

CHEMICAL REACTION ENGINEERING (SKF3223)

Chapter 4: Isothermal Reaction Design

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The general mole balance equation:

 $F_{A0} - F_A + \int r_A dV = \frac{dN_A}{dt}$

Apply mole balance to specific reactor design equations:

 $Batch: N_{A0} \frac{dX}{dt} = -r_A V$ $CSTR: V = \frac{F_{A0} X}{-r_A}$ $PFR: F_{A0} \frac{dX}{dV} = -r_A$ $PBR: F_{A0} \frac{dX}{dW} = -r_A$



 $-r_A = f(X)$

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4 Determine the rate law in terms of the concentration of the reacting species:

$$-r_A = k \left(C_A C_B - \frac{C_C}{K_C} \right)$$

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Use stoichiometry to express concentration as a function of conversion

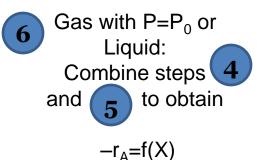
(i) Liquid-phase or constant volume batch:

$$C_{A} = C_{A0}(1 - X)$$
$$C_{B} = C_{A0} \left(\Theta_{B} - \frac{b}{a} X \right)$$

(ii) Gas-phase $T=T_0$:

$$C_A = C_{A0} \frac{(1-X)}{(1+\varepsilon X)} \frac{P}{P_0}$$





Gas phase reaction with pressure drop: $\frac{dy}{dW} = -\frac{\alpha}{2y}(1 + \varepsilon X)$ $y = \frac{P}{P_0}$

Semibatch reactors:

 $V = V_0 + v_0 t$

Combine mole balance, rate law and stoichiometry, transport law, and pressure drop term in an ordinary differential equation solver (ODE solver Polymath)

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<u>ALGORITHM FOR</u> ISOTHERMAL REACTOR

- 1. Mole balance
 - 2. Rate law
- 3. Stoichiometry (specify whether the reaction is gas or

liquid phase)

4. Combine

5. Evaluate (Analytically-Appendix A1, Graphically-Chapter 2, Numerically-Appendix A4, Polymath software)

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Mole balance		$\frac{dX}{dt} = \frac{-r_A}{N_{A0}}V$	
Rate law	$-r_A = kC_A$		$-r_A = kC_A^2$
Stoichiometry V=V ₀		$C_A = \frac{N_A}{V_0} = C_{A0}(1 - X)$	
Combine	$\frac{dX}{dt} = k(1 - X)$		$\frac{dX}{dt} = kC_{A0}(1-X)^2$
Integrate $t = N_{A0} \int_{0}^{X} \frac{dX}{-r_{A}V}$	$t = \frac{1}{k} \ln \frac{1}{1 - X}$		$t = \frac{X}{kC_{A0}(1-X)}$



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ALGORITHM - CSTR

Mole balance	$V = \frac{F_{A0}.X}{-r_A}$		
Rate law	$-r_A = kC_A$		$-r_A = kC_A^2$
Space time		$\tau \equiv \frac{V}{\nu_0}$	
Conversion	1 st order:	1 st order:	2 nd order:
	Parallel	Series	$X = \frac{(1+2Da) - \sqrt{1+4Da}}{2Da}$
	$X = \frac{\tau . k}{1 + \tau . k} = \frac{Da}{1 + Da}$	$X = 1 - \frac{1}{\left(1 + \tau . k\right)^n}$	2Da
n= number of reactors	$Da = \tau k$	$C_{An} = \frac{C_{A0}}{\left(1 + Da\right)^n}$	$Da = \tau k C_{A0}$
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- One of important characteristic in CSTR equation is Damköhler number, Da
- Da number is a dimensionless

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If Da < 0.1, then X < 0.1If Da > 10, then X > 0.9

$$Da = \frac{-r_{A0}.V}{F_{A0}}$$

Rate of reaction at entrance
 Entering flow rate of A

$$Da = \frac{kC_{A0}.V}{\nu_0.C_{A0}} = \tau.k$$

First order irreversible reaction

$$Da = \frac{kC_{A0}.V}{\nu_0 C_{A0}} = \tau .kC_{A0}$$

Second order irreversible reaction



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<u>ALGORITHM – TUBULAR</u> (GAS-PHASE) (No pressure drop)

