

#### 4. TUBULAR REACTORS WITH LAMINAR FLOW

Tubular reactors in which homogeneous reactions are conducted can be empty or packed conduits of various cross-sectional shape. Pipes, i.e. tubular vessels of cylindrical shape, dominate. The flow can be turbulent or laminar. The questions arise as to how to interpret the performance of tubular reactors and how to measure their departure from plug flow behavior.

We will start by considering a cylindrical pipe with fully developed laminar flow. For a Newtonian fluid the velocity profile is given by

$$u = 2\bar{u} \left( 1 - \left( \frac{r}{R} \right)^2 \right) \quad (1)$$

where  $\bar{u} = \frac{u_{max}}{2}$  is the mean velocity. By making a balance on species A, which may be a reactant or a tracer, we arrive at the following equation:

$$D \frac{\partial^2 C_A}{\partial z^2} - u \frac{\partial C_A}{\partial z} + \frac{D}{r} \frac{\partial}{\partial r} \left( r \frac{\partial C_A}{\partial r} \right) - r_{A-} = \frac{\partial C_A}{\partial t} \quad (2)$$

For an exercise this equation should be derived by making a balance on an annular cylindrical region of length  $\Delta z$ , inner radius  $r$  and outer radius  $\Delta r$ . We should render this equation dimensionless by defining:

$$\xi = \frac{z}{L}; \eta = \frac{r}{R}; \theta = \frac{t}{\bar{t}}; c = \frac{C_A}{C_{A_0}}$$

where  $L$  is the pipe length of interest,  $R$  is the pipe radius,  $\bar{t} = L / \bar{u}$  is the mean residence time,  $C_{A_0}$  is some reference concentration. Let us assume an n-th order rate form.

The above equation (2) now reads:

$$\frac{D}{\bar{u}L} \frac{\partial^2 c}{\partial \xi^2} - 2(1-\eta^2) \frac{\partial c}{\partial \xi} + \frac{D}{\bar{u}R} \left( \frac{L}{R} \right) \frac{1}{\eta} \frac{\partial}{\partial \eta} \left( \eta \frac{\partial c}{\partial \eta} \right) - k C_{A_0}^{n-1} \bar{t} c^n = \frac{\partial c}{\partial \theta} \quad (2a)$$

We define:

$$Pe_a = \frac{\bar{u}L}{D} = \frac{L^2/D}{L/\bar{u}} = \frac{\text{characteristic diffusion time (axial)}}{\text{process time (characteristic convection time)}} \quad (3)$$

= axial Peclet number.

$$Pe_r = \frac{\bar{u}R}{D} = \text{radial Peclet number} \quad (4)$$

$$\frac{\bar{u}R^2}{DL} = \frac{R^2 / D}{L / \bar{u}} = \frac{\text{characteristic radial diffusion time}}{\text{characteristic convection time}} =$$

$$Pe_r \times \left(\frac{R}{L}\right) = (\text{radial Peclet number}) \times (\text{pipe aspect ratio})$$

$$Da_n = k C_{A_0}^{n-1} \bar{t} = \frac{L / \bar{u}}{\frac{1}{k C_{A_0}^{n-1}}} = \frac{\text{process time}}{\text{characteristic reaction time}} \quad (5)$$

In terms of the above dimensionless groups which involve: characteristic reaction time, characteristic diffusion time, characteristic convection or process time and the aspect ratio, we can rewrite the above equations as:

$$Pe_a = \frac{\bar{u}L}{D} = \frac{\bar{u}d_t}{D} \cdot \frac{L}{d_t} = Pe \left(\frac{L}{d_t}\right) \quad (3a)$$

$$Pe_r = \frac{\bar{u}R}{D} = \frac{\bar{u}d_t}{D} \frac{R}{d_t} = \frac{Pe}{2} \quad (4a)$$

$$\begin{aligned} & \underbrace{\frac{1}{Pe_a}}_{\frac{1}{Pe} \left(\frac{d_t}{L}\right)} \frac{\partial^2 c}{\partial \xi^2} - 2(1-\eta^2) \frac{\partial c}{\partial \xi} + \underbrace{\frac{1}{Pe_r} \left(\frac{L}{R}\right)}_{\frac{1}{Pe} \left(\frac{4L}{d_t}\right)} \frac{1}{\eta} \frac{\partial}{\partial \eta} \left( \eta \frac{\partial c}{\partial \xi} \right) - Da_n c^n = \frac{\partial c}{\partial \xi} \\ & \underbrace{\frac{1}{Pe_a}}_{\frac{1}{Pe} \left(\frac{d_t}{L}\right)} \frac{\partial^2 c}{\partial \xi^2} - 2(1-\eta^2) \frac{\partial c}{\partial \xi} + \underbrace{\frac{1}{Pe_r} \left(\frac{L}{R}\right)}_{\frac{1}{Pe} \left(\frac{4L}{d_t}\right)} \frac{1}{\eta} \frac{\partial}{\partial \eta} \left( \eta \frac{\partial c}{\partial \xi} \right) - Da_n c^n = \frac{\partial c}{\partial \theta} \end{aligned} \quad (2b)$$

where  $Pe = \frac{\bar{u}d_t}{D} = Re \cdot Sc$  is the Peclet number for the flow. In terms of  $Pe_a$  and  $Pe_r$  we have:

$$\frac{1}{Pe_a} \frac{\partial^2 c}{\partial \xi^2} - 2(1-\eta^2) \frac{\partial c}{\partial \xi} + \frac{1}{Pe_r} \left(\frac{L}{R}\right) \frac{1}{\eta} \frac{\partial}{\partial \eta} \left( \eta \frac{\partial c}{\partial \eta} \right) - Da_n c^n = \frac{\partial c}{\partial \theta} \quad (2c)$$

The above equation can be simplified if we deal with a steady state reactor problem for which  $\frac{\partial c}{\partial \theta} = 0$ , or if we deal with a nonreactive tracer dynamic response, for which  $Da_n = 0$ . In either

event we need to solve a cumbersome partial differential equation and need two boundary conditions in axial coordinate  $\xi$  and two in radial coordinate  $\eta$ .

