Dynamic Simulation and Optimization using EMSO

– Lecture 5 –

Simulation of tubular reactors.

Argimiro R. Secchi

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Solutions for Process Control and Optimization

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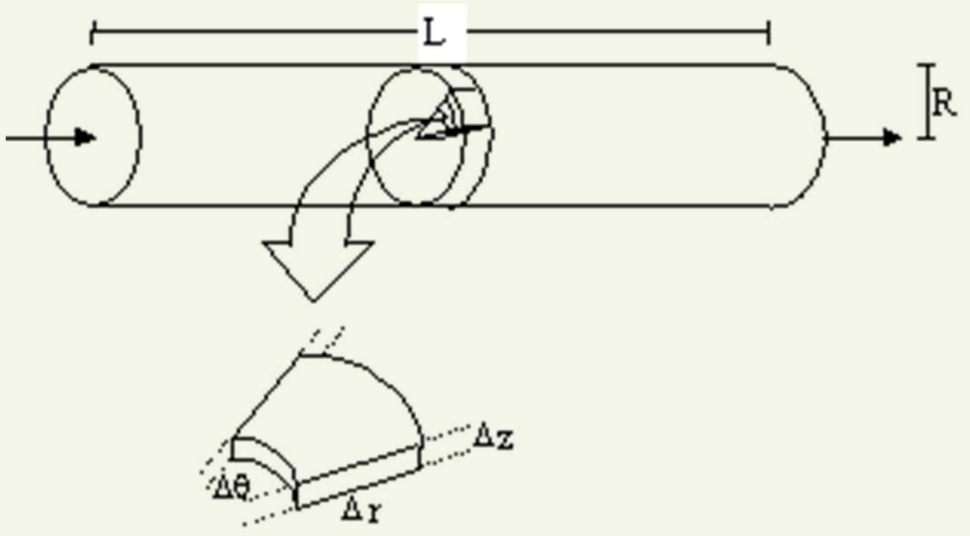
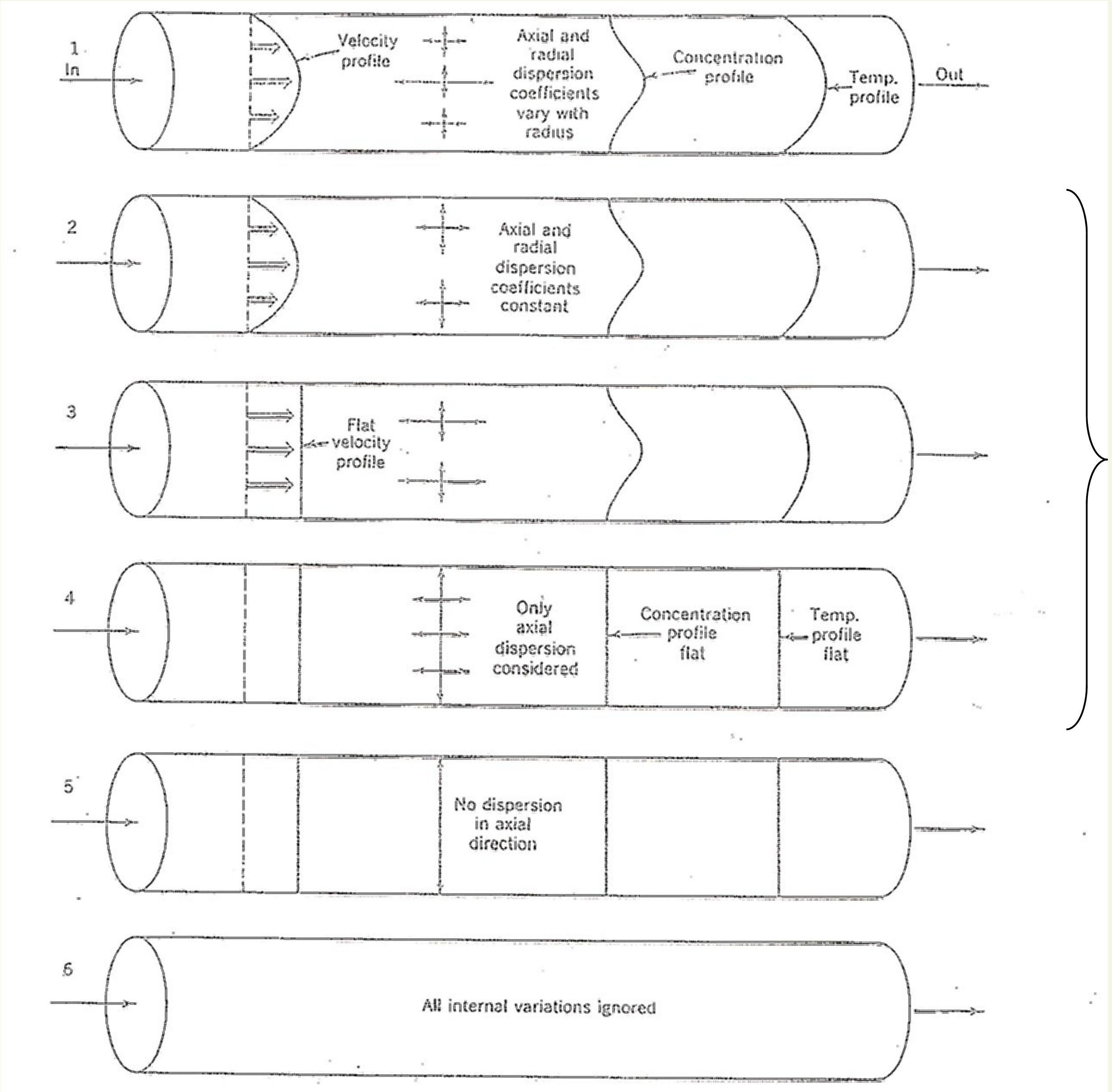
Dynamic Modeling of Tubular Reactors



