is required to represent flow, heat transfer, mass transfer and reaction in order to determine the distributions of temperature and concentration. In the case of corrugated or coiled tubes, the induced secondary flow improves mixing at low Reynolds number flow. However, modeling such systems can be rather complex and demand the use of CFD (computational fluid dynamics) tools to represent particular geometries and flow patterns (Bergles and Joshi, 1983; Barba *et al.*, 2002; Solano *et al.*, 2012; Pimenta and Campos, 2013).

The objective of this work is to present an alternative approach to model enhanced heat and mass transfer in LFR or heat exchangers. Besides coiled and corrugated tubes, heat and mass transfer augmentation are also associated with high relative wall roughness, mechanical vibration or the presence of fittings and curves. The vessel can be modeled as a straight smooth tube with developed laminar flow and the heat and mass transfer enhancement can be introduced in the model as an "effective" radial diffusivity that promotes mixing. This approach can lead to a good approximation of the heat and mass transfer in the vessel using a simpler mathematical model (Kechichian *et al.*, 2012; Dantas *et al.*, 2013). This would introduce two empirical parameters in the model to be determined through experiments: the effective radial diffusivity of heat α_{eff} and the effective radial diffusivity of mass D_{eff}. The procedure to estimate these parameters is derived in this work and a case study is presented to validate this modeling approach.

2. MATHEMATICAL MODELING

The Laminar Flow Reactor (LFR) or equivalent tubular system is modeled as a straight smooth tube with circular cross section, length L and internal radius R. Average thermophysical properties of the fluid are considered. Single phase, incompressible, inelastic and developed flow is assumed with the following power-law velocity profile:

$$v_z(r) = v_m \left(\frac{3n+1}{n+1} \right) \left[1 - \left(\frac{r}{R} \right)^{\frac{n+1}{n}} \right] \tag{1}$$

where v_m is the bulk velocity and n is the flow behavior index; which is unitary for a Newtonian fluid (Chhabra and Richardson, 2008).

The model should take into account enhanced mass and heat transfer that are associated with corrugated tubes, coils, high relative wall roughness, mechanical vibration or the presence of fittings and curves. The empirical parameters for heat transfer (α_{eff}) and mass transfer (D_{eff}) can be determined through separate heat transfer and residence time distribution (RTD) experiments, respectively. Modeling for these two cases are derived as follows and parameter determination from experimental data is explained in Section 3.

2.1. Heat Transfer Model

For the experimental determination of the effective thermal diffusivity, a uniform wall temperature will be assumed for the modeling. In this case, the energy conservation in the LFR leads to the known Graetz problem for heat transfer in tube under laminar flow:

$$v_z \frac{\partial T}{\partial z} - \alpha \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial T}{\partial r} \right) = 0 \tag{2}$$

where T is the temperature and α is the thermal diffusivity. This equation takes into account the axial advection and the radial diffusion of heat under steady state conditions. Boundary conditions are: specification of the inlet temperature T_{in}, symmetry at the center of the tube and specification of the wall temperature T_w (Chhabra and Richardson, 2008).

By introducing the power-law velocity profile from Equation 1, Equation 2 can be expressed in dimensionless form:

$$\left(\frac{3n+1}{n+1} \right) \left(1 - Y^{\frac{n+1}{n}} \right) \frac{\partial \Phi}{\partial Z} - \frac{1}{Pe_H} \frac{1}{Y} \frac{\partial}{\partial Y} \left(Y \frac{\partial \Phi}{\partial Y} \right) = 0 \tag{3}$$

where the dimensionless variables are $\Phi = (T - T_w)/(T_{in} - T_w)$, $Z = z/L$ and $Y = r/R$. Equation 3 depends on two parameters: the flow behavior index n and the modified reactor Peclet number for enhanced heat transfer:

$$Pe_H = \frac{L\, v_m}{\alpha_{eff}}\left(\frac{R}{L}\right)^2 \tag{4}$$

The mixing cup average temperature along the tube Φ_m can be calculated as:

$$\Phi_m(Z) = 2\left(\frac{3\,n+1}{n+1}\right)\int_0^1\left(1 - Y^{\frac{n+1}{n}}\right)\Phi(Z,Y)\,Y\,dY \tag{5}$$