#### 4.1 Segregated Flow Model

The question arises whether we really need to solve the above equation (2c) numerically at all times or whether we can find simple solutions which are valid under certain conditions. Since reactant or tracer dimensionless concentration,  $c$ , is a smooth function, based on physical arguments, the value of the function and its derivatives is of similar order of magnitude except perhaps at a finite number of points. It can be shown then that when  $Pe_a \gg 1$  and  $Pe_r \frac{R}{L} \gg 1$  the first and third term of eq (2c) can be neglected. For a steady state reactor problem this results in the following equation:

$$-2(1-\eta^2)\frac{\partial c}{\partial \xi} - Da_n c^n = 0 \quad (6)$$

$$\xi = 0 \quad ; \quad c = 1 \quad (7)$$

The exit concentration  $c(\xi = 1, \eta)$  is a function of radial position, i.e of the stream line on which the reactant travels. The overall average exit concentration, or mixing cup concentration, is obtained as

$$\bar{c}_{ex} = \frac{\bar{C}_{A_{exit}}}{C_{A_0}} = \frac{\int_0^R 2\pi r u C_A(z=L, r) dr}{\int_0^R 2\pi r u C_{A_0} dr} = \frac{4\pi \int_0^R r \bar{u} \left(1 - \left(\frac{r}{R}\right)^2\right) C_A(L, r) dr}{\pi R^2 \bar{u} C_{A_0}}$$

where  $u = 2\bar{u} \left(1 - \left(\frac{r}{R}\right)^2\right)$ . Using dimensionless variables we get:

$$\bar{c}_{ex} = 4 \int_0^1 \eta (1 - \eta^2) c(1, \eta) d\eta \quad (8)$$

For a first order reaction ( $n = 1$ ) we readily find from eqs. (6) and (7) that

$$c(\xi = 1, \eta) = e^{-\frac{Da}{2(1-\eta^2)}} \quad (9)$$

Using eqn (9) in eqn (8) we obtain the exit mixing cup concentration:

$$\bar{c}_{ex} = 4 \int_0^1 \eta (1 - \eta^2) e^{-\frac{Da}{2(1-\eta^2)}} d\eta \quad (10)$$

Change variables to  $\frac{1}{1-\eta^2} = u$  ;  $\frac{2\eta d\eta}{(1-\eta^2)^2} = du$  to get

$$\bar{c}_{ex} = 4 \int_1^\infty \frac{\eta(1-\eta^2)^3}{2\eta} e^{-\frac{Da}{2}u} du = 2 \int_1^\infty \frac{e^{-\frac{Da}{2}u}}{u^3} du = 2 E_3\left(\frac{Da}{2}\right) \quad (10a)$$

where  $E_n(x) = \int_1^\infty \frac{e^{-xu}}{u^n} du$  is the n-th exponential integral.

In contrast, the cross-sectional average concentration is:

$$\begin{aligned} \tilde{c}_{ex} &= 2 \int_0^1 \eta c(1, \eta) d\eta = 2 \int_0^1 \eta e^{-\frac{Da}{2(1-\eta^2)}} d\eta \\ &= \int_1^\infty \frac{e^{-\frac{Da}{2}u}}{u^2} du = E_2\left(\frac{Da}{2}\right) \end{aligned} \quad (11)$$

$$\tilde{c}_{ex} = E_2\left(\frac{Da}{2}\right) = e^{-\frac{Da}{2}} - \frac{Da}{2} E_1\left(\frac{Da}{2}\right) \quad (11a)$$

Thus, if we measure by an instrument the cross-sectional average concentration, and try to infer reactant conversion from it, our results may be in error since conversion is only obtainable from mixing cup (flow averaged) concentration and clearly there is a discrepancy between equation (11a) and eq (10a).. You should examine the deviation of eq (11a) compared to eq (10a) and plot it as a function of  $Da$  . The needed exponential integral are tabulated by Abramowitz and Stegun (Handbook of Mathematical Functions, Dover Publ. 1964).

Using the following relationship among exponential integrals

$$E_{n+1}(z) = \frac{1}{n} \left[ e^{-z} - z E_n(z) \right] \quad (12)$$

we get the following expression for conversion from eqn (10a)

$$1 - x_A = \bar{c}_{ex} = \left(1 - \frac{Da}{2}\right) e^{-\frac{Da}{2}} + \frac{Da^2}{4} E_1\left(\frac{Da}{2}\right) \quad (10b)$$

$$\text{where } E_1(x) = \int_1^{\infty} \frac{e^{-xu}}{u} du = \int_x^{\infty} \frac{e^{-t}}{t} dt$$

We should realize immediately, upon reflection, that by eliminating the diffusion terms in eq (2c) and in arriving at eq (6-7) we deal with the segregated flow system. Indeed, in absence of diffusional effects there is no mixing among various stream lines. The reactant that enters on a particular stream line exits on the same stream line, i.e. at the same radial position  $\eta$ , and hence is surrounded by elements of the same age as its own at all times during its journey through the reactor. Every stream line has a different residence time. The shortest residence time is experienced by the elements on the center line ( $\bar{t}/2$ ), the mean residence time ( $\bar{t}$ ) is experienced by the fluid traveling on the stream line that has the mean velocity, i.e. at  $\eta = 1/\sqrt{2} = 0.707$ , while infinite residence time is experienced by the elements on the stream line at the wall ( $\eta = 1$ ). However, since the stream line at the wall receives infinitesimal amount of new fluid the mean residence time for the system exists and is  $\bar{t}$ . We recall that for the segregated flow condition and first order reaction the exit concentration can be written as:

$$\bar{c}_{ex} = \int_0^{\infty} e^{-Da\theta} E_{\theta}(\theta) d\theta = \bar{E}_{\theta}(s) \Big|_{s=Da} \quad (13)$$

This means that we have found the Laplace transform of the exit age density function for fully developed laminar flow of Newtonian fluid in a pipe

$$L\{E_{\theta}(\theta)\} = \bar{E}_{\theta}(s) = 2 E_3\left(\frac{s}{2}\right) = \left(1 - \frac{s}{2}\right) e^{-\frac{s}{2}} + \frac{s^2}{4} E_1\left(\frac{s}{2}\right) \quad (14)$$

However, even the extensive transform pairs of Bateman (Tables of Integral Transform Vol. 1) do not show this transform.

We can, however, derive the RTD or the F function for laminar flow readily based on physical arguments. Let us imagine that we have switched from white to red fluid at the inlet at  $t = 0$ . Red fluid will appear at the outlet at  $z = L$  only starting at time  $\bar{t}/2$ . The fraction of the outflow that is younger than  $t$  is given by the fraction of the fluid which is red. This is obtained by integrating the flow from the center stream line, where the red fluid is present at the outlet from time  $\bar{t}/2$ , to the stream line at position  $r$  at which red fluid just at the outlet at time  $t$  and by dividing this flow rate by the total flow rate.

