k = mass transfer coefficient on the high pressure side of the membrane, cm./sec.
L = longitudinal length over which mixing is considered, cm.
M = quantity defined by Equation (13)
P = operating pressure, atm.
PeB = quantity defined by Equation (14)
$\overline{U}$ = $\overline{u}/\overline{u}^0$
$\overline{u}$ = average fluid velocity in the transverse length of the channel at a given x, cm./sec.
V_w = v_w/v_w^*
v_w = fluid velocity component in the direction perpendicular to the membrane surface, cm./sec.
v_w^* = AP/c, cm./sec.
X = quantity defined by Equation (12)
X_A = mole fraction of solute
$\overline{X}_A$ = average mole fraction of solute
x = longitudinal distance from chemical entrance, cm.

Greek Letters

γ = quantity defined by Equation (1)
Δ = quantity defined by Equation (11)
θ = quantity defined by Equation (2)
λ = quantity defined by Equation (3)
$\pi(X_A)$ = osmotic pressure corresponding to X_A, atm.

Subscripts

1 = bulk solution
2 = concentrated boundary solution
3 = membrane permeated product solution

Superscript

0 = condition at channel entrance

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Optimal Design of Jacketed Tubular Reactor with Taylor Diffusion

VINCENT P. LEUNG and KUN S. CHANG

University of Waterloo, Waterloo, Ontario, Canada

For plug flow tubular reactors, steady state ideal optimal temperature policies for various reaction schemes are available [for example, (*1* to *5*)]. In the physical implementation, such perfect ideal policies provide theoretical upper bounds and may not exactly be achieved due to the nonideal flow behavior and the physical limitations in design and operation. The nonlinear behavior can be explained by Taylor diffusion and various engineering problems with such diffusion have been posed and solved in the past for isothermal and few nonisothermal tubular reactors (*7* to *17*). Isothermal reactors in particular have received attention and the effect of axial diffusion on the yield was studied (*13, 14, 16*). The ideal optimal temperature profile was also obtained for an ideal tubular reactor (*14*). The treatment of optimal heat flux in a tubular reactor with radial diffusion is available (*18*). The physical limitations in design and operation, on the other hand, are severe if the reactions are exothermic or endothermic. Under these circumstances, optimal jacketed tubular reactors may be used (*6*). This mode of configuration introduces additional constraints on the energy balance and leads in some cases to the reduction of an infinite dimensional optimization problem to a finite dimensional one.

JACKETED TUBULAR REACTOR WITH DIFFUSION

A steady state jacketed tubular reactor with Taylor diffusion in which $(n-1)$ independent chemical species are involved in r independent reactions can be described by a system of n differential equations:

$$D_i \frac{d^2 v_i}{dz^2} - \nu \frac{dv_i}{dz} + \frac{\delta_{in}}{\alpha}[u - v_n] + f_i(k_1, \ldots, k_r, v_1, \ldots, v_n) = 0 \qquad (1)$$

$$(i = 1, 2, \ldots, n)$$

where the nth equation represents the energy balance of the reacting fluid. Here the jacket side coolant flow rate is assumed to be sufficiently large so that the variation in jacket temperature u along z is negligible. The boundary conditions at the entrance and the exit of the reactor are given by (*7* to *10*)

$$\nu v_{i0} = \nu v_i - D_i \frac{dv_i}{dz} \qquad \text{at} \quad z = 0 \qquad (2)$$

and

$$\frac{dv_i}{dz} = 0 \qquad \text{at} \quad z = L \qquad (3)$$

The rate constants are all assumed to follow Arrhenius' expression

$$k_j = k_{j0} \exp[-E_j/(R \cdot v_n)] \quad (j = 1, 2, \ldots, r)$$

For a given chemical feed, it is to design the reactor optimally in some well posed sense. An objective function may be given. Then the optimization has to be carried out over the open variables. These are L and α in design and ν, u, and v_{n0} in operation. Among these variables, α and u appear only in the process equation, ν in both process equations and boundary conditions, v_{n0} in the boundary conditions, and L implicitly in the choice of ν. Since $\alpha = [r \cdot C_p \cdot \rho/(2 \cdot U)]$ and U is approximately proportional to $(\nu^{1/3}/r)$ for laminar flow and to $(\nu^{0.8}/r)$ for turbulent flow in a tube, α is proportional to $(r^2/\nu^{1/3})$ for laminar and to $(r^2/\nu^{0.8})$ for turbulent flow, respectively. Therefore if C_p and ρ do not change appreciably along z then for a given ν, α depends only on the choice of r and is constant along z. Consequently, the choice of α corresponds to fixing the radius. If the values for some of these variables are specified, then the optimization may be carried out for the remaining open variables. For this nonlinear two point boundary value problem, an analytical solution is not in sight and the solution technique is invariably a numerical one. However, the numerical technique poses some difficulty for this type of problem [for example, (*15, 19, 20* to *22, 24*)]. One of the efficient techniques that give a short computing time and a stable iterative computation

Vincent P. Leung is with the Imperial Oil Co., Sarnia, Ontario, Canada.