2.2. Mass Transfer Model

Information regarding mass diffusivity in the LFR can be determined through residence time distribution (RTD) experiments. To model the dispersion of a tracer along the vessel, a transient mass conservation equation is needed:

$$\frac{\partial c_t}{\partial t} + v_z\,\frac{\partial c_t}{\partial z} - D_t\left[\frac{\partial^2 c_t}{\partial z^2} + \frac{1}{r}\frac{\partial}{\partial r}\left(r\,\frac{\partial c_t}{\partial r}\right)\right] = 0 \tag{6}$$

where $c_t(z,r,t)$ is the tracer concentration (Dantas *et al.*, 2014). A step experiment is considered, where the tracer continuously enters the vessel with $c_t = c_{t,0}$ and there is no tracer an $t = 0$ (initial condition). Boundary conditions are: symmetry at the center of the tube, no diffusion at the tube wall and uniform inlet of tracer at the tube entrance.

Equation 6 can be expressed in dimensionless form including the velocity profile (Equation 1):

$$\frac{\partial C}{\partial \theta} + \left(\frac{3\,n+1}{n+1}\right)\left(1 - Y^{\frac{n+1}{n}}\right)\frac{\partial C}{\partial Z} - \frac{1}{Pe_M}\frac{1}{Y}\frac{\partial}{\partial Y}\left(Y\,\frac{\partial C}{\partial Y}\right) = 0 \tag{7}$$

where the new dimensionless variables are $C = c_t/c_{t,0}$ and $\theta = t/\tau$. Parameter $\tau = L/v_m$ is the characteristic time of the tube (ratio between internal volume and volumetric flow rate). The mass dispersion in the vessel is expressed through the modified reactor Peclet number for enhanced mass transfer:

$$Pe_M = \frac{L\, v_m}{D_{eff}}\left(\frac{R}{L}\right)^2 \tag{8}$$

The mixing cup average concentration along the tube C_m can be calculated as:

$$C_m(Z,\theta) = 2\left(\frac{3\,n+1}{n+1}\right)\int_0^1\left(1 - Y^{\frac{n+1}{n}}\right)C(Z,Y,\theta)\,Y\,dY \tag{9}$$

3. MATERIALS AND METHODS

For the experiments a laboratory scale thermal processing unit for liquid foods was used. This unit comprises two stainless steel double-pipe heat exchangers (heating and cooling) and a holding tube, as shown in Figure 1. Each heat exchanger has four straight tubes with an effective length of 1.675 m available for heat transfer. Internal and external diameters of the inner tube are 4.0 and 6.0 mm. Internal and external diameters of the outer tube are 2.2 and 2.5 cm. The heating section is connected to a pressurized hot water circuit (H.Cidade, Brazil) and the cooling section is connected to a 5.8 kW chiller (Mecalor, Brazil). An eccentric screw pump (Netzsch, Brazil) provides fluid flow and tested flow rates were 10, 20, 30, 40 and 50 L/h. Tested fluids were a 80/20(w/w) glycerin/water mixture (Newtonian) and a 1.0%(w/w) solution of carboxymethylcellulose (CMC), which is pseudoplastic (Dantas *et al.*, 2014). Generalized Reynolds number for power law fluids in tubes was calculated according to Carezzato *et al.* (2007). Physical properties of fluids were calculated at average temperature (Dantas *et al.*, 2014).
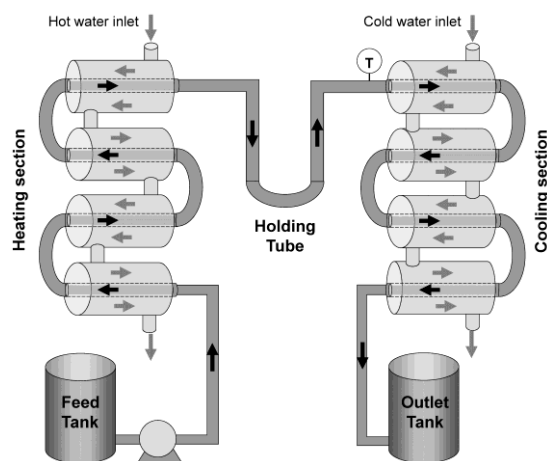