Mole balance		$V = F_{A0} \int_{0}^{X} \frac{dX}{-r_{A}}$	
Rate law	$-r_A = kC_A$		$-r_A = kC_A^2$
Stoichio- metry		$C_A = C_{A0} \frac{(1-X)}{(1+\varepsilon X)}$	
combine	$V = \frac{F_{A0}}{kC_{A0}} \int_{0}^{X} \frac{(1 + \varepsilon X)}{(1 - X)} dX$		$V = \frac{F_{A0}}{kC_{A0}^{2}} \int_{0}^{X} \frac{(1 + \varepsilon X)^{2}}{(1 - X)^{2}} dX$
	$= \frac{F_{A0}}{kC_{A0}} \left[(1+\varepsilon) \ln \frac{1}{1-X} - \varepsilon X \right]$		080 EX NO 5

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PRESSURE DROPS IN REACTORS

Liquid-phase reaction: pressure drop can totally ignored

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- □ Gas-phase reaction: Pressure drop will be considered is very
- important key factor in the success or failure of the reactor operation
- □ For an ideal gas, the concentration of reacting species i:

$$C_{i} = C_{A0} \left(\frac{\Theta_{i} + v_{i}X}{1 + \varepsilon X} \right) \frac{P}{P_{0}} \frac{T_{0}}{T}$$

$$\Theta_{i} = \frac{F_{i0}}{F_{A0}}, \varepsilon = y_{A0}\delta, v = stoichiometric coefficient$$

□ When $P \neq P_0$, use the differential forms of the PFR/PBR design equations



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$$2A \rightarrow B + C$$



dV

1. Mole balance - Differential form:

$$F_{A0} \frac{dX}{dW} = -r_{A}$$

2. Rate law:
$$-r_A' = kC_A^2$$

3. From stoichiometry for gas-phase reactions:

$$r_{A} = f(X) - r_{A}' = k \left[\frac{C_{A0}(1-X)}{1+\varepsilon X} \frac{P}{P_{0}} \frac{T_{0}}{T} \right]^{2}$$

 $C_A = \frac{C_{A0}(1-X)}{1+\varepsilon X} \frac{P}{P_0} \frac{T_0}{T}$

Isothermal, $T = T_0$

4. Combine

$$F_{A0}\frac{dX}{dW} = k\left[\frac{C_{A0}(1-X)}{1+\varepsilon X}\right]^2 \left(\frac{P}{P_0}\right)^2 \quad \Longrightarrow \quad \frac{dX}{dW} = \frac{kC_{A0}}{v_0}\left[\frac{(1-X)}{1+\varepsilon X}\right]^2 \left(\frac{P}{P_0}\right)^2$$

5. Relate the pressure drop (P/P_0) to the catalyst weight \rightarrow to determine X as a function of catalyst weight





PRESSURE DROPS IN PBR

Ergun Equation:

$$\frac{dP}{dz} = \frac{-G}{\rho g_C D_P} \left(\frac{1-\phi}{\phi^3}\right) \left[\frac{150(1-\phi)\mu}{D_P} + 1.75G\right]$$

1 Variable Gas Density:
$$\rho = \rho_0 \frac{P}{P_0} \frac{T_0}{T} \frac{F_{T0}}{F_T}$$

 $P = Ib_f/ft^2$ (kPa) Φ = porosity = void fraction = volume of void total bed volume $1-\Phi =$ volume of solid total bed volume $gc = 32.174 \ lb_m ft/s^2 lb_f$ (conversion factor) Dp = diameter of particle in the bed, ft(m) μ = viscosity of gas, Ib_m/ft.h (kg/m.s) z =length of PBR pipe, ft (m) u = superficial velocity = volumetric flow/cross section, ft/h (m/s) ρ = gas density, ib/ft³ (kg/m³) $G = \rho u =$ superficial mass velocity, Ibm/ft²h (kg/m²s)

$$\frac{dP}{dz} = \frac{-G}{\rho_0 g_C D_P} \left(\frac{1-\phi}{\phi^3}\right) \left[\frac{150(1-\phi)\mu}{D_P} + 1.75G\right] \frac{P_0}{P} \frac{T}{T_0} \frac{F_T}{F_{T0}}$$



