

# A Tubular Reactor of Axial Dispersion and Micromixing

Yu-Shu Chien

Department of Chemical Engineering

## Abstract

In this paper, we extend the MURM (Modified Universe Reaction Model) of a CSTR to a tubular reactor with axial dispersion. Furthermore, we use this mixing model to study the effect of the micromixing on the steady state multiplicity for an autocatalytical reaction of tubular reactor.

## 1. Introduction

The exact multiplicity criteria for autocatalytic reactions in a perfectly mixed CSTR was been published [2,3]. However, these criteria can only be applied toward the situation in which CSTR is perfect mixing. Recently, the effect of macromixing and micromixing of two unpremixed feeds on the necessary and sufficient conditions for multiplicity in a CSTR have been studied by Chien and Liou [1] by using the modified Cholett's model and Liou and Chien [4] by MURM (Modified Universal Reaction Model), respectively.

The original idea of MURM from URM (Universe Reaction Model) was developed by Miiyawaki et al. [7]. The merit of the URM includes its corresponding physical meaning, ease of use and no loss of accuracy. Furthermore, URM do not needs an initial guess of mean concentration by using the IEM (Interaction by exchange with the mean) model. However, the shortcoming of URM lies in the fact that it considers each chemical species has the same mixing time constant, therefore, the URM cannot be applied towards understanding the phenomena of steady-state multiplicity with micromixing. MURM considers that each of the chemical species having its own mixing time constant. The MURM can also be simplified to the URM once the values of the mixing time constants become the same.

On the other hand, the deviation from ideality occurs due to axial mixing caused by various factors such as velocity profile, eddy and molecular diffusion, presence of packing, etc. The axial dispersion model is a widely used approximation of flow and mixing phenomena of a tubular reactor.

This note combines MURM and axial dispersion model in a tubular reactor. Basic equations obtained are solved analytically under Danckwert' boundary condition. Our model not only considers axial dispersion but also consider the effect of incomplete mixing between two reactive feeds of **A** and **B** in a tubular reactor. By virtue of our deriving model, we find that the micromixing indeed influence the unique criteria of an isothermal autocatalytical reaction of a tubular reactor.

## 2. Mixing model

A schematic diagram of **MURM** of an axial dispersion tubular reactor with two feeds are illustrated in Fig. 1. The **MURM** was derived from **URM** by Miyawaki et al. [7] and was used to discuss that the effect of the micromixing of two unpremixed feeds on the necessary and sufficient conditions for multiplicity of an isothermal autocatalytical reaction in a CSTR [4]. Here, we extend the concept of the **MURM** to a tubular reactor with axial dispersion model.

The case is first considered in which two miscible solutions containing chemical species (labelled **A** and **B**, respectively) are mixed by turbulence and simultaneously react with each other. The two solutions are divided into several lumps of a small scale. These lumps are twisted and divided a much smaller scale by turbulence, with the solutions finally being mixed to a molecular scale and then reacted with each other.

In Fig. 1, a distribution concentration-area in a lump of our model by modified **MURM**, is assumed for each cross section in a tubular reactor. In a thickness  $dZ$  (the cross section area  $A$  is constant), the area of the cross section is divided into three regions. There are two completely segregated regions for components **A** and **B**,  $A_2A_a$  and  $B_2A_b$ , respectively, and another molecular mixed region is the combination of  $A_1A_a$  and  $B_1A_b$ . The chemical reaction is assumed to occur only in the molecular-mixed region. The degree of micromixing,  $a_m A$ , is defined as the relative area ratio of the summation of  $A_1A_a$  and  $B_1A_b$  to the cross section system. This definition is true in the light of the fact that the molecular-mixed regions are homogeneous. We define that  $a_{ma}$ ,  $a_{mb}$  and  $a_m$  are the mixing degree for components **A**, **B** and overall in the cross section area. At steady state, the relations of

$$\begin{aligned}
 A_a(A_1 + A_2) &= A_a \\
 A_b(B_1 + B_2) &= A_b \\
 A_2 &= 1 - a_{ma}, A_{11} = a_{ma} \\
 B_2 &= 1 - a_{mb}, B_{11} = a_{mb} \\
 a_m A &= A_1A_a + B_1A_b = 1 - (1 - a_{ma})A_a - (1 - a_{mb})A_b
 \end{aligned} \tag{1}$$

exist in  $dZ$  (the cross section area  $A$  is constant).