Figure 1 – Scheme of the thermal processing unit used in the experiments (Dantas *et al.*, 2014)

3.1. Heat Transfer Experiments

For the heat transfer experiments, the cooling section of the thermal processing unit was used. The flow rate of the cooling water was kept at its maximum to provide a uniform wall temperature around the internal tube of the exchanger (the velocity in the annulus was approximately 9 m/s). The temperature of the external surface of the tube was assumed equal to the average temperature of the cooling water and the internal surface temperature was calculated based on the heat load of the exchanger and the thermal resistance of the tube wall.

Temperatures at the inlet and outlet of the exchanger were measured with inserted thermocouples and a data acquisition system (National Instruments, USA). Once steady state conditions were verified, temperatures were acquired for 30 s and average values calculated.

Equation (3) was used to simulate the heat transfer experiment. Numerical solution using a finite difference discretization method was implemented in software gPROMS 3.2 (PSE, UK). The outlet average temperature of the tube $\Phi_m(Z=1)$ was specified based on the experimental data and the Peclet number Pe_H was defined as a free variable to yield zero degrees of freedom. Obtained results were correlated with Reynolds number.

3.2. Mass Transfer Experiments

For the RTD experiments, only one section of the holding tube was used with flow at room temperature (21±1 °C). This section is equivalent to one hairpin of the heat exchanger with a total length of 3.64 m and internal volume of 45.7 mL (two straight tubes and a 180° bend with 12.5 cm diameter). Injected tracer was 0.35 mL of 160 ppm methylene blue in the test fluid, which was assessed using a spectrophotometer (Femto, Brazil) at 665 nm (absorbance was under 0.1). Outlet fluid was collected every 1 or 2 s and each sample was homogenized prior to assessment.

Five experiments were conducted for each flow rate. The mean residence time and the dimensionless E-curve $E_\theta(\theta)$ were determined according to Levenspiel (1989) and Fogler (2006). The active volume of the vessel (V_{active}) was obtained from the plot of the mean residence time versus flow rate as $t_m = V_{active}/Q$. The active volume is smaller than the internal volume because of the presence of stagnation or recirculation areas, consequently, $t_m \leq \tau$.

In order to determine Pe_M for each run, the obtained experimental curve $E_\theta(\theta)$ was compared with the results from model simulation for different values of Pe_M. The plot of the sum of squared errors on $E_\theta(\theta)$ versus $\log(Pe_M)$ provided the modified Peclet with the minimum error. The adjusted values of Pe_M were correlated with Reynolds number.

The dispersion of the tracer was modeled through Equation 7, which was solved using a finite difference method in software gPROMS 3.2 (PSE, UK). Equation 9 for $Z = 1$ (end of the tube) is actually the RTD F-curve: $F = C_m(Z=1,\theta)$. Consequently, the theoretical E-curve was numerically determined as $E_\theta(\theta) = \partial F/\partial\theta$.