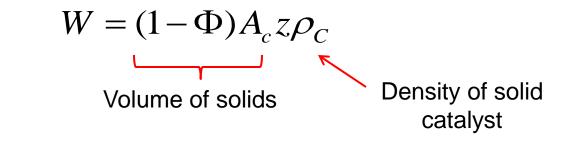


□ Simplifying yields:

$$\frac{dP}{dz} = -\beta_0 \frac{P_0}{P} \frac{T}{T_0} \frac{F_T}{F_{T_0}}$$
$$\beta_0 = \frac{G(1-\Phi)}{\rho_0 g_c D_p \Phi} \left[\frac{150(1-\Phi)}{D_p} + 1.75G \right]$$

 $\begin{array}{c} \beta_0 \text{ is a constant} \\ \text{down the reactor} \\ \text{that depends only} \\ \text{on the properties of} \\ \text{the PBR and the} \\ \text{entrance conditions} \end{array}$

□ The catalyst weight up to a distance of z down the reactor is:



Bulk density:

 $\rho_b = \rho_c (1 - \phi)$



□ Ergun equation in terms of catalyst weight:

$$\frac{dP}{dW} = -\frac{\beta_0}{A_C(1-\phi)\rho_C} \frac{P_0}{P} \left(\frac{T}{T_0}\right) \frac{F_T}{F_{T0}}$$

□ Further simplification yields:

$$\frac{dP}{dW} = -\frac{\alpha}{2} \frac{T}{T_0} \frac{P_0}{P/P_0} \left(\frac{F_T}{F_{T0}}\right) \qquad \alpha = \frac{2\beta_0}{A_c \rho_c (1-\phi)P_0} \qquad y = \frac{P}{P_0}$$

Use for multiple reactions and membrane reactors

$$\frac{dy}{dW} = -\frac{\alpha}{2y} \frac{T}{T_0} \left(\frac{F_T}{F_{T0}}\right)$$

Use for single reactions in PBR

$$\frac{dy}{dW} = -\frac{\alpha}{2y}(1 + \varepsilon X)\frac{T}{T_0}$$

$$F_{T} = F_{T0} + F_{A0} \delta X = F_{T0} \left[1 + \frac{F_{A0}}{F_{T0}} \delta X \right]$$

$$\frac{F_T}{F_{T0}} = 1 + \varepsilon X \qquad \varepsilon = y_{A0} \delta = \frac{F_{A0}}{F_{T0}} \delta$$



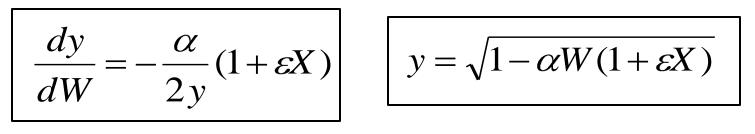




□ ONLY for isothermal with T/T₀=0, ϵ = 0:

$$y = \frac{P}{P_0} = \sqrt{(1 - \alpha W)}$$

□ For isothermal with T/T₀=0, $\epsilon \neq 0$:



$$y = \frac{P}{P_0} = \left(1 - \frac{2\beta_0 z}{P_0}\right)^{1/2}$$

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$\underline{\mathsf{EXAMPLE}} \qquad A \longrightarrow B$

$$\mathbf{1} \quad F_{A0} \frac{dX}{dW} = -r_{A}$$

$$2 - r_A' = kC_A^2$$

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$$C_A = C_{A0}(1-X)\frac{P}{P_0} = C_{A0}(1-X)y$$

$$y = \frac{P}{P_0} = \sqrt{(1 - \alpha W)}$$

$$-r_{A}' = kC_{A0}^{2}(1-X)^{2}(\sqrt{(1-\alpha W)})^{2}$$
$$-r_{A}' = kC_{A0}^{2}(1-X)^{2}(1-\alpha W)$$

Could now solve for X given W, or for W given X.

$$\frac{dX}{dW} = \frac{kC_{A0}^{2}(1-X)^{2}(1-\alpha W)}{F_{A0}}$$
$$\left[\frac{1}{(1-X)^{2}}\right]dX = \left[\frac{kC_{A0}^{2}}{F_{A0}}(1-\alpha W)\right]dW$$
$$\left[\frac{X}{1-X} = \frac{kC_{A0}^{2}}{F_{A0}}\left[W - \frac{\alpha W^{2}}{2}\right]\right]$$
$$\left[\frac{kC_{A0}W}{1-X}\left(1 - \frac{\alpha W}{2}\right)\right]$$
$$\left[\frac{kC_{A0}W}{1 + \frac{kC_{A0}W}{v_{0}}\left(1 - \frac{\alpha W}{2}\right)}\right]$$
$$\left[\frac{1 - \sqrt{1 - \left[\frac{(2v_{0}\alpha)}{kC_{A0}}\right]\left[\frac{X}{1-X}\right]}}{\alpha}\right]$$