Assumptions:

- Constant physical properties (ρ, μ, c_p);
- Newtonian fluid;
- Angular symmetry;
- $v_r = v_\theta = 0$

Model classification based on physical-chemistry principles



Microscopic Model

Multiple Gradients Model

Maximum Gradient Model

Macroscopic Model

Microscopic Model

$$(\nabla.\overline{v}) = 0 \quad \longrightarrow \quad \overline{v}_z = \overline{v}_z(r,t)$$

$$\frac{\partial \overline{C}_i}{\partial t} = -\overline{v}_z(r,t)\frac{\partial \overline{C}_i}{\partial z} - (\nabla \cdot \overline{C_i' v'}) + (\nabla \cdot \mathrm{D}^{(l)}\, \nabla \overline{C}_i) + R_i$$

$$\rho\, c_p \frac{\partial \overline{T}}{\partial t} = -\rho\, c_p\, \overline{v}_z(r,t)\frac{\partial \overline{T}}{\partial z} - \rho\, c_p (\nabla \cdot \overline{v' T'}) + (\nabla \cdot k^{(l)}\, \nabla \overline{T}) + (\mu^{(l)}\, \phi_v^{(l)} + \mu^{(t)} \phi_v^{(t)}) + \overline{S}_r$$

$$\rho \frac{\partial \overline{v}}{\partial t} = -\rho \overline{v} \cdot \nabla \overline{v} - \rho[\nabla \cdot \overline{v' v'}] - \nabla \overline{P} + \mu^{(l)}\, \nabla^2 \overline{v} + \rho\, g$$

Turbulence Model (simple example):

$$\overline{v' c_i'} = J_i^{(t)} = -\mathrm{D}^{(t)}\, \nabla \overline{C}_i$$

$$\rho\, c_p \overline{v' T'} = q^{(t)} = -k^{(t)} \nabla \overline{T}$$

$$\rho\, \overline{v' v'} = \tau^{(t)} = -\mu^{(t)}\, \nabla \overline{v}$$

Computational Fluid Dynamic (CFD)

Recommended Numerical Method

Finite Volume:

Consist in carry out balances of properties in elementary volumes (finite volumes), or in an equivalent form in the integration over the elementary volume of the differential equation in the conservative form (or divergent form, where the fluxes appearing in the derivatives).

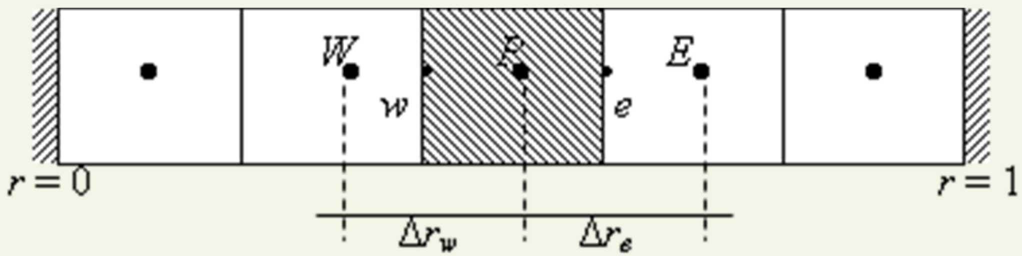
→ Use of CFD software (Computational Fluid Dynamics)

Simple example using finite-volume method:

Reaction-diffusion equations of a spherical catalytic particle

$$\begin{cases} \dfrac{\partial C}{\partial t} = \dfrac{1}{r^2}\dfrac{\partial}{\partial r}\left(r^2 \dfrac{\partial C}{\partial r}\right) - \Phi^2 C(r,t) \\ \left.\dfrac{\partial C}{\partial r}\right|_{r=0} = 0 \;;\; C(1,t) = 1 \\ C(r,0) = C_0 \end{cases}$$