Recall that

$$t = \frac{L}{u} = \frac{L}{2\bar{u}\left(1 - \left(\frac{r}{R}\right)^2\right)}$$

By definition the F curve is given by:

$$F(t) = \frac{\int_0^r 2\pi r' u dr'}{Q} = \frac{\int_0^r 4\pi r' \bar{u} \left(1 - \left(\frac{r'}{R}\right)^2\right) dr'}{\pi R^2 \bar{u}} \quad (15)$$

$$F(t) = 4 \int_0^\eta \eta' (1 - \eta'^2) d\eta' , \text{ for } t \geq \frac{\bar{t}}{2} \quad (15a)$$

where  $t = \frac{\bar{t}}{2(1 - \eta^2)}$  or  $\eta = \left(1 - \frac{\bar{t}}{2t}\right)^{1/2}$ . Hence,

$$F(t) = 4 \left[ \frac{\eta^2}{2} - \frac{\eta^4}{4} \right] = 2\eta^2 \left[ 1 - \frac{\eta^2}{2} \right] \text{ for } t \geq \frac{\bar{t}}{2} \quad (16)$$

$$F(t) = 2 \left(1 - \frac{\bar{t}}{2t}\right) \left(1 - \frac{1 - \frac{\bar{t}}{2t}}{2}\right) = \left(1 - \frac{\bar{t}}{2t}\right) \left(1 + \frac{\bar{t}}{2t}\right) = 1 - \frac{\bar{t}^2}{4t^2}$$

$$F(t) = \begin{cases} 0 & t < \frac{\bar{t}}{2} \\ 1 - \frac{\bar{t}^2}{4t^2} & t \geq \frac{\bar{t}}{2} \end{cases} \quad (17)$$

or

$$F(t) = \left(1 - \frac{\bar{t}^2}{4t^2}\right) H\left(t - \frac{\bar{t}}{2}\right) \quad (17a)$$

and

$$F(\theta) = \left(1 - \frac{1}{4\theta^2}\right) H\left(\theta - \frac{1}{2}\right) \quad (17b)$$

The exit age density function is:

$$E(t) = \frac{dF}{dt} = \frac{\bar{t}^2}{2t^3} H\left(t - \frac{\bar{t}}{2}\right) \quad (18)$$

The dimensionless exit age density function is:

$$E_\theta(\theta) = \bar{t}E(\bar{t}\theta) = \frac{1}{2\theta^3} H\left(\theta - \frac{1}{2}\right) \quad (18a)$$

For any reaction order we can then write for segregated flow laminar flow

$$\bar{c}_{ex} = \int_0^\infty c_{batch}(\theta) \frac{1}{2\theta^3} H\left(\theta - \frac{1}{2}\right) d\theta = \int_{\frac{1}{2}}^\infty \frac{c_{batch}(\theta)}{2\theta^3} d\theta \quad (19)$$

Previous studies have shown that this model is valid when  $Pe > 1000$  and  $Pe_r \frac{R}{L} > 85$ . Recall that  $Pe_a = Pe \frac{L}{dt}$ ,  $Pe_r = \frac{Pe}{2}$ .

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### 4.1.1 Use of Segregated Flow Model in Laminar Flow

When (Anathakrishnan et al. AIChE J., 11, 1063 (1963), 12, 906 (1966), 13, 939 (1968))

$$Pe_r = \frac{\bar{u}R}{D} > 500$$

$$Pe_r \frac{R}{L} = \frac{\bar{u}R}{D} \frac{R}{L} = \frac{R^2 / D}{\bar{t}} > 85$$

convection is the only important mode of transport and the laminar flow reactor behaves as in segregated flow since diffusion effects are not felt. Conversion is then given for Newtonian fluid in a cylindrical pipe by

$$x_A = 1 - \int_{\frac{1}{2}}^{\infty} \frac{C_{batch}/C_{Ao}}{2\theta^3} d\theta = 1 - \int_{1/2}^{\infty} \frac{C_{batch}}{2\theta^3} d\theta \quad (20)$$

Several points should be made:

- i) Segregated flow is most likely to occur in polymeric systems due to low diffusivities encountered in such systems. Because such systems often behave as non-Newtonian, new  $E(\theta)$  curves based on velocity profiles for non-Newtonian fluids should be derived. Such expressions can be obtained for power law fluids, Bingham fluids, etc., and actual deviations are left for the exercises.
- ii) Since the conditions for segregated flow to hold require

$$\frac{L}{dt} < \frac{Pe_r}{170} \quad ; \quad Pe_r > 500 \quad \text{or} \quad \frac{L}{dt} < \frac{Pe}{340} \quad ; \quad Pe > 1000$$

and the conditions for laminar flow are  $Re = \frac{\bar{u}d_t}{\nu} < 2,100$ . Recall that  $Pe_r = \frac{1}{2}(Sc)(Re)$  and that we must ensure that the flow is truly fully developed before it enters the reactor section. The entrance length,  $L_e$ , is of the order  $L_e = 0.035 d_t Re$  ( $L_e \approx 0.0288 d_t Re$  also is used).

For example if  $Re = 100$  and  $Sc = 1000$ ,  $Pe_r = 50,000$ . Then  $Pe = 100,000$  and  $\frac{L}{d_t} < 294$  while  $L_e = 3.5 d_t$ . If actual reactor length  $L = 250 d_t$  the conditions for segregated flow are satisfied and the entry length represents only 1.4% of the total length and can be neglected. However, if  $Re = 1,000$  and  $Sc = 1,000$ ,  $Pe_r = 500,000$  and  $L/d_t < 2940$  while  $L_e \approx 35d_t$  if



$L = 250 d_t$  segregated flow conditions are satisfied but now the entry length is 14% of the total length and might not be negligible any more in its effect. Then the entry length problem, i.e the region of developing laminar profile needs full attention. However, the model is now valid for  $L = 2500 d_t$  and entry length effects are now negligible.

iii) Often even in laminar flow it is important to create a narrow exit age density function or a steeper RTD (F curve) so that deviations from plug flow are minimized and plug flow performance approached. Narrower RTD's are useful in certain type of consecutive reactions where intermediates are the desired product, and when it is necessary to prevent undesired reactions of large reaction times to occur in a fluid with residence times much larger than the mean. Narrower E curves or, equivalently, sharper F or W curves can be obtained by using

- parallel plates configurations
- annular flow
- helical coiled pipes
- static mixers

For example, the fully developed velocity profile for annular flow is:

$$u(\eta) = \frac{2\bar{u}(1-\rho^2)\ell n\rho}{(1-\rho^4)\ell n\rho + (1-\rho^2)^2} \left[ 1 - \eta^2 - \frac{1-\rho}{\ell n\rho} \ell n\eta \right] \quad (21)$$

$$\eta = \frac{r}{R} \quad ; \quad \rho = \frac{R_{in}}{R_{out}}$$

Then

$$F(t) = 1 - W(t) = \int_{\eta_1}^{\eta_2} \eta \frac{u}{\bar{u}} d\eta \quad ; \quad \text{for } t \geq t_{min}$$

$$F(t) = \frac{2(1-\rho^2)\ell n\rho}{(1-\rho^4)\ell n\rho + (1-\rho^2)^2} \left[ \frac{\eta^2}{2} - \frac{\eta^4}{4} - \frac{1-\rho^2}{\ell n\rho} \left( \frac{\eta^2}{2} \ell n\eta - \frac{\eta^2}{4} \right) \right]_{\eta_1}^{\eta_2} \quad (22)$$

where  $\eta_1, \eta_2$  are given by the solution of the following transcendental equation

$$t = \frac{\bar{t} \left[ (1 - \rho^4) \ell n \rho + (1 - \rho^2)^2 \right]}{2(1 - \rho^2) \ell n \rho \left[ 1 - \eta^2 - \frac{1 - \rho^2}{\ell n \rho} \ell n \eta \right]} \quad (23)$$