 $T/T_0 = 0$, $\epsilon = 0$, with ΔP



$$T/T_0 = 0$$
, $\epsilon = 0$, in the absence of ΔP , ($\alpha = 0$)

$$-r_{A}' = kC_{A}^{2}$$

$$\frac{1}{1-X} = \frac{kC_{A0}^2}{F_{A0}}W$$

$$X = \frac{\frac{kC_{A0}W}{v_0}}{1+\frac{kC_{A0}W}{v_0}}$$

$$With \Delta P, (\alpha \neq 0)$$

$$\alpha = \frac{2\beta_0}{A_C\rho_C(1-\phi)P_0}$$

$$\alpha = \frac{2\beta_0}{A_C\rho_bP_0}$$

- X decreased with ΔP
- ✤ If we increase catalyst size, laminar terms = 0 (Ergun equation)
- ↔ By increasing the particle diameter, we decrease the pressure drop parameter (β_0) and thus increase the $-r_A$ and the X.





PRESSURE DROP IN PIPES

□ Pressure drop along the length of the pipe:

$$\frac{dP}{dL} = -G\frac{du}{dL} - \frac{2fG^2}{\rho D}$$

D = pipe diameter, cm u = average velocity of gas, cm/s f = Fanning friction factor G = ρ u, g/cm²s (mass velocity, constant along the length of the pipe)



EXAMPLE: <u>Calculating X in a Reactor With ∆P (gas=phase)</u>

$$\begin{vmatrix} A + \frac{1}{2}B \rightarrow C \\ P_{A0} \frac{dX}{dW} = -r_{A}' \\ P_{A} = kP_{A}^{1/3}P_{B}^{2/3} \\ P_{A} = C_{A}RT \\ -r_{A}' = k(C_{A}RT)^{1/3}(C_{B}RT)^{2/3} \\ -r_{A}' = k(C_{A}RT)^{1/3}(C_{B}RT)^{2/3} \\ -r_{A}' = kRTC_{A}^{1/3}C_{B}^{2/3} \\ P_{A} = \frac{C_{A0}(1-X)}{1+\varepsilon X} \left(\frac{P}{P_{0}}\right) = \frac{C_{A0}(1-X)y}{1+\varepsilon X} \\ -r_{A}' = k\frac{P_{A0}}{C_{A0}} \left[\frac{C_{A0}(1-X)y}{1+\varepsilon X}\right]^{1/3} \left[\frac{C_{A0}(1-X)y}{2(1+\varepsilon X)}\right]^{2/3} \\ -r_{A}' = kRTC_{A}^{1/3}C_{B}^{2/3} \\ -r_{A}' = kRTC_{A}^{1/3}C_{B}^{2/3} \\ -r_{A}' = kRTC_{A}^{1/3}C_{B}^{2/3} \\ R_{A} = \frac{C_{A0}(1-X)}{1+\varepsilon X} \left(\frac{P}{P_{0}}\right) = \frac{C_{A0}(1-X)y}{1+\varepsilon X} \\ K' = kP_{A0}(1/2)^{2/3} = 0.63kP_{A0} \\ K' = kP_{A0}(1/2)^{2/3} = 0.63kP_{A0} \\ R_{A} = \frac{C_{A0}(\Theta_{B} - X/2)y}{1+\varepsilon X} = \frac{C_{A0}(1-X)y}{2(1+\varepsilon X)} \\ R_{A} = \frac{C_{A0}(\Theta_{B} - X/2)y}{1+\varepsilon X} = \frac{C_{A0}(1-X)y}{2(1+\varepsilon X)} \\ R_{A} = \frac{C_{A0}(\Theta_{B} - X/2)y}{1+\varepsilon X} = \frac{C_{A0}(1-X)y}{2(1+\varepsilon X)} \\ R_{A} = \frac{C_{A0}(\Theta_{B} - X/2)y}{1+\varepsilon X} = \frac{C_{A0}(1-X)y}{2(1+\varepsilon X)} \\ R_{A} = \frac{C_{A0}(1/2)^{2/3}}{1+\varepsilon X} = 0.63kP_{A0} \\ R_{A} = \frac{C_{A0}(\Theta_{B} - X/2)y}{1+\varepsilon X} \\ R_{A} = \frac{C_{A0}(1-X)}{1+\varepsilon X} \\ R_{A} = \frac{C_{A0}(1-X)}{1$$