First-order selections



$$dV = 4\pi r^2 dr$$

$$\int_w^e \frac{\partial C}{\partial t} r^2 dr = \int_w^e \partial\left(r^2 \frac{\partial C}{\partial r}\right) - \int_w^e \Phi^2 r^2 C\, dr$$

Simple example using finite-volume method:

$$C_p = \frac{\int_w^e C\, r^2 dr}{\int_w^e r^2 dr} = \frac{3}{\left(r_e^2 - r_w^2\right)} \int_w^e C\, r^2 dr$$

mean value

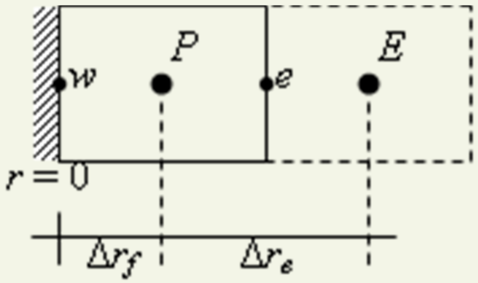
$$\frac{dC_p}{dt} = \frac{3}{\left(r_e^3 - r_w^3\right)} \left[r^2 \frac{\partial C}{\partial r} \right]_w^e - \Phi^2 C_p \qquad \left. \frac{\partial C}{\partial r} \right|_{r=r_e} = \frac{C_E - C_p}{\Delta r_e} \qquad \left. \frac{\partial C}{\partial r} \right|_{r=r_w} = \frac{C_p - C_W}{\Delta r_w}$$

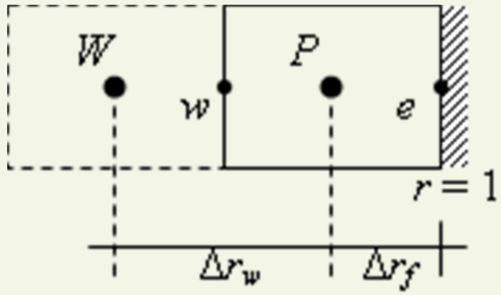
$$\frac{dC_p}{dt} = A_W C_W + A_p C_p + A_E C_E - \Phi^2 C_p \qquad p = 2, \ldots, N-1$$

$$A_W = \frac{3r_w^2}{\left(r_e^3 - r_w^3\right)\Delta r_w} \qquad A_p = \frac{-3}{\left(r_e^3 - r_w^3\right)} \left(\frac{r_e^2}{\Delta r_e} + \frac{r_w^2}{\Delta r_w} \right) \qquad A_E = \frac{3r_e^2}{\left(r_e^3 - r_w^3\right)\Delta r_e}$$

Simple example using finite-volume method:

Boundary conditions





$r_w = 0$

$r_e = 1$

$$\left.\frac{\partial C}{\partial r}\right|_{r_w} = 0$$

$$\left.\frac{\partial C}{\partial r}\right|_{r_e} = \frac{C_e - C_p}{\Delta r_f} = \frac{1 - C_p}{\Delta r_f}$$

Resulting systems: $\frac{d\underset{\sim}{C}}{dt} = A\underset{\sim}{C} + \underset{\sim}{b}$ where A is a tridiagonal matrix

Or a non-linear system for reaction of order ≠ 1: $\frac{d\underset{\sim}{C}}{dt} = \underset{\sim}{F}(\underset{\sim}{C})$

Multiple Gradients Model

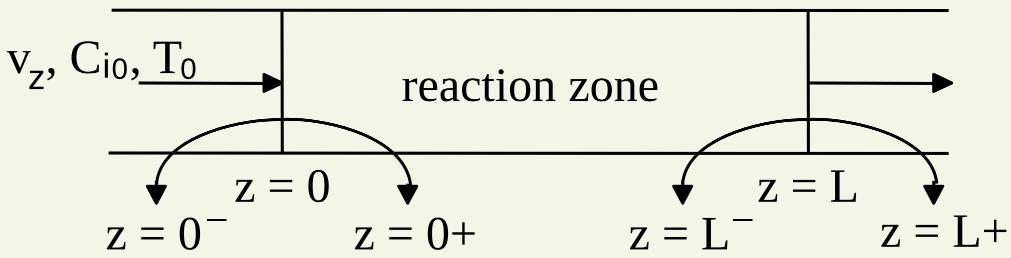
$$\bar{\mathrm{D}} = \mathrm{D}^{(t)} + \mathrm{D}^{(l)} \qquad \bar{k} = k^{(t)} + k^{(l)} \qquad \bar{\mu} = \mu^{(t)} + \mu^{(l)}$$

$$\frac{\partial \bar{C}_i}{\partial t} = -\bar{v}_z(r,t)\frac{\partial \bar{C}_i}{\partial z} + (\nabla \cdot \bar{\mathrm{D}} \nabla \bar{C}_i) + R_i$$

$$\rho c_p \frac{\partial \bar{T}}{\partial t} = -\rho c_p \bar{v}_z(r,t)\frac{\partial \bar{T}}{\partial z} + (\nabla \cdot \bar{k} \nabla \bar{T}) + \bar{\mu}\bar{\phi}_v + \bar{S}_r$$

$$\rho \frac{\partial \bar{v}_z}{\partial t} = -\nabla \bar{P} + \bar{\mu}\nabla^2 \bar{v}_z + \rho g$$