4. RESULTS AND DISCUSSION

Figure 2 brings simulation results of the thermal model for a Newtonian fluid ($n = 1$). The effect of the modified Peclet number Pe_H on the heat transfer can be observed. Lower values of Pe_H imply on higher radial heat dispersion and, consequently, $\Phi_m(Z=1) \to 0.0$. It was verified that pseudo-plasticity slightly improved heat transfer, but he influence of the flow behavior index n on the temperature distribution was small.
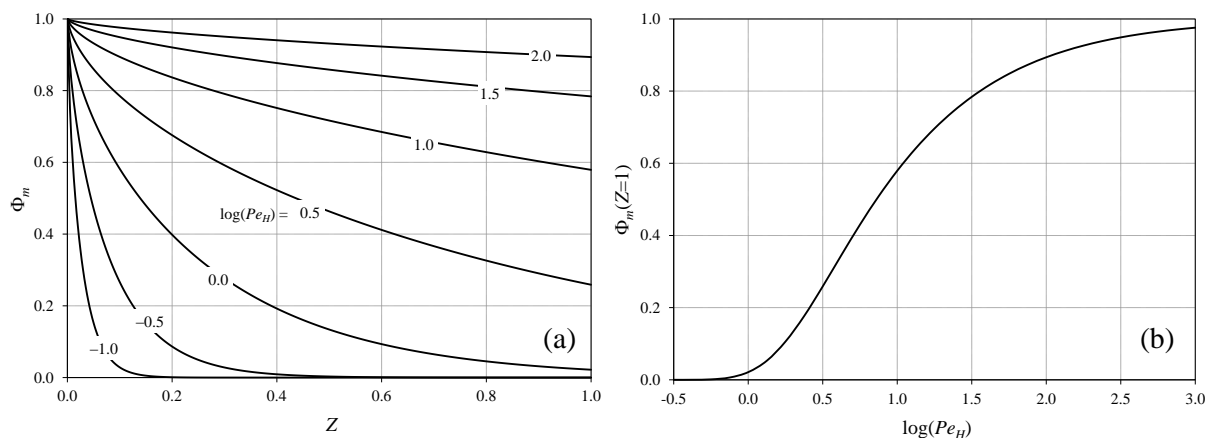

Figure 2 – Simulation results of the heat transfer model for $n = 1$ and variable Peclet number (Pe_H): a) Average temperature versus tube length (Z); b) Outlet temperature versus Pe_H.

In order to determine Pe_H from experimental data, the average temperature must be known on at least one axial position Z apart from the entrance. In this work, only the outlet temperature was measured. By specifying $\Phi_m(Z=1)$, the model simulation provided the corresponding value of Pe_H. From Equation 4, the effective radial diffusivity of heat α_{eff} can be calculated. Thus, the improvement in heat transfer can be expressed by means of a heat transfer enhancement factor F_H = α_{eff} /α. Figure 3 presents the obtained values of F_H for the experiments with the 80/20 glycerin/water mixture ($n = 1.0$) and the 1.0% CMC solution (n between 0.43 and 0.47, depending on average temperature). It can be seen that F_H approaches unity for low Reynolds number and that F_H improves with the flow rate because of the increased turbulence.

$$F_H = 0.437\ Re^{0.287}$$
$$R^2 = 0.947$$
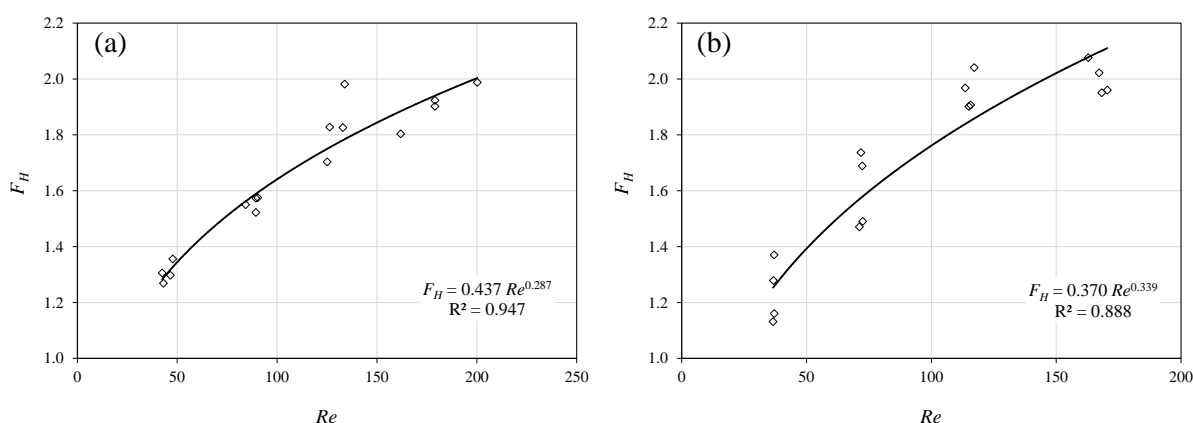
$$F_H = 0.370\ Re^{0.339}$$
$$R^2 = 0.888$$