Considering Fig. 1, each one of the complete segregated regions of components **A** and **B** is decreasing with increasing the axial direction. On the contrary, the mixing degree  $a_{ma}$ ,  $a_{mb}$ , and  $a_m$  increase along with the axial direction. There are two input fluxes for component **A**, one is the chemical species **A** by convection ( $v_a A_2 A_a C_{as}$ ) and the other is the axial dispersion [ $D_a A_2 A_a (\mathcal{I}C_{as}/\mathcal{I}z)$ ], feed in completely segregated regions  $A_2 A_a$  at the position  $Z$ . ( $v_a$  is the average velocity and  $D_a$  is the axial dispersion coefficient) On the other hand, three fluxes flow out of  $A_2 A_a$  for component **A** at the position  $Z + dZ$ , one is the chemical species **A** by convection ( $v_a A_2 A_a [C_{as} + (\mathcal{I}C_{as}/\mathcal{I}z)dZ]$ ),

another is the axial dispersion ( $D_a A_2 A_a \{ \int / \int z [ C_{as} + (\int C_{as} / \int z) dz ] \}$ ) and the other is the mass diffusion rate [ $(\int V_{sa} / dt) C_{AS}$ ].

The magnitude of the diffusion rate from the segregated region ( $A_2 A_a$ ) to the molecular-mixed region ( $\mathbf{a}_m \mathbf{A}$ ) can be determined for the chemical species  $\mathbf{A}$ ,

$$\int V_{sa} / dt = -K_a A_a A_a dz \quad (2a)$$

where  $K_a$  is the inversion of the mixing time constant for the component  $\mathbf{A}$ .

Simultaneously, the fluxes for the component  $\mathbf{B}$  in the cross section  $B_2 A_b$  also have the same statements above only the component  $\mathbf{A}$  is replaced by the component  $\mathbf{B}$ .

As to the section of the molecular mixed region ( $\mathbf{a}_m \mathbf{A}$ ), there are four input fluxes for component  $\mathbf{A}$ , one is the chemical species  $\mathbf{A}$  by convection ( $v_a \mathbf{a}_m A C_{as}$ ) and another is that of the axial dispersion [ $D_a \mathbf{a}_m A (\int C_{as} / \int z)$ ], feed in  $\mathbf{a}_m \mathbf{A}$  at the position  $Z$ . The other two output fluxes are the mass diffusion rates [ $(\int V_{sa} / dt) C_{as}$ ] and [ $(\int V_{sb} / dt) C_{bs}$ ] from  $A_2 A_a$  and  $B_2 A_b$ , respectively. Note that the diffusion rates are different for chemical species  $\mathbf{A}$  and  $\mathbf{B}$  since each of the species has its own time constant and volume of the complete segregate region.

Follow the definition of ( $\int V_{sa} / dt$ ), we obtain

$$\int V_{sb} / dt = -K_b B_2 A_b dz \quad (2b)$$

where  $K_b$  is the inversion of the mixing time constant for  $\mathbf{B}$  component. The discussions that how to obtain  $K_a$  and  $K_b$  can be founded in Liou and Chien [4]. On the other hand, there are two fluxes flow out of  $\mathbf{a}_m \mathbf{A}$  for component  $\mathbf{A}$  at the position  $Z + dz$ , one is the chemical species  $\mathbf{A}$  by convection ( $v_a \mathbf{a}_m A [C_{as} + (\int C_{as} / \int z) dz]$ ), another is that of the axial dispersion ( $D_m \mathbf{a}_m A \{ \int / \int z [ C_{am} + (\int C_{am} / \int z) dz ] \}$ )