Boundary Conditions:



Component Mass Balance

(removing time average notation)

$$\frac{\partial C_i}{\partial t} = \mathrm{D}_z(r,t)\frac{\partial^2 C_i}{\partial z^2} + \frac{1}{r}\frac{\partial}{\partial r}\left(r\,\mathrm{D}_R(r,t)\frac{\partial C_i}{\partial r}\right) - v_z(r,t)\frac{\partial C_i}{\partial z} + R_i$$

$$v_z(r,t)\,C_{i0}(t) = v_z(r,t)\,C_i(0,r,t) - \mathrm{D}_z(r,t)\frac{\partial C_i(0,r,t)}{\partial z}$$

$$\frac{\partial C_i}{\partial z}(L,r,t) = 0$$

no reaction at exit

$$\frac{\partial C_i}{\partial r}(z,0,t) = 0$$

Symmetry

$$\frac{\partial C_i}{\partial r}(z,R,t) = 0$$

Impermeable wall

$$C_i(z,r,0) = C_{i\_in}(z,r)$$

Initial condition

Energy Balance

(removing time average notation)

$$\rho c_p \frac{\partial T}{\partial t} = k_z(r,t)\frac{\partial^2 T}{\partial z^2} + \frac{1}{r}\frac{\partial}{\partial r}\left(r\, k_R(r,t)\frac{\partial T}{\partial r}\right) - \rho c_p\, v_z(r,t)\frac{\partial T}{\partial z} + \Delta H_r\, R_A$$

$$v_z(r,t)T_0(t) = v_z(r,t)T(0,r,t) - \frac{k_z(r,t)}{\rho c_p}\frac{\partial T(0,r,t)}{\partial z}$$

$$\frac{\partial T}{\partial z}(L,r,t) = 0$$

No reaction at exit

$$\frac{\partial T}{\partial r}(z,0,t) = 0$$

Symmetry

$$k_R(R,t)\frac{\partial T}{\partial r}(z,R,t) = U\left[T_w - T(z,R,t)\right]$$

Heat exchange with the wall

$$T(z,r,0) = T_{in}(z,r)$$

Initial condition

Momentum Balance

(removing time average notation)

$$\rho \frac{\partial v_z}{\partial t} = -\frac{\partial P}{\partial z} + \mu \frac{1}{r} \frac{\partial}{\partial r}\left(r \frac{\partial v_z}{\partial r} \right)$$

$$\frac{\partial v_z}{\partial r}(0,t) = 0$$ **Symmetry**

$$v_z(R,t) = 0$$ **Fixed wall**

$$v_z(r,0) = v_{z\_in}(r)$$ **Initial Condition**

Additional assumptions:

- constant effective diffusive coefficients
- constant velocity

$$\frac{\partial C_i}{\partial t} = \mathrm{D}_L \frac{\partial^2 C_i}{\partial z^2} + \frac{\mathrm{D}_R}{r}\frac{\partial}{\partial r}\left(r \frac{\partial C_i}{\partial r} \right) - v_z \frac{\partial C_i}{\partial z} + R_i$$

$$\rho c_p \frac{\partial T}{\partial t} = k_L \frac{\partial^2 T}{\partial z^2} + \frac{k_R}{r}\frac{\partial}{\partial r}\left(r \frac{\partial T}{\partial r} \right) - \rho c_p v_z \frac{\partial T}{\partial z} + \Delta H_r R_A$$

$$v_z C_{i0}(t) = v_z C_i(0,r,t) - \mathrm{D}_L \frac{\partial C_i(0,r,t)}{\partial z}$$

$$\frac{\partial C_i}{\partial z}(L,r,t) = 0$$

$$\frac{\partial C_i}{\partial r}(z,0,t) = 0$$

$$\frac{\partial C_i}{\partial r}(z,R,t) = 0$$

$$C_i(z,r,0) = C_{i\_in}(z,r)$$

$$v_z T_0(t) = v_z T(0,r,t) - \frac{k_L}{\rho c_p} \frac{\partial T(0,r,t)}{\partial z}$$

$$\frac{\partial T}{\partial z}(L,r,t) = 0$$

$$\frac{\partial T}{\partial r}(z,0,t) = 0$$

$$k_R \frac{\partial T}{\partial r}(z,R,t) = U\left[T_w - T(z,R,t) \right]$$

$$T(z,r,0) = T_{in}(z,r)$$

Method of Lines

- spatial discretization (finite differences, finite volumes, finite elements)