Thus,  $F(t)$  must be evaluated numerically. The maximum velocity occurs at

$$\eta_{max} = \left( \frac{1 - \rho^2}{\ell n(1 / \rho^2)} \right)^{1/2} \quad (24)$$

and the earliest appearance time is at

$$t_{min} = \frac{\bar{t} \left[ (1 - \rho^4) \ell n \rho + (1 - \rho^2)^2 \right]}{2(1 - \rho^2) \ell n \rho \left[ 1 - \eta_{max}^2 - \frac{1 - \rho^2}{\ell n \rho} \ell n \eta_{max} \right]} \quad (25)$$

The sketches of the washout curve for a number of cases are given in the attached figure taken from Nauman. Clearly, as  $\rho$  is increased one departs more and more from the circular tube  $W(t)$  curve and approaches that for flow among parallel plates which is closer to plug flow. Remarkably, adding even a thin wire in the center of the pipe such as  $R_{wire} / R_{pipe} = \frac{1}{1000} = \rho$  makes the RTD much closer to that of plug flow!

For a helical coil, the solution for  $F(t)$  is lengthy, complex and numerical. However, a good approximation is obtained by

$$W(t) = \begin{cases} \frac{0.2010}{(t/\bar{t})^{2.84}} & t < 0.613 \bar{t} \\ \frac{0.07244}{(t/\bar{t})^2} & t > 0.613 \bar{t} \end{cases} \quad (26)$$

A single screw extruder also gives a rather narrow E curve with  $t_{min} = 0.75 \bar{t}$ . For details and references consult Nauman and Buffham (Mixing in Continuous Flow Systems, Wiley 1983). Static mixers also create a narrower E curve and approximate analysis has been presented by Nauman.

- iv) The final point to be made is that in laminar segregated flow in order to properly interpret tracer information the tracer must be injected at the injection plane proportionally to flow

at each point, and at the exit the mixing cup concentration must be measured (mean flow concentration).

Mathematically, if  $i(r,t)$  ( $\frac{g}{cm^2s}$ ) is the local flux density of the tracer at position  $\vec{r}$  of the injection plane, and  $v(r)$  is the normal velocity at point  $\vec{r}$  of the injection plane, proper flow tagging requires

$$i(r,t) = c(t) v(r) \text{ for all } \vec{r}. \quad (27)$$

For a step input this means

$$i_s(r,t) = C_o v(r) \quad (27a)$$

Mixing cup, or mean flow concentration is:

$$\bar{c} = \frac{\int_A v(r) c(r,t) dA}{\int_A v(r) dA} \quad (28)$$

where  $c(r,t)$  is the concentration at the exit plane,  $v(r)$  is the velocity normal to the exit plane at point  $\vec{r}$  and  $A$  is the cross-sectional area of the exit plane.

If however one either uses cross sectional area tagging

$$i(r,t) = i_o(t) \quad (29)$$

which for a step input is

$$i_s(r,t) = i_o \quad (29a)$$

or monitors cross-sectional average concentration

$$\tilde{c} = \frac{\int c(r,t) dA}{A} \quad (30)$$

erroneous results in terms of interpreting a step tracer response as an F curve are obtained as discussed below.

### 4.1.2 Limitations on the Tracer Method in Laminar Flow

Let us define the following quantities:

$v(\vec{r}, t)$  = velocity of the moving fluid perpendicular to the cross-sectional area  $S$ ,  $v(\vec{r}, t) = \vec{v} \cdot \vec{s}$

Indicator flux density (i.e flux) per unit area  $i(\vec{r}, t)$

$$i(\vec{r}, t) = c(\vec{r}, t)v(\vec{r}, t) \quad (31)$$

Amount of indicator collected at the outflow between  $t_o$  and  $t_o + T$ ,  $I$

$$I = \int_{t_o}^{t_o + T} \int_S i(\vec{r}, t) dA dt = \int_{t_o}^{t_o + T} \int_S c(\vec{r}, t) v(\vec{r}, t) dA dt \quad (32)$$

The flow of indicator across  $S$  at time  $t$  is

$$\int_S c(\vec{r}, t)v(\vec{r}, t) dA = \bar{c}(t)Q(t)$$

Mean flow concentration  $\bar{c}(t)$ :

$$\bar{c}(t) = \frac{\int_S c(\vec{r}, t)v(\vec{r}, t) dA}{\int_S v(\vec{r}, t) dA} = \frac{\int_S i(\vec{r}, t) dA}{Q(t)} \quad (33)$$

Now

$$I = \int_{t_o}^{t_o + T} \bar{c}(t)Q(t) dt \quad (34)$$

where  $Q(t) = \int_S v(\vec{r}, t) dA$  is the volumetric flow rate.

In the time interval  $(t_o, t_o + T)$  the mean flow concentration is:

$$\bar{c} = \frac{\int_{t_o}^{t_o + T} \bar{c}(t) Q(t) dt}{\int_{t_o}^{t_o + T} Q(t) dt} \quad (35)$$

Mean flow is

$$\bar{Q} = \frac{1}{T} \int_{t_o}^{t_o + T} Q(t) dt \quad (36)$$

In steady state flow  $\bar{Q} = Q = \text{const.}$

**Mean Cross-sectional Concentration**  $\tilde{c}(t)$ :

$$\tilde{c}(t) = \frac{\int_S c(\vec{r}, t) dA}{\int_S dA} \quad (37)$$

**Two Ways of Injecting Tracer into Steady Flow:**

**Flow tagging** -during a time interval the indicator is injected at (or flows through) the cross section ( $z = 0$ ) in such a way that for any time  $t$  in this interval

$$i(\vec{r}, t) = \mu(t)v(\vec{r}) \quad (38)$$

For all  $\vec{r}$  in the cross section

$$c(\vec{r}, t) = \mu(t) \quad (38a)$$

The above injection is proportional to flow, hence, the name flow tagging.