Figure 3 – Heat transfer enhancement factor versus Reynolds number for experiments with: a) 80/20 Glycerin/water mixture; and b) 1.0% CMC solution.

In the experiments, the effective thermal diffusivity was up to two times higher than the fluid thermal diffusivity. This heat transfer enhancement can mainly be attributed to the high relative roughness of the tube wall (approximately 0.01). Moreover, the 180° bends also contributed with the fluid mixing. Considering the uncertainty in the experimental data in Figure 3, it is not possible to identity different behaviors between the two fluids.
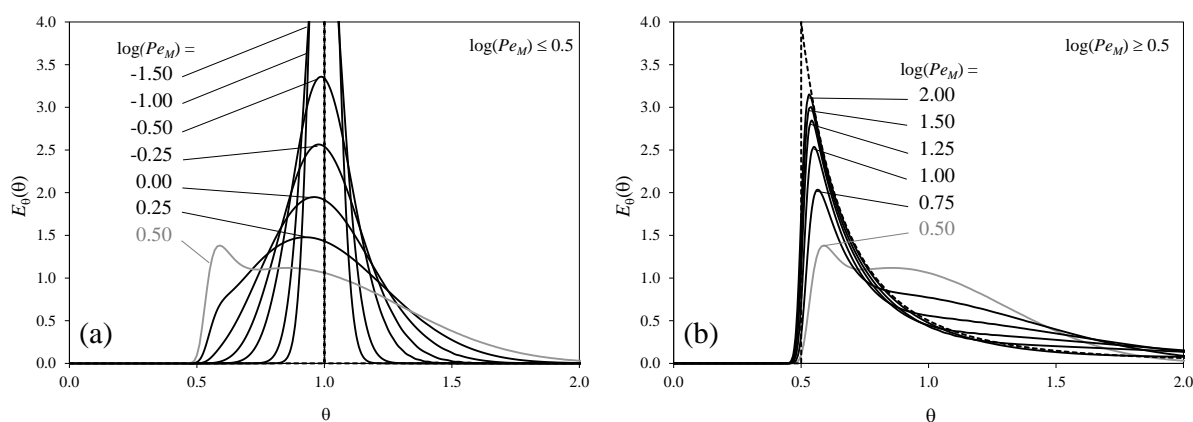
Figure 4 – Simulation results of the mass transfer model for $n = 1$ and variable Peclet number (Pe_M): a) E-curves for $\log(Pe_M) \leq 0.5$; b) E-curves for $\log(Pe_M) \geq 0.5$ (Dantas *et al.*, 2014).

Figure 4 presents simulation results of the mass transfer model for a Newtonian fluid ($n = 1$) in the form of theoretical E-curves for the dispersion of a tracer. Results were split in two figures because two distinct behaviors were observed. For $\log(Pe_M) \leq 0.5$, the E-curve shows a peak near $\theta = 1.0$ that is characteristic of turbulent flow (high dispersion). On the other hand, for $\log(Pe_M) \geq 0.5$, the E-curve shows a peak around $\theta = 0.5$ that is characteristic of laminar flow with low radial dispersion. Pseudo-plasticity narrowed the tracer dispersion because the minimum residence time was higher than $\theta = 0.5$ (Newtonian). The limiting case in Figure 4(b) (dashed line) corresponds to segregated laminar flow. For $\log(Pe_M) \approx 0.5$, the velocity profile and the radial dispersion play important roles on the tracer dispersion and the E-curve has two peaks.