- time integration

Example: finite differences in axial direction and orthogonal collocation in radial

$$\frac{dC_{i,j,k}}{dt} = \mathrm{D}_L \left(\frac{C_{i,j+1,k} - 2C_{i,j,k} + C_{i,j-1,k}}{\Delta z^2} \right) + 4\,\mathrm{D}_R \left[\sum_{m=0}^{n+1} \left(u_k B_{k,m} + A_{k,m} \right) C_{i,j,m} \right] - v_z \left(\frac{C_{i,j+1,k} - C_{i,j-1,k}}{2\Delta z} \right) + R_{i,j,k}$$

$$\rho c_p \frac{dT_{j,k}}{dt} = k_L \left(\frac{T_{j+1,k} - 2T_{j,k} + T_{j-1,k}}{\Delta z^2} \right) + 4 k_R \left[\sum_{m=0}^{n+1} \left(u_k B_{k,m} + A_{k,m} \right) T_{j,m} \right] - \rho c_p v_z \left(\frac{T_{j+1,k} - T_{j-1,k}}{2\Delta z} \right) + \Delta H_r R_{A,j,k}$$

where:

$$u = r^2 \qquad l_m(u) = \prod_{\substack{p=0 \\ p \neq m}}^{n+1} \frac{u - u_p}{u_m - u_p} \qquad A_{k.m} = \frac{dl_m(u_k)}{du} \qquad B_{k,m} = \frac{d^2 l_m(u_k)}{du^2}$$

$$j = 1,2,\ldots,N \qquad k = 1,2,\ldots,n \qquad y(u) \cong P_{n+1}^{(\alpha,\beta)}(u) = \sum_{m=0}^{n+1} l_m(u)\, y_m$$

Method of Lines

Boundary conditions

$$v_z\, C_{i0}(t) = v_z\, C_{i,1,k} - \mathrm{D}_L \left(\frac{C_{i,2,k} - C_{i0}(t)}{2\Delta z} \right)$$

$$v_z\, T_0(t) = v_z\, T_{1,k} - \frac{k_L}{\rho\, c_p} \left(\frac{T_{2,k} - T_0(t)}{2\Delta z} \right)$$

$$C_{i,N+1,k} - C_{i,N,k} = 0$$

$$T_{N+1,k} - T_{N,k} = 0$$

$$\sum_{m=0}^{n+1} A_{0,m} C_{i,j,m} = 0$$

$$\sum_{m=0}^{n+1} A_{0,m} T_{j,m} = 0$$

$$\sum_{m=0}^{n+1} A_{n+1,m} C_{i,j,m} = 0$$

$$k_R \sum_{m=0}^{n+1} A_{n+1,m} T_{j,m} = U \left[T_w - T_{j,n+1} \right]$$

$$C_{i,j,k} = C_{i\_in,j,k}$$

$$T_{j,k} = T_{in,j,k}$$

$$j = 1,2,\dots,N \qquad k = 1,2,\dots,n$$

$\longrightarrow$ Results in a DAE system!

Another Multiple Gradients Model

(ignoring radial gradients) – PFR with axial dispersion

$$\frac{\partial C_i}{\partial t} = D_L \frac{\partial^2 C_i}{\partial z^2} - v_z \frac{\partial C_i}{\partial z} + R_i \longleftarrow \bar{C}_i(z,t) = \frac{\int_0^R C_i(r,z,t)\, r\, dr}{\int_0^R r\, dr}$$

$$v_z C_{i0}(t) = v_z C_i(0,t) - D_L \frac{\partial C_i(0,t)}{\partial z}$$

$$\frac{\partial C_i}{\partial z}(L,t) = 0$$

$$C_i(z,0) = C_{i\_in}(z)$$

$$\rho c_p \frac{\partial T}{\partial t} = k_L \frac{\partial^2 T}{\partial z^2} - \rho c_p v_z \frac{\partial T}{\partial z} + \frac{2U}{R}\left(T_w - T\right) + \Delta H_r R_A \longleftarrow \bar{T}(z,t) = \frac{\int_0^R T(r,z,t)\, r\, dr}{\int_0^R r\, dr}$$

$$v_z T_0(t) = v_z T(0,t) - \frac{k_L}{\rho c_p} \frac{\partial T(0,t)}{\partial z}$$

$$\frac{\partial T}{\partial z}(L,t) = 0$$

$$T(z,0) = T_{in}(z)$$

Method of Lines

- spatial discretization (F.D., F.V., orthogonal collocation on finite elements)
- time integration