**Cross-sectional tagging** - the indicator is injected at a rate  $\lambda(t)$  uniform on the cross-section  $y = 0$  i.e if for very time  $t$  in the interval in question

$$i(\vec{r}, t) = \lambda(t). \quad (39)$$

Then for any  $\bar{r}$  at  $z = 0$ ,  $c(\bar{r}, t) = \frac{\lambda(t)}{v(\bar{r})}$  (39a)

In fully developed Newtonian laminar flow the velocity profile in a cylindrical tube is

$$\begin{aligned} v(r) &= \frac{2Q}{\pi R^4}(R^2 - r^2) = \frac{2\bar{u}}{R^2}(R^2 - r^2) = 2\bar{u}\left(1 - \left(\frac{r}{R}\right)^2\right) \\ &= u_o\left(1 - \left(\frac{r}{R}\right)^2\right) \quad 0 \leq r \leq R \quad \text{with } u_o = \frac{2Q}{\pi R^2} \end{aligned}$$

Mean Flow Concentration at  $z = L$

$$\bar{c}(t) = \frac{2\pi}{Q} \int_0^R r c(r, t) v(r) dr \quad (40)$$

Mean Cross-sectional Concentration at  $z = L$

$$\tilde{c}(t) = \frac{2}{R^2} \int_0^R r c(r, t) dr \quad (41)$$

For a particle with radial coordinate  $r$  that at time  $t'$  was at  $z = 0$  and at time  $t$  at  $z = L$  the following relation holds:

$$\begin{aligned} (t - t')v(r) &= L = (t - t') \frac{2Q}{\pi R^4}(R^2 - r^2) = t_{o_a} \frac{2Q}{\pi R^4} \\ t - t' &\geq t_{o_a} = \frac{L}{u_o} \\ L &= v(r)(t - t') \\ L &= t_{o_a} u_o \\ \frac{v(r)}{u_o} &= \frac{t_{o_a}}{t - t'} = 1 - \left(\frac{r}{R}\right)^2 \\ r &= R \left(1 - \frac{t_{o_a}}{t - t'}\right)^{1/2} \quad t' \leq t - t_{o_a} \end{aligned} \quad (42)$$

For a fixed  $t$  this defines  $r$  as a function of  $t'$ . Hence for  $t' = 0$

$$r = R \left(1 - \frac{t_{o_a}}{t}\right)^{1/2} \quad (42a)$$

$$dr = \frac{R}{2} \left(1 - \frac{t_{o_a}}{t}\right)^{-1/2} t_{o_a} t^{-2} \quad (42b)$$

Consider now various combinations of tagging at the injection plane and concentration monitoring at the exit plane.

a) Flow tagging step input, mean flow concentration at sampling site,  $c(z = 0, t) = \mu(t) = \mu H(t)$

$$\bar{c}(t) = \begin{cases} 0 & 0 < t < t_{o_a} \\ \frac{2\pi}{Q} \int_0^{R\left(1-\frac{t_{o_a}}{t}\right)^{1/2}} \mu r v(r) dr = \mu \left(1 - \frac{t_{o_a}^2}{t^2}\right) & \text{for } t \geq t_{o_a} \end{cases} \quad (43)$$

$$E(t) = \frac{d}{dt} \left( \frac{\bar{c}(t)}{\mu} \right) = 2 t_{o_a}^2 t^{-3} \quad t \geq t_{o_a} \quad (44)$$

$$\int_{t_{o_a}}^{\infty} E(t) dt = 2 t_{o_a}^2 \int_{t_{o_a}}^{\infty} t^{-3} dt = -2 t_{o_a}^2 \frac{t^{-2}}{2} \Big|_{t_{o_a}}^{\infty} = 1 \quad (45)$$

$$\bar{t} = \int_{t_{o_a}}^{\infty} t E(t) dt = 2 t_{o_a}^2 \int_{t_{o_a}}^{\infty} t^{-2} dt = 2 t_{o_a}^2 t^{-1} \Big|_{t_{o_a}}^{\infty} = 2 t_{o_a} \quad (46)$$

Indeed an E curve is obtained since both the mass balance, i.e zeroth moment, and the central volume principle, i.e first moment, are satisfied.

b) Flow tagging step input, mean cross-sectional concentration at  $z = L$ ,  $c(y = 0, t) = \mu H(t)$

$$\tilde{c}(t) = \frac{2}{R^2} \int_0^{R\left(1-\frac{t_{o_a}}{t}\right)^{1/2}} r \mu dr$$

$$\tilde{c}(t) = \mu \left(1 - \frac{t_{o_a}}{t}\right) \quad t \geq t_{o_a} \quad (47)$$

$$\tilde{E}(t) = \frac{d(\tilde{c}/\mu)}{dt} = \frac{t_{o_a}}{t^2} \quad \text{for } t \geq t_{o_a} \quad (48)$$

$$\int_{t_{o_a}}^{\infty} \tilde{E}(t) dt = t_{o_a} \int_{t_{o_a}}^{\infty} t^{-2} dt = -t_{o_a} t^{-1} \Big|_{t_{o_a}}^{\infty} = 1 \quad (49)$$

$$\bar{t} = \int_{t_{o_a}}^{\infty} t \tilde{E}(t) dt = t_{o_a} \int_{t_{o_a}}^{\infty} t^{-1} dt = t_{o_a} \ell n t \Big|_{t_{o_a}}^{\infty} = \infty \quad (50)$$

The obtained impulse response,  $\tilde{E}$ , clearly is not an E curve since the mean does not exist.

- c) Cross-sectional tagging step input, mean flow concentration,  $i(z = 0, t) = \lambda H(t)$

$$c(r, t) \Big|_{z=0} = \frac{\lambda}{v(r)} \quad t > 0 \quad (51)$$

$$\bar{c}(t) = \frac{2\pi}{Q} \int_0^{R \left(1 - \frac{t_{o_a}}{t}\right)^{1/2}} r c(r, t) v(r) dr = \frac{\lambda \pi R^2}{Q} \left(1 - \frac{t_{o_a}}{t}\right) \quad t \geq t_{o_a} \quad (52)$$

$$\text{Now } \bar{F}(t) = \frac{Q \bar{c}(t)}{\lambda \pi R^2}; \quad \bar{E}(t) = \frac{d\bar{F}}{dt}$$

$\bar{E}(t)$  is the same as  $\tilde{E}(t)$  in b). It cannot be an E-curve since  $\bar{t} \rightarrow \infty$ .

- d) Cross-sectional tagging step input, mean cross-sectional concentration,  $i(z = 0, t) = \lambda H(t)$

$$\tilde{c}(t) = \frac{2\lambda}{R^2} \int_0^{R \left(1 - \frac{t_{o_a}}{t}\right)^{1/2}} \frac{r}{v(r)} dr = \frac{2\lambda}{R^2} \int_0^{R \left(1 - \frac{t_{o_a}}{t}\right)^{1/2}} \frac{r \pi R^2 dr}{2Q(R^2 - r^2)}$$

$$\tilde{c}(t) = \frac{\pi \lambda R^2}{Q} \left[ -\frac{1}{2} \ell n(R^2 - r^2) \right]_0^{R \left(1 - \frac{t_{o_a}}{t}\right)^{1/2}} = \frac{\pi \lambda R^2}{2Q} \ell n \frac{t}{t_{o_a}} \quad \text{for } t \geq t_{o_a} \quad (53)$$

$$\tilde{E}(t) = \frac{1}{2} t^{-1} \quad \text{and} \quad \int_{t_{o_a}}^{\infty} E(t) dt = \frac{1}{2} \int_{t_{o_a}}^{\infty} t^{-1} dt = \frac{1}{2} \ell n t \Big|_{t_{o_a}}^{\infty} = \infty \quad (54)$$

This certainly cannot be an E-curve since the area under the curve is not finite!