By taking the mass balance in the cross section area,  $A_2 A_a$  at steady state (the density is constant)

$$\begin{aligned} q_a (1 - \mathbf{a}_{ma}) - [q_a (1 - \mathbf{a}_{ma}) + \frac{d}{dz} q_a (1 - \mathbf{a}_{ma}) dz] - D_a \frac{d}{dz} A_a (1 - \mathbf{a}_{ma}) + \\ D_a \frac{d}{dz} [A_a (1 - \mathbf{a}_{ma}) + \frac{d}{dz} A_a (1 - \mathbf{a}_{ma}) dz] - (\int V_{sa} / dt) = 0 \end{aligned} \quad (3)$$

Based on the relation ( $V_a / V = q_a / q = A_a / A$ ) of URM (Miyawaki, 1975), we obtain

$$A_a (1 - \mathbf{a}_{ma}) = A \frac{A_a}{A} (1 - \mathbf{a}_{ma}) = A \frac{V_a}{V} (1 - \mathbf{a}_{ma}) = A \frac{q_a}{q} (1 - \mathbf{a}_{ma}) \quad (4)$$

Substituting Eqs. (2) and (4) into Eq. (3), we obtain

$$\frac{q_a}{q} \left( -v_a \frac{d}{dz} (1 - \mathbf{a}_{ma}) + D_a \frac{d^2(1 - \mathbf{a}_{ma})}{dz^2} + K_a (1 - \mathbf{a}_{ma}) \right) = 0 \quad (5)$$

By taking Danckwert's boundary condition

$$z = 0, v_a (1 - \mathbf{a}_{ma}) = v_a (1 - \mathbf{a}_{ma}) - D_a \frac{d(1 - \mathbf{a}_{ma})}{dz} \quad (6)$$

$$z = L, \frac{d(1 - \mathbf{a}_{ma})}{dz} = 0$$

and using

$$Z = z / L$$

$$v_a L / D_a = Pe_e \quad (7)$$

$$L / v_a = \mathbf{t}_a$$

Eqs. (5) and (6) are rearranged as

$$\frac{1}{Pe_e} \frac{d^2(1 - \mathbf{a}_{ma})}{dZ^2} - \frac{d}{dZ} (1 - \mathbf{a}_{ma}) + K_a \mathbf{t}_a (1 - \mathbf{a}_{ma}) = 0. \quad (8)$$

and

$$Z = 0, 1 - \mathbf{a}_{ma0} = (1 - \mathbf{a}_{ma}) - \frac{1}{Pe_e} \frac{d}{dZ} (1 - \mathbf{a}_{ma}) \quad (9)$$

$$Z = 1, \frac{d}{dZ} (1 - \mathbf{a}_{ma}) = 0$$

If we consider  $\mathbf{a}_{ma0} = 0$ , the solution of Eqs. (7) to (9) is

$$1 - \mathbf{a}_{ma} = \frac{4\underline{P}_a}{(1 + \underline{P}_a)^2 \exp[\underline{P}_a(1 + \underline{P}_a/2)] - (1 - \underline{P}_a)^2 \exp[\underline{P}_a(1 - \underline{P}_a/2)]} \quad (10)$$

where

$$P_a = (1 + 4K_a \mathbf{t}_a / Pe_e) \quad (11)$$

Consider two limiting situations

$$(1) Pe_e \rightarrow \infty$$

Eq. (10) can be reduce to

$$1 - \mathbf{a}_{ma} = \exp(-K_a \mathbf{t}_a) = A_2 \quad (12a)$$

and

$$\mathbf{a}_{ma} = 1 - \exp(-K_a \mathbf{t}_a) = A_1 \quad (12b)$$

The above results are the same as that of the tubular reactor [6].

$$(2) Pe_e \rightarrow 0$$

Eq. (10) can be reduce to

$$1 - \mathbf{a}_{ma} = 1/(1 + K_a \mathbf{t}_a) = A_2 \quad (13a)$$

and

$$\mathbf{a}_{ma} = K_a \mathbf{t}_a / (1 + K_a \mathbf{t}_a) = A_1 \quad (13b)$$

The above results are the same as that of the CSTR (Liou and Chien, 1995).