Example: finite differences in axial direction

$$\frac{dC_{i,j}}{dt} = \mathrm{D}_L \left(\frac{C_{i,j+1} - 2C_{i,j} + C_{i,j-1}}{\Delta z^2} \right) - v_z \left(\frac{C_{i,j+1} - C_{i,j-1}}{2\Delta z} \right) + R_{i,j}$$

$$j = 1,2,\ldots,N$$

$$\rho\, c_p \frac{dT_j}{dt} = k_L \left(\frac{T_{j+1} - 2T_j + T_{j-1}}{\Delta z^2} \right) - \rho\, c_p\, v_z \left(\frac{T_{j+1} - T_{j-1}}{2\Delta z} \right) + \frac{2U}{R}\left(T_w - T_j\right) + \Delta H_r\, R_{A,j}$$

$$v_z\, C_{i0}(t) = v_z\, C_{i,1} - \mathrm{D}_L \left(\frac{C_{i,2} - C_{i0}(t)}{2\Delta z} \right)$$

$$v_z\, T_0(t) = v_z\, T_1 - \frac{k_L}{\rho\, c_p} \left(\frac{T_2 - T_0(t)}{2\Delta z} \right)$$

$$C_{i,N+1} - C_{i,N} = 0$$

$$T_{N+1} - T_N = 0$$

$$C_{i,j} = C_{i\_in,j}$$

$$T_j = T_{in,j}$$

Maximum Gradient Model

(ignoring axial dispersion) – PFR without axial dispersion

$$\frac{\partial C_i}{\partial t} = -v_z \frac{\partial C_i}{\partial z} + R_i$$

$$C_i(0,t) = C_{i0}(t)$$

$$C_i(z,0) = C_{i\_in}(z)$$

$$\rho\, c_p \frac{\partial T}{\partial t} = -\rho\, c_p\, v_z \frac{\partial T}{\partial z} + \frac{2U}{R}\left(T_w - T\right) + \Delta H_r\, R_A$$

$$T(0,t) = T_0(t)$$

$$T(z,0) = T_{in}(z)$$

→ Results in an EDO system!

Macroscopic Model

$$V\frac{dC_i}{dt} = C_{i0}(t)\,v_z\,S - C_i\,v_z\,S + \overline{R}_i\,V \longleftarrow \overline{C}_i(t) = \frac{\int_0^L C_i(z,t)\,S\,dz}{\int_0^L S\,dz}$$

$$C_i(0) = \overline{C}_{i\_in}$$

$$\rho\,c_p\,V\frac{dT}{dt} = \rho\,c_p\,v_z\,S\,T_0(t) - \rho\,c_p\,v_z\,S\,T + U\,A_t\,\overline{(T_w - T)} + \Delta H_r\,\overline{R}_A\,V \longleftarrow \overline{T}(t) = \frac{\int_0^L T(z,t)\,S\,dz}{\int_0^L S\,dz}$$

$$T(0) = \overline{T}_{in}$$

$\longrightarrow$ Results in a EDO system!

Method of Orthogonal Collocation with EMSO

- Boundaries
- Internal Points
- Alfa and Beta

EMSO
Simulator

Plugin

Roots
A and **B** Matrices

- Jacobi roots
- **A** and **B** matrices

DD as Plugin (Type="OCFEM", Boundary="BOTH", InternalPoints=5 alfa=1, beta=1)

Plugin: ocfem_emso.dll

Fixed-bed Reactor with Axial Dispersion

(reaction of order *m*)

$$\frac{\partial y}{\partial \tau} + \frac{\partial y}{\partial x} = \frac{1}{Pe}\frac{\partial^2 y}{\partial x^2} - Da\, y^m$$

Boundary conditions:

$$-\frac{1}{Pe}\left.\frac{\partial y}{\partial x}\right|_{x=0} = 1 - y(\tau, 0) \qquad \text{or} \qquad y(\tau, 0) = 1$$

$$\left.\frac{\partial y}{\partial x}\right|_{x=1} = 0$$

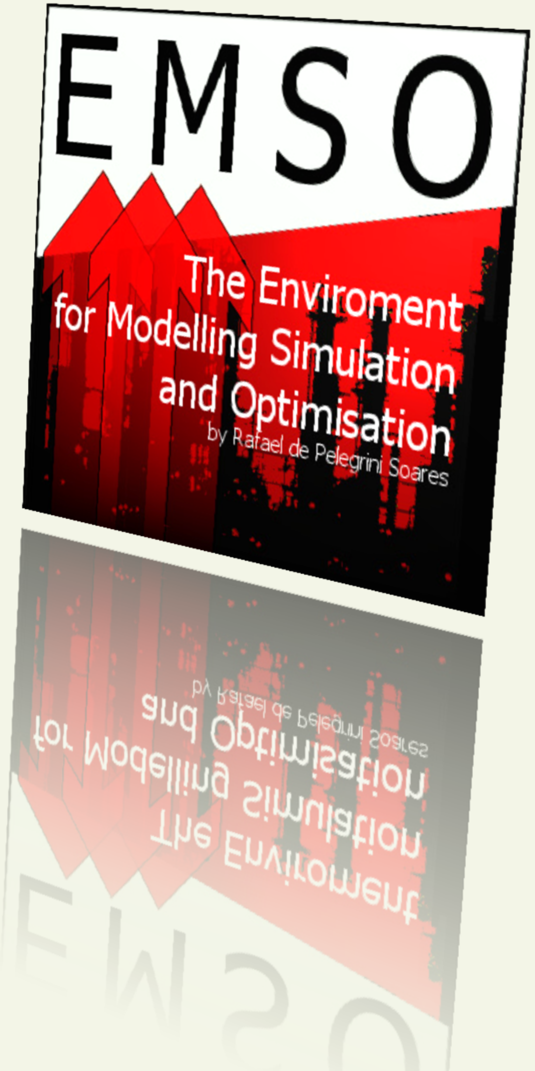
Initial conditions:

$$y(0, x) = 0$$

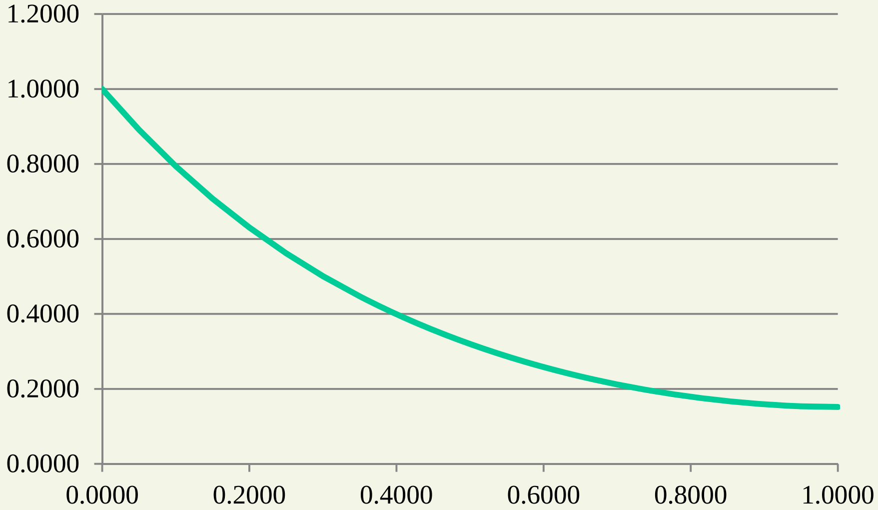
PDE

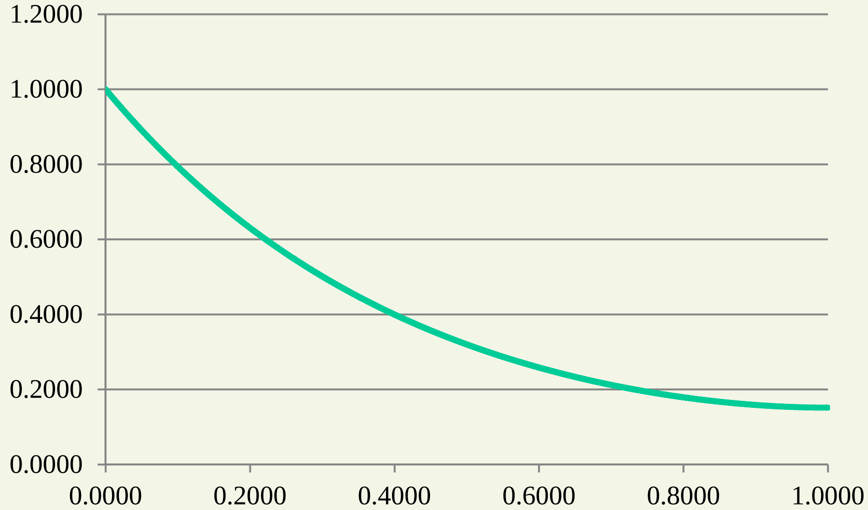
Method of Lines: D.F. and Orthogonal Collocation

Example: add Plugin ocfem_emso.dll and execute flowsheets of files FDM_ss.mso, OCM_ss.mso and OCFEM_ss.mso, and compare results of discretizations. Repeat for the dynamic simulation in files FDM_din.mso e OCM_din.mso.



Comparing Results





OCM by EMSO $\alpha = 1 \quad \beta = 1$
Number of internal points: 5

y(x=1) = 0.151475 (error of 0.038%)

Method of Finite Differences
Number of internal points: 6000

y(x=1) = 0.15155 (error of 0.087%)

y(x=1) = 0.151418 (exact)

Case Study

- **Production of acetic anhydride in adiabatic PFR**
  - Acetic anhydride is often produced by reacting acetic acid with ketene, obtained by heating acetone at 700-770^{o}C.
  - A important step is the vapor phase cracking of acetone to ketene and methane:

$$CH_3COCH_3 \rightarrow CH_2CO + CH_4$$

  - The second step is the reaction of ketene with acetic acid.