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## 4.2 Taylor Diffusion and the Axial Dispersion Model

When  $Pe_r > 500$  but  $L/d_t > Pe_r/170$  the conditions for the segregated flow model are violated. Now  $Pe_a = Pe_r \cdot \frac{2L}{dt} > \frac{Pe_r^2}{85} \gg 1$  so that axial diffusion can be neglected compared to radial diffusion and convection terms. Since  $\frac{L}{d_t}$  is large, radial diffusion has sufficient time to be felt and cannot be neglected since  $\frac{R^2/D}{\bar{t}}$  is now less than 85, i.e. characteristic radial diffusion time,  $R^2/D$ , becomes more comparable to the characteristic convection time,  $\bar{t}$ .

For a reactor at steady state the following problem would have to be solved:

$$\frac{1}{Pe_r} \left( \frac{L}{R} \right) \frac{1}{\eta} \frac{\partial}{\partial \eta} \left( \eta \frac{\partial c}{\partial \eta} \right) - 2(1-\eta^2) \frac{\partial c}{\partial \xi} - Da_n c^n = 0 \quad (55)$$

while the inert tracer response is described by:

$$\frac{1}{Pe_r} \left( \frac{L}{R} \right) \frac{1}{\eta} \frac{\partial}{\partial \eta} \left( \eta \frac{\partial c}{\partial \eta} \right) - 2(1-\eta^2) \frac{\partial c}{\partial \xi} = \frac{\partial c}{\partial \theta} \quad (56)$$

Both eq (55) and (56) are subject to the appropriate boundary conditions (B.C.). Again a complex PDE needs to be solved and it would be helpful to find an approximate solution. The idea of utilizing the B.C.'s in  $\eta$  i.e.  $\eta = 0, \frac{\partial c}{\partial \eta} = 0$  and  $\eta = 1, \frac{\partial c}{\partial \eta} = 0$  by defining a cross-sectional mean concentration  $\int_0^1 \eta c d\eta$  is not immediately fruitful because of the  $(1-\eta^2)$  term multiplying  $\frac{\partial c}{\partial \xi}$ .

Some time ago G.I. Taylor made an experiment by injecting a dye into laminar flow. He observed that the slug of dye traveled together and spread out as it moved downstream rather uniformly across the tube diameter. It did not form a paraboloid of dye as expected. While stream lines close to the center tend to move the dye faster than those close to the walls, a concentration gradient develops in the lateral direction, and the dye is transported by diffusion from the centrally located stream lines to others at the leading edge of the front and from the stream lines close to the wall to centrally located ones at the trailing edge.

G.I. Taylor (Proc. Royal Society, London, A 219, 186 (1953); A 223, 446 (1954), A 224, 473 (1954)) described this behavior mathematically by fully utilizing the experimental observations.

He noticed that the centroid of the dye slug moves at the mean velocity of flow. Hence, a transformation of coordinates to a moving coordinate system at mean flow velocity is useful.

This requires:

$$\theta' = \theta, \quad \zeta = \xi - \theta$$

Thus

$$\begin{aligned} \frac{\partial}{\partial \theta} &= \frac{\partial}{\partial \theta'} \frac{\partial \theta'}{\partial \theta} + \frac{\partial}{\partial \zeta} \frac{\partial \zeta}{\partial \theta} = \frac{\partial}{\partial \theta'} - \frac{\partial}{\partial \xi} \\ \frac{\partial}{\partial \xi} &= \frac{\partial}{\partial \zeta} \frac{\partial \zeta}{\partial \xi} + \frac{\partial}{\partial \theta'} \frac{\partial \theta'}{\partial \xi} = \frac{\partial}{\partial \zeta} \end{aligned}$$

which transforms eq (56) for the tracer response to:

$$\frac{1}{Pe_r} \left( \frac{L}{R} \right) \frac{1}{\eta} \frac{\partial}{\partial \eta} \left( \eta \frac{\partial c}{\partial \eta} \right) - (1 - 2\eta^2) \frac{\partial c}{\partial \zeta} = \frac{\partial c}{\partial \theta'} \quad (57)$$

Furthermore, experimental observations indicated that the concentration at a point moving at the mean flow velocity varies extremely slowly in time, hence  $\frac{\partial c}{\partial \theta'} \approx 0$ . Finally, G.I. Taylor assumed that the axial concentration gradient is independent of radial position, again as supported by experimental observations.

With these assumptions eq (57) becomes:

$$\frac{\partial}{\partial \eta} \left( \eta \frac{\partial c}{\partial \eta} \right) = Pe_r \frac{R}{L} (\eta - 2\eta^3) \frac{\partial c}{\partial \zeta} \quad (58)$$

Integrate from  $\eta = 1; \frac{\partial c}{\partial \eta} = 0$  to  $\eta$

$$\frac{\partial c}{\partial \eta} = Pe_r \left( \frac{R}{L} \right) \left( \frac{\eta}{2} - \frac{\eta^3}{2} \right) \frac{\partial c}{\partial \zeta} \quad (59)$$

Now integrate between  $\eta = 0, \frac{\partial c}{\partial \eta} = 0$  and  $\eta$  again

$$c - c(\eta = 0) = Pe_r \left( \frac{R}{L} \right) \left( \frac{\eta^2}{4} - \frac{\eta^4}{8} \right) \frac{\partial c}{\partial \xi} \quad (60)$$

The concentration at the center line is unknown,  $c_o = c(\eta = 0)$  and should be eliminated in terms of the mean cross sectional area concentration

$$\begin{aligned} \tilde{c} &= 2 \int_0^1 \eta c \, d\eta = 2c_o \int_0^1 \eta \, d\eta + \frac{Pe_r}{2} \left( \frac{R}{L} \right) \int_0^1 \left( \eta^3 - \frac{\eta^5}{2} \right) \frac{\partial c}{\partial \xi} \, d\eta \\ \tilde{c} &= c_o + \frac{Pe_r}{2} \left( \frac{R}{L} \right) \left[ \frac{\eta^4}{4} - \frac{\eta^6}{12} \right] \frac{\partial c}{\partial \xi} = c_o + \frac{Pe_r}{12} \left( \frac{R}{L} \right) \frac{\partial c}{\partial \xi} \end{aligned} \quad (61)$$

Eliminating  $c_o$  in eq (60) using eq (61) gives:

$$c = \tilde{c} + \frac{Pe_r}{4} \left( \frac{R}{L} \right) \left[ \eta^2 - \frac{\eta^4}{2} - \frac{1}{3} \right] \frac{\partial c}{\partial \xi} \quad (62)$$