MOLE BALANCES FOR LIQUID-PHASE REACTIONS

$$A + \frac{b}{a}B \to \frac{c}{a}C + \frac{d}{a}D$$

Batch	$\frac{dC_A}{dt} = r_A$	$\frac{dC_B}{dt} = \frac{b}{a} r_A$
CSTR	$V = \frac{v_0 (C_{A0} - C_A)}{-r_A}$	$V = \frac{v_0 (C_{B0} - C_B)}{-(b/a)r_A}$
PFR	$v_0 \frac{dC_A}{dV} = r_A$	$v_0 \frac{dC_B}{dV} = \frac{b}{a} r_A$
PBR	$v_0 \frac{dC_A}{dW} = r_A$	$v_0 \frac{dC_B}{dW} = \frac{b}{a} r_A$



MOLE BALANCES FOR GAS-PHASE REACTIONS

$$A + \frac{b}{a}B \to \frac{c}{a}C + \frac{d}{a}D$$

BATCH	CSTR	PFR
$\frac{dN_A}{dt} = r_A V$	$V = \frac{F_{A0} - F_A}{-r_A}$	$\frac{dF_A}{dV} = r_A$
$\frac{dN_B}{dt} = r_B V$	$V = \frac{F_{B0} - F_B}{-r_B}$	$\frac{dF_B}{dV} = r_B$
$\frac{dN_C}{dt} = r_C V$	$V = \frac{F_{C0} - F_C}{-r_C}$	$\frac{dF_C}{dV} = r_C$
$\frac{dN_D}{dt} = r_D V$	$V = \frac{F_{D0} - F_D}{-r_D}$	$\frac{dF_D}{dV} = r_D$

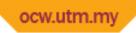




GAS-PHASE REACTIONS

$$\begin{array}{c} \mathbf{1} \quad \frac{dF_{j}}{dV} = r_{j} \\ \mathbf{3} \quad C_{j} = C_{T0} \frac{F_{j}}{F_{T}} \frac{T_{0}}{T} y \\ C_{A} = C_{T0} \frac{F_{A}}{F_{T}} \frac{T_{0}}{T} y \\ C_{B} = C_{T0} \frac{F_{B}}{F_{T}} \frac{T_{0}}{T} y \\ F_{T} = F_{A} + F_{B} + F_{C} + F_{D} + F_{I} \\ \frac{dy}{dW} = \frac{-\alpha}{2y} \frac{F_{T}}{F_{T0}} \end{array}$$

$$\begin{array}{c} \mathbf{2} \quad -r_{A} = k_{A} C_{A}^{\alpha} C_{B}^{\beta} \\ \frac{r_{A}}{-a} = \frac{r_{B}}{-b} = \frac{r_{C}}{c} = \frac{r_{D}}{d} \\ \frac{dF_{A}}{-a} = \frac{r_{B}}{-b} = \frac{r_{C}}{c} = \frac{r_{D}}{d} \\ \frac{dF_{A}}{dV} = r_{A} \\ \frac{dF_{A}}{dV} = -k_{A} C_{T0}^{\alpha\beta} \left(\frac{F_{A}}{F_{T}} \frac{T_{0}}{T} y\right)^{\alpha} \left(\frac{F_{B}}{F_{T}} \frac{T_{0}}{T} y\right)^{\beta} \\ F_{T} = F_{A} + F_{B} + F_{C} + F_{D} + F_{I} \\ \frac{dF_{A}}{dV} = -k_{A} C_{T0}^{\alpha\beta} \left(\frac{F_{A}}{F_{T}} y\right)^{\alpha} \left(\frac{F_{B}}{F_{T}} y\right)^{\beta} \\ \end{array}$$