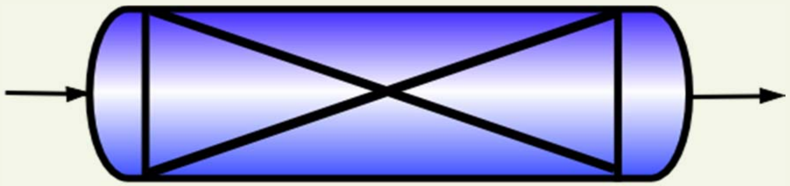
$$CH_2CO + CH_3COOH \rightarrow (CH_3CO)O$$

Ref: G. V. Jeffreys, *A Problem in Chemical Engineering Design: The Manufacture of Acetic Anhydride*, 2nd ed. (London: Institution of Chemical Engineers, 1964)

Case Study

Problem Definition

– The first production step is carried out in a vapor phase reaction of acetone in an adiabatic PFR.



where A = acetone; B = ketene and C = methane

$$A \rightarrow B + C$$

– The reaction is of 1ª order in relation to acetone in the cracking reaction, with Arrhenius constant given by:

- k – seconds^{-1}
- T – Kelvin

$$k = \exp\left(34.34 - \frac{34222}{T}\right)$$

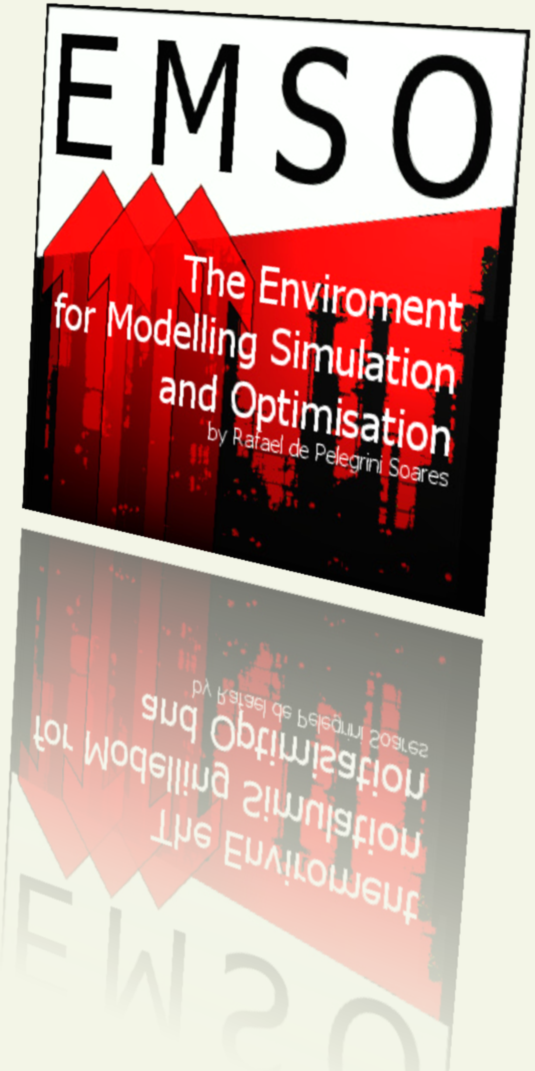
Case Study

Process Description

- Reactor geometry
  - adiabatic continuous tubular reactor;
  - bank of 1000 tubes of 1 in sch. 40 with cross section of 0.557 m^2;
  - total length of 2.28 m;
- Operating conditions
  - feed temperature 762^oC (1035 K);
  - operating pressure: 1.6 atm
  - feed flow rate of 8000 kg/h (137.9 kmol/h);
- Composition
  - acetone, ketene and methane
  - feed of pure acetone
- Kinetics
  - first order reaction,
  - pre-exponential factor (k_0): 8.2 x 10^{14} s^{-1}
  - activation energy (E/R): 34222 K
  - heat of reaction: -80.77 kJ/mol

Case study
– Production of acetic anhydride –

Example: run FlowSheet in file PFR_Adiabatico.mso and plot steady-state temperature and composition profiles. Show also the evolution of the temperature profile. Discuss the type and quality of discretization.



Exercise

Solve the reaction-diffusion problem in a spherical catalytic particle, given by:

$$\frac{\partial y}{\partial t} = \frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2 \frac{\partial y}{\partial r}\right) - \Phi^2 y^{1/2}$$

$$\left.\frac{\partial y}{\partial r}\right|_{r=0} = 0 \qquad \left. y(t,r)\right|_{r=1} = 1 \qquad \left. y(t,r)\right|_{t=0} = 0$$

$$\Phi = 2 \quad \text{(Thiele modulus)}$$

References

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Apoio:
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BR PETROBRAS Braskem COPESUL innova Ipiranga REFAP Alberto Pasqualini - Refap S/A

For supporting the ALSOC Project.

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... thank you for your attention!

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Process Modeling, Simulation and Control Lab

- Prof. Argimiro Resende Secchi, D.Sc.
- Phone: +55-21-2562-8307
- E-mail: arge@peq.coppe.ufrj.br
- http://www.peq.coppe.ufrj.br/Areas/Modelagem_e_simulacao.html