The dimensionless flux of tracer that crosses the plane that moves at the mean velocity of flow is:

$$\begin{aligned} j_{td} &= 2 \int_0^1 \eta c (1 - 2\eta^2) \, d\eta = 2 \int_0^1 (\eta - 2\eta^3) \tilde{c} \, d\eta + \\ &\quad \frac{Pe_r}{2} \left( \frac{R}{L} \right) \int_0^1 (\eta - 2\eta^3) \left( \eta^2 - \frac{\eta^4}{2} - \frac{1}{3} \right) \frac{\partial c}{\partial \xi} \, d\eta \\ j_{td} &= 2\tilde{c} \int_0^1 (\eta - 2\eta^3) \, d\eta + \frac{Pe_r}{2} \left( \frac{R}{L} \right) \frac{\partial c}{\partial \xi} \int_0^1 \left( -\frac{\eta}{3} + \frac{5}{3}\eta^5 - \frac{5}{2}\eta^5 + \eta^7 \right) \, d\eta \\ &= 2\tilde{c} \left( \frac{1}{2} - \frac{2}{4} \right) + \frac{Pe_r}{2} \left( \frac{R}{L} \right) \frac{\partial c}{\partial \xi} \left[ -\frac{1}{6} + \frac{5}{12} - \frac{5}{12} + \frac{1}{8} \right] \\ &= 0 - \frac{Pe_r}{2} \left( \frac{R}{L} \right) \frac{\partial c}{\partial \xi} \frac{1}{24} = -\frac{Pe_r}{48} \left( \frac{R}{L} \right) \frac{\partial c}{\partial \xi} \end{aligned} \quad (64)$$

Based on the previous assumptions  $\frac{\partial \tilde{c}}{\partial \xi} = \frac{\partial c}{\partial \xi}$  since  $\frac{\partial c}{\partial \xi}$  is independent of  $\eta$ . The dimensionless flux density of tracer across the plane moving at the mean velocity of flow is:

$$j_{td} = -\frac{Pe_r}{48} \left(\frac{R}{L}\right) \frac{\partial \tilde{c}}{\partial \xi}$$

This yields the following expression for the dimensional tracer flux density,  $J_t$

$$j_{td} = \frac{J_t}{\bar{u} C_o} = -\frac{Pe_r}{48} \left(\frac{R}{L}\right) \frac{\partial \tilde{c}}{\partial \xi}$$

$$J_t = -\bar{u} \frac{\bar{u} R}{48 D} \left(\frac{R}{L}\right) \frac{\partial C_t}{\partial z} = -\frac{\bar{u}^2 R^2}{48 D} \frac{\partial C_t}{\partial z} \quad (65)$$

Equation (65) for the relative tracer flux density in the axial direction with respect to the plane moving at the mean flow velocity has the form of Fick's law:

$$J_t = -D_{app} \frac{\partial C_t}{\partial z} \quad (66)$$

The apparent diffusion coefficient, called the axial dispersion coefficient,  $D_{app}$ , by comparison of eq (65) and eq (66) has the form

$$D_{app} = \frac{\bar{u}^2 R^2}{48 D} \quad (67)$$

This is the famous formula for Taylor diffusivity for laminar flow of Newtonian fluid in a circular pipe. Since the formula depends on the velocity profile, it is clear that different  $D_{app}$  is obtained for different geometries (parallel planes, rectangular cross-section, annular flow) or for Non-Newtonian fluids.

A reader who is not familiar with the above representation of the dimensionless tracer flux density with respect to the moving coordinate system should rederive eq (65) starting from the beginning.

Total tracer that passes a plane in the stagnant coordinate system per unit time is

$$\dot{m}_t = \int_0^R 2\pi r \times 2\bar{u} \left(1 - \left(\frac{r}{R}\right)^2\right) C(r,L) dr \text{ where } C = C_o \cdot c \quad (68)$$

The flux density of tracer with respect to stagnant coordinates is

$$\dot{N}_t = \frac{\dot{m}_t}{\pi R^2} = 4 \bar{u} C_o \int_0^1 \eta (1 - \eta^2) c(\eta, \xi) d\eta \quad (69)$$

Upon substitution of eq (62) into eq (69) and integration one gets:

$$\begin{aligned} \dot{N}_t &= \bar{u} C_o - \frac{\bar{u} Pe_r \left( \frac{R}{L} \right) \frac{\partial \tilde{C}}{\partial \xi}}{48} \\ &= \bar{u} C_o - \frac{\bar{u}^2 R^2}{48 D} \frac{\partial \tilde{C}}{\partial z} = \bar{u} C_o - D_{app} \frac{\partial \tilde{C}}{\partial z} \end{aligned} \quad (70)$$

The first term is a convective term and the second is the already established dispersion term. Thus, the flux density with respect to the stagnant coordinate system equals the flux with respect to the moving coordinate system plus the convective flux.

Later, Aris (Proc. Roy. Soc., A 235, 67 (1956)) showed that the apparent diffusivity or effective dispersion coefficient should take the following form:

$$D_{app} = D + \frac{\bar{u}^2 R^2}{48 D} \quad (71)$$

Clearly the molecular diffusion term is negligible when the second term is much larger. The axial dispersion coefficient,  $D_{app}$ , combines the effect of molecular diffusion and of the velocity profile. The net result is that the effects of the velocity profile and of radial diffusion can be expressed by an equivalent axial dispersion term. Eq. (56) can now be rewritten as:

$$\frac{1}{Pe_{app}} \frac{\partial^2 c}{\partial \xi^2} - \frac{\partial c}{\partial \xi} - D a_n c^n = \frac{\partial c}{\partial \theta} \quad (72)$$

which represents the axial dispersion model with

$$Pe_{app} = \frac{\bar{u} L}{D_{app}} \quad ; \quad \frac{1}{Pe_{app}} = \frac{D_{app}}{\bar{u} L} \quad (73)$$

The appropriate boundary conditions for the model require flux continuity at the entrance and at the exit. In addition, concentration must be continuous at the exit. Since inlet lines are normally of much lesser diameter than the reactor, or contain packing in order to intermix the feed, the  $D_{app}$  for these lines is usually very small and the dispersion flux can be neglected compared to the convective flux. The inlet boundary condition then is:

$$\xi = 0 \quad ; \quad c - \frac{1}{Pe_{app}} \frac{\partial c}{\partial \xi} = c_o(\theta) \quad (74)$$

At the exit

$$\xi = 1 \quad ; \quad \frac{\partial c}{\partial \xi} = 0 \quad (75)$$

The initial condition is:

$$\theta = 0 \quad ; \quad c = c^i(\xi) \quad (76)$$

where

$$c_o(\theta) = \frac{C_{inlet}(\theta)}{C_o}, \quad c^i(\xi) = \frac{C_{initial}(\xi)}{C_o}, \quad Pe_{app} = \frac{\bar{u} L}{D_{app}} \quad (77)$$

Please note that the new Peclet number, or the axial dispersion Peclet number, is defined now in terms of the apparent or effective dispersion coefficient.

$$D_{app} = D + \frac{\bar{u}^2 R^2}{48D} \quad (71)$$

In circular pipes for Newtonian fluids in laminar flow this can be expressed as:

$$Pe_{app} = \frac{192 Re Sc}{192 + Re^2 Sc^2} \cdot \left( \frac{L}{d_t} \right) \quad (78)$$

#### 4.2.1 Region of Validity

The above Taylor diffusion model with the axial dispersion coefficient given by eq (67) is valid when

- 1) The characteristic radial diffusion time < characteristic convection time
- 2) molecular axial diffusivity << axial dispersion coefficient

This implies  $Re < 2,100$  (to guarantee laminar flow),

$$\frac{L}{d_t} > 0.08 Pe_r \quad or \quad \frac{L}{d_t} > 0.08 \frac{\bar{u} R}{D} \quad (79)$$

and

$$\frac{Pe}{2} = Pe_r > 6.9 \quad (80)$$

according to G.I. Taylor.