<u>UNSTEADY-STATE OPERATION:</u> <u>CSTR & SEMIBATCH</u>

- To determine the time necessary to reach steady-state operation
- To predict the concentration, C and X as a function of time
- ✤ Analytical solutions: zero and 1st order reactions
- ODE solvers: other reaction orders





LIQUID-PHASE REACTIONS (CSTR)

$$v = v_o$$
, $V = V_o$, $\tau = V_0/v_0$

1
$$F_{A0} - F_A + r_A V = \frac{dN_A}{dt}$$
$$C_{A0} - C_A + r_A \tau = \tau \frac{dC_A}{dt}$$
$$-r_A = kC_A$$
$$\frac{dC_A}{dt} + \frac{1 + \tau k}{\tau} C_A = \frac{C_{A0}}{\tau}$$

3

Initial condition:
$$C_A = C_{A0}$$
 at t = 0:

$$C_{A} = \frac{C_{A0}}{1 + \tau k} \{1 - \exp[-(1 + \tau k)\frac{t}{\tau}]\}$$

 t_{s} = the time necessary to reach 99% of the steadystate concentration, C_{AS} : $(C_{A} = 0.99C_{AS})$

$$C_{AS} = \frac{C_{A0}}{1 + \tau k}$$

Slow reactions with small k (1>>τk): $t_s = 4.6\tau$

Rapid reactions with large k (τk >> 1):

$$t_s = \frac{4.6}{k}$$





<u>SEMIBATCH REACTOR – in terms of</u> <u>concentration, C</u>

General mole balance:

$$r_A V(t) = \frac{dN_A}{dt}$$

In terms of concentration:
$$r_A V = \frac{d(C_A V)}{dt} = \frac{V dC_A}{dt} + C_A \frac{dV}{dt}$$

Semibatch reactor volume as a function of time:

$$V = V_0 + v_0 t$$

Mole balance on A:

$$\frac{dC_A}{dt} = r_A - \frac{\upsilon_0}{V}C_A$$

Mole balance on B:

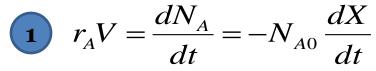
$$\frac{dC_B}{dt} = r_B + \frac{\upsilon_0 (C_{B0} - C_B)}{V}$$





<u>SEMIBATCH REACTOR – in terms of</u> conversion, X

$A + B \leftrightarrow C + D$



For constant molar feed rate and no B initially in system:

$$N_{\scriptscriptstyle B} = F_{\scriptscriptstyle B0} t - N_{\scriptscriptstyle A0} X$$

$$\begin{split} C_{B} = & \frac{N_{B}}{V} = \frac{N_{Bi} + F_{B0}t - N_{A0}X}{V_{0} + v_{0}t} \\ C_{D} = & \frac{N_{D}}{V} = \frac{N_{A0}X}{V_{0} + v_{0}t} \end{split}$$

$$2 - r_A = k \left(C_A C_B - \frac{C_C C_D}{K_C} \right)$$

B
$$C_A = \frac{N_A}{V} = \frac{N_{A0}(1-X)}{V_0 + v_0 t}$$

$$C_{C} = \frac{N_{C}}{V} = \frac{N_{A0}X}{V_{0} + v_{0}t}$$







$$\frac{dX}{dt} = \frac{k[(1-X)(N_{Bi} + F_{B0}t - N_{A0}X) - (N_{A0}X^2 / K_C)]}{V_0 + v_0 t}$$

5

At equilibrium after feedings pecies B for a time, t, the equilibrium conversion could be:

$$K_{C} = \frac{C_{Ce}C_{De}}{C_{Ae}C_{Be}} = \frac{N_{Ce}N_{De}}{N_{Ae}N_{Be}} = \frac{(N_{A0}X_{e})(N_{A0}X_{e})}{N_{A0}(1-X_{e})(F_{B0}t-N_{A0}X_{e})}$$
$$t = \frac{N_{A0}}{K_{C}F_{B0}} \left(K_{C}X_{e} + \frac{X_{e}^{2}}{1-X_{e}}\right)$$

$$X_{e} = \frac{K_{c} \left(1 + \frac{F_{B0}t}{N_{A0}}\right) - \sqrt{\left[K_{c} \left(1 + \frac{F_{B0}t}{N_{A0}}\right)\right]^{2} - 4(K_{c} - 1)K_{c} \frac{tF_{B0}}{N_{A0}}}{2(K_{c} - 1)}$$



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