The E-curves obtained from the experiments in the equipment were compared to the model simulation results and the error was minimized in order to determine the corresponding value of the modified Peclet Pe_M. Results are presented in Figure 5 for the experiments with the 80/20 glycerin/water mixture ($n = 1.0$) and the 1.0% CMC solution ($n = 0.4$ at $T = 21\ °C$) as the effective radial mass diffusivity versus Reynolds number. Considering the uncertainty on the diffusivities, a linear correlation could be adjusted and similar behavior was observed for both fluids. Average values of the Modified Peclet number were $\log(Pe_M) = 0.26$ for the glycerin/water mixture and $\log(Pe_M) = 0.52$ for the CMC solution. From Figure 4 it can be concluded that radial diffusion was important to the dispersion of the tracer for these values of Pe_M.
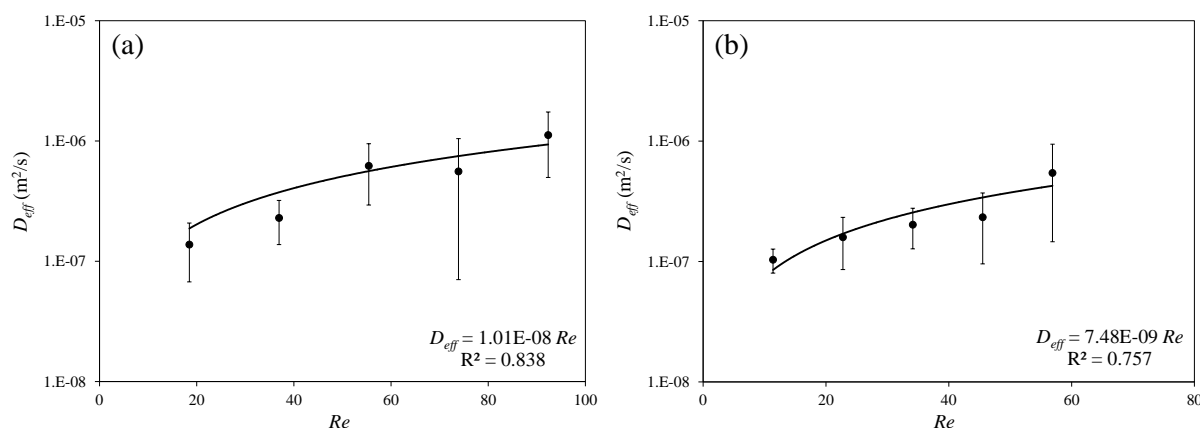

Figure 5 – Effective radial mass diffusivity versus Reynolds number for experiments with: a) 80/20 Glycerin/water mixture; and b) 1.0% CMC solution.

Another import result that was obtained from the RTD experiments is the active volume of the vessel. The adjusted values from the glycerin/water experiments and the CMC solution experiments were 91% and 89%, respectively. This indicates that approximately 10% of the internal volume of the tube is related with stagnation or recirculation zones or the presence of deposits at the tube wall. Since the equipment was used for thermal processing of liquid foods, fooling in the holding tube is a plausible explanation.

5. CONCLUSIONS

Two parameters were introduced in the Laminar Flow Reactor (LFR) model in order to simulate enhanced heat and mass transfer: the effective radial diffusivity of heat α_{eff} and the effective radial diffusivity of mass D_{eff}. Through heat transfer and RTD experiments, it was

possible to estimate these parameters for a liquid food thermal processing unit that has a high relative roughness on the tube wall. For this case study, the adjusted parameters can be applied to simulate the temperature distribution and the microbial inactivation of the process using the model developed by Kechichian *et al.* (2012). This modeling approach of the LFR provides a simple form to take into account heat and/or mass transfer enhancement in tubular laminar flow problems.

6. ACKNOWLEDGEMENTS

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