Comparison with numerical solutions and experiments indicates that the range of applicability is more like

$$Re < 2,000 \quad ; \quad 12 \frac{L}{d_t} > Pe_r > 50 \quad (81)$$

Again for a laminar flow reactor one should check whether the entry length  $L_e$  is small compared to reactor  $L$  in order for the above model to be applicable.

Additional checks of the Taylor-Aris diffusion model against numerical solutions have been made. Wen and Fan summarize the findings in the enclosed graph which shows the applicability of various limiting cases. Presumably in the region labeled dispersion model the Taylor-Aris expression is not valid but other forms for  $D_{app}$  have to be fitted to empirical data. Other than the regimes already mentioned, there is a case of negligible convection and strict one dimensional diffusion which is not of great practical significance.

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## 4.2.2 Addenda

It is of interest to point out the following facts.

1. G.I. Taylor based on his experimental evidence reasoned that "the time necessary for appreciable effects to appear owing to convective transport is long compared with the time of decay during which radial variations of concentration are reduced to a fraction of their initial value through the action of molecular diffusion".

The characteristic time for radial diffusion is obtained by solving

$$\frac{\partial c}{\partial \theta} = \frac{1}{Pe_r} \left( \frac{L}{R} \right) \frac{1}{\eta} \frac{\partial}{\partial \eta} \left( \eta \frac{\partial c}{\partial \eta} \right) \quad (82)$$

$$\eta = 0, \quad \eta \frac{\partial c}{\partial \eta} = 0 \quad (82a)$$

$$\eta = 1, \quad \frac{\partial c}{\partial \eta} = 0 \quad (82b)$$

The solution is:

$$c = \sum_{n=1}^{\infty} A_n e^{-\alpha_n^2 \theta} J_0 \left( \frac{\alpha_n \eta}{\left( \frac{L}{R Pe_r} \right)^{1/2}} \right) \quad (83)$$

where  $\alpha_n$  are the eigenvalues that satisfy the following equation:

$$J_1 \left( \frac{\alpha_n}{\left( \frac{L}{R Pe_r} \right)^{1/2}} \right) = 0 \quad (84)$$

If one represents the above series solution for concentration by its leading term, in anticipation of good convergence, and assumes that the dye was initially present only at the center line, then



$$c = e^{-\alpha_1^2 \theta} J_0 \left( \frac{\alpha_1 \eta}{\left( \frac{L}{R Pe_r} \right)^{1/2}} \right) \quad (85)$$

The first root of  $J_1$  is 3.83 so that

$$\frac{\alpha_1}{\left( \frac{L}{R Pe_r} \right)^{1/2}} = 3.83 \quad (86)$$

$$\alpha_1 = 3.83 \left( \frac{L}{R Pe_r} \right)^{1/2} = 3.83 \left( \frac{4L}{d_t Pe} \right)^{1/2} \quad (87)$$

By convention, the characteristic diffusion time is taken as the one when the concentration of unity at the center line has dropped to  $e^{-1}$  of its original value i.e.

$$\begin{aligned} \alpha_1^2 \theta_D &= 1 \\ \theta_D &= \frac{1}{\alpha_1^2} = \frac{1}{(3.83)^2} \frac{R}{L} Pe_r = 0.0682 \frac{R}{L} Pe_r = \frac{t_D}{\bar{t}} = 0.0682 \frac{d_t Pe}{4L} \end{aligned} \quad (88)$$

Therefore, the characteristic time for convective change must be long compared to the characteristic radial diffusion time.

$$t_c = \frac{L}{u_{max}} = \frac{L}{2\bar{u}} = \frac{\bar{t}}{2} \quad (89)$$

$$\theta_c = \frac{1}{2} \quad (89a)$$

Thus

$$\frac{1}{2} > 0.0682 \frac{R}{L} Pe_r \quad (90)$$

$$\frac{L}{d_t} > 0.0682 Pe_r \quad ; \quad \frac{L}{d_t} > 0.0341 Pe \quad (91)$$

$$14.7 \frac{L}{d_t} > Pe_r \quad ; \quad 29.4 \frac{L}{d_t} > Pe \quad (92)$$

Practice shows that the requirement  $12 \frac{L}{d_t} > Pe_r$  is sufficient ; often  $8 \frac{L}{d_t} > Pe_r$  is also sufficient.

The other condition arises from the requirement that the molecular diffusion be small compared to the Taylor diffusion effect

$$\frac{\bar{u}^2 R^2}{48 D} > D$$

$$\frac{\bar{u}^2 R^2}{D^2} > 48$$

$$Pe_r^2 > 48$$

or  $Pe_r > \sqrt{48} = 6.9 \quad Pe > 13.8$

Practice and comparison with numerical solutions show that  $Pe_r > 20$  or preferably 50 are required for the perfect match between approximate Taylor solution and data or the numerical solution of the exact equation. It is important, however, to understand how the above criteria for validity of the Taylor solution were established. The other important thing to realize is that Taylor approach provides the expression for the behavior of the cross-sectional average concentration

$$\frac{\partial \tilde{c}}{\partial \theta'} = \frac{1}{Pe} \frac{\partial^2 \tilde{c}}{\partial \xi^2} \quad (93)$$

in terms of the moving coordinate system, and not of the mixing cup concentration. Thus, interpreting the results in terms of the mixing cup concentration might be in error.

### Summary: Laminar Flow on Tubular Reactors

Convective model (Segregated Flow) is valid for

$$\frac{\bar{u} d_t}{D} > 1000 \quad \text{and} \quad \frac{L}{d_t} < \frac{1}{340} \frac{\bar{u} d_t}{D}$$

Taylor Diffusivity Model for Axial Dispersion is valid when

$$Pe_r > \sqrt{48} = 6.9 \quad Pe_r = \frac{\bar{u} R}{D}$$

$$\frac{L}{d_t} > 0.0682 Pe_r$$

$$\text{or } \frac{\bar{u} d_t}{D} > 13.8 \text{ and } \frac{L}{d_t} > 0.0341 \frac{\bar{u} d_t}{D}$$

Need at least  $L/d_t > 10$  for axial dispersion model.

Pure diffusion

$$\frac{\bar{u} L}{D} < 1 \text{ or } \frac{L}{d_t} < \frac{1}{\frac{\bar{u} d_t}{D}}$$

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