Follow the same procedures, we also get the similar results for component **B**. In additional, we take mass balance for the cross section,  $A_m \mathbf{a}_m$  at steady state to obtain

$$\frac{1}{Pe_m} \frac{d^2 \mathbf{a}_m}{dz^2} - \frac{d\mathbf{a}_m}{dz} + K_a \mathbf{t}_a \mathbf{a}_{ma} \frac{q_a}{q} + K_b \mathbf{t}_b \mathbf{a}_{mb} \frac{q_b}{q} = 0 \quad (14)$$

and

$$\begin{aligned} z = 0, \mathbf{a}_{m0} &= \mathbf{a}_m - \frac{1}{Pe_m} \frac{d\mathbf{a}_m}{dz} \\ z = 1, \frac{d\mathbf{a}}{dz} &= 0 \end{aligned} \quad (15)$$

Based on the relations ( $V_a/V = q_a/q = A_a/A$ ) and  $\mathbf{a}_m A = A_1 A_a + B_1 A_b$  of URM [7], we obtain

$\mathbf{a}_m = (q_a/q) \mathbf{a}_{ma} + (q_b/q) \mathbf{a}_{mb}$  and  $\mathbf{t} = (L/v_m) = \mathbf{t}_a = \mathbf{t}_b$  is a limitation.

We take the mass balance of component **A** in  $A_2 A_a$

$$\frac{q_a}{q} \left( D_a \frac{d^2 (1 - \mathbf{a}_{ma}) C_{as}}{dz^2} - v_a \frac{d(1 - \mathbf{a}_{ma}) C_{as}}{dz} + K_a (1 - \mathbf{a}_{ma}) C_{as} \right) = 0 \quad (16)$$

Using Eq. (5), Eq. (16) is written as

$$(1 - \mathbf{a}_{ma}) \left( D_a \frac{d^2 C_{as}}{dz^2} - v_a \frac{dC_{as}}{dz} \right) = 0 \quad (17)$$

The solution is

$$C_{as} = C_1 (-v_a/D_a) + C_2 \text{Exp}(v_a/D_a)z \quad (18)$$

One of boundary condition  $z = L, dC_{as}/dz = 0$  make  $C_2 = 0$  in Eq. (18), another boundary

condition  $z = 0, v_a (C_{a0} - C_{as}) = D_a \frac{dC_{as}}{dz}$  make  $C_1 = (-D_a/V_a) C_{a0}$  in Eq. (18). Thus, we get

$$C_{as} = C_{a0} \quad (19)$$

Similarity, we take the mass balance of component **B** in  $B_2 B_b$  to obtain

$$C_{bs} = C_{b0} \quad (20)$$

We take the mass balance of component **A** in  $\mathbf{a}_{mv} V$  region and obtain

$$D_m \frac{d^2 \mathbf{a}_m C_{am}}{dz^2} - v_m \frac{d\mathbf{a}_m C_{am}}{dz} + K_a (1 - \mathbf{a}_{ma}) C_{as} \frac{q_a}{q} + \text{rate} \mathbf{a}_m = 0 \quad (21)$$

Use dimensionless variable and Eq. (8), we obtain

$$\begin{aligned} & \mathbf{a}_m D_m \frac{d^2 C_{am}}{dz^2} - \mathbf{a}_m v_m \frac{dC_{am}}{dz} + K_a (1 - \mathbf{a}_{ma}) C_{as} \frac{q_a}{q} \\ & - \left( K_a (1 - \mathbf{a}_{ma}) \frac{q_b}{q} + K_b (1 - \mathbf{a}_{mb}) \frac{q_b}{q} \right) C_{am} + \text{rate}_a \mathbf{a}_m = 0 \end{aligned} \quad (22)$$

We use the mean concentration in the cross-section area to solve our problem.

The mass balance for component **A** in  $B_2 A_b$  region at steady state is

$$\frac{q_b}{q} \left( D_b \frac{d^2 (1 - \mathbf{a}_{mb}) 0}{dz^2} - v_b \frac{d(1 - \mathbf{a}_{mb}) 0}{dz} + K_b (1 - \mathbf{a}_{mb}) 0 \right) = 0 \quad (23)$$

If we consider  $D_a = D_b = D_m$ , we sum Eqs. (16), (21) and (23) and use

$$q \mathbf{a}_m C_{am} + q_a (1 - \mathbf{a}_{ma}) C_{as} + q_b (1 - \mathbf{a}_{mb}) 0 = q \bar{C}_a \quad (24)$$

to obtain

$$D_m \frac{d^2 \bar{C}_a}{dz^2} - v_m \frac{d\bar{C}_a}{dz} + \text{rate}_a \mathbf{a}_m = 0 \quad (25)$$

Respectively, boundary conditions of Eqs. (16), (22) and (23) are

$$z = 0, v_a \mathbf{a}_{ma} 0 C_{as0} = v_a \mathbf{a}_{ma} C_{as} - D_a \frac{d\mathbf{a}_{ma} C_{as}}{dz} \quad (26a)$$

$$z = L, \frac{d\mathbf{a}_{ma} C_{as}}{dz} = 0$$

$$z = 0, v_m \mathbf{a}_{m0} C_{am0} = v_m \mathbf{a}_m C_{am} - D_m \frac{d\mathbf{a}_m C_{am}}{dz} \quad (26b)$$

$$z = L, \frac{d\mathbf{a}_m C_{am}}{dz} = 0$$

and

$$z = 0, v_b \mathbf{a}_{mb} 0 = v_b \mathbf{a}_{mb} 0 - D_b \frac{d\mathbf{a}_{mb} 0}{dz} \quad (26c)$$

$$z = L, \frac{d\mathbf{a}_{mb} 0}{dz} = 0$$

By virtue of Eq. (24), Eq. (26) can be written as

$$z = 0, v_m \bar{C}_{a0} = v_m \bar{C}_a - D_m \frac{d\bar{C}_a}{dz} \quad (27)$$

$$z = L, \frac{d\bar{C}_a}{dz} = 0$$

Follow the same steps for component **B**, we can obtain

$$D_m \frac{d^2 \bar{C}_b}{dz^2} - v_m \frac{d\bar{C}_b}{dz} + \text{rate}_b \mathbf{a}_m = 0 \quad (28)$$

where

$$q\mathbf{a}_m C_{bm} + q_a(1 - \mathbf{a}_{ma})0 + q_b(1 - \mathbf{a}_{mb})C_{bm} = q\overline{C}_b \quad (29)$$

and

$$\begin{aligned} z = 0, v_m \overline{C}_{b0} &= v_m \overline{C}_b - D_m \frac{d\overline{C}_b}{dz} \\ z = L, \frac{d\overline{C}_b}{dz} &= 0 \end{aligned} \quad (30)$$

Thus, Eqs. (25) and (28) with boundary conditions (27) and (30) for components **A** and **B**, respectively are the design equations by combining the axial dispersion and micromixing models of a tubular reactor.

### 3. The effort of the micromixing on the sufficient condition for uniqueness autocatalytical reaction in a tubular reactor

Considering an autocatalytical reaction,  $A + B \xrightarrow{k} (h+1)B + \text{product}$

which rate expression is  $-\mathbf{g}_a = kC_a^p C_b^r$ , take place in a tubular reactor.

We define

$$\overline{Y}_a = \frac{\overline{C}_{a0} - \overline{C}_a}{\overline{C}_{a0}} \quad \text{or} \quad \overline{C}_a = \overline{C}_{a0}(1 - \overline{Y}_a) \quad (31)$$

where  $\overline{C}_{a0} = q C_{a0} / q$ . From the stoichiometry of component **A** and **B**, the relation

$$\overline{C}_b = \overline{C}_{b0} + h(\overline{C}_{a0} - \overline{C}_a) \quad (32)$$

where  $\overline{C}_{b0} = q_b C_{b0} / q$ . (33)

Thus, we apply the axial dispersion and the micromixing models to the above reaction system to obtain that the design equation for component **A** is

$$D_m \frac{d^2 \overline{C}_a}{dz^2} - v_m \frac{d\overline{C}_a}{dz} - k C_{am}^p C_{bm}^r \mathbf{a}_m = 0 \quad (34)$$

where

$$C_{am} = \overline{C}_{a0}(\mathbf{a}_{ma} - \overline{Y}_a) / \mathbf{a}_m \quad (35)$$

$$C_{bm} = (\overline{C}_{b0} \mathbf{a}_{mb} + h \overline{C}_{a0} \overline{Y}_a) / \mathbf{a}_m \quad (36)$$

From Eqs. (34), (35), (36), we get

$$\frac{1}{Pe_m} \frac{d^2 \overline{Y}_a}{dz^2} - \frac{d\overline{Y}_a}{dz} + \mathbf{q} \left( \frac{1}{\mathbf{a}_m} \right)^{p+r-1} (\mathbf{a}_{ma} - \overline{Y}_a)^p (\mathbf{l} \mathbf{a}_{mb} + \overline{Y}_a)^r = 0 \quad (37)$$

where

$$\mathbf{q} = kt \overline{C}_{a0}^{p+r-1} \quad (38)$$

$$\mathbf{l} = \overline{C}_{b0} / h \overline{C}_{a0} \quad (39)$$

The boundary conditions are

$$\begin{aligned} z = 0, \bar{Y}_a &= \frac{1}{Pe} \frac{d\bar{Y}_a}{dz} \\ z = 1, \frac{d\bar{Y}_a}{dz} &= 0 \end{aligned} \quad (40)$$

We use the method of Luss [5] and the sufficient condition for uniqueness as follows: (The detail deriving is omitted.)

$$\text{Sup}_{0 \leq \bar{Y}_a \leq \mathbf{a}_{ma}} \bar{Y}_a \frac{d \ln f(\bar{Y}_a(z))}{d\bar{Y}_a} \leq 1 \quad (41)$$

where

$$f(\bar{Y}_a) = \mathbf{q} \left( \frac{1}{\mathbf{a}_m} \right)^{p+r-1} (\mathbf{a}_m - \bar{Y}_a)^p (\mathbf{d}\mathbf{a}_{mb} + \bar{Y}_a) \quad (42)$$

We found that the micromixing indeed effect on the sufficient condition for uniqueness because  $f(\bar{Y}_a)$  depends on  $\mathbf{a}_m, \mathbf{a}_{ma}$  and  $\mathbf{a}_{mb}$ . The ideal mixing is hardly attained in practice, we hope that our mixing model is important for chemical reactor design.

## References

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## Nomenclature

- A cross section area  
 A chemical species for A



- B** chemical species for B
- C** concentrations,  $\text{mol/m}^3$
- D** axial dispersion coefficient
- k** reaction rate constant,  $(\text{mol/ m}^3)^{1-p-r}$
- $K_a$**  inversion of the mixing time constant of chemical species A,  $s^{-1}$
- $K_b$**  inversion of the mixing time constant of chemical species B,  $s^{-1}$
- Pe** peclet Number,  $(vL/D)$
- $p$**  reaction order
- $r$**  reaction order
- V** volume
- $v$**  fluid velocity
- Y** conversion
- z** the distance of the axial direction

***Greek letters***

- $a_m$**  degree of complete mixing
- $a_{ma}$**  degree of micromixing for A
- $a_{mb}$**  degree of micromixing for B
- $-g_a$**  reaction rate per unit of reactant volume in the region of complete mixing
- $l$**  define in Eq. (39)
- $h$**  stoichiometry coefficient
- $q$**  define in Eq. (38)
- $t$**  mean residence time

***Superscripts***

- $-$**  average in the cross-section area **A**

***Subscripts***

- 0** feed concentration
- 1** complete segregated region
- 2** maximum mixedness region
- $a$**  chemical species A
- $b$**  chemical species B
- $m$**  in maximum mixedness region
- $s$**  in completely segregated region



# 具有軸向混合與微觀混合之管式反應器

錢玉樹

化學工程系

## 摘 要

在本文我們延伸 CSTR 的修正單一反應模式 (Modified Universe Reaction Model) 至具有軸向混合與微觀混合之管式反應器。更進一步，我們使用上述模式研究微觀混合對於自身催化反應在管式反應器多重穩態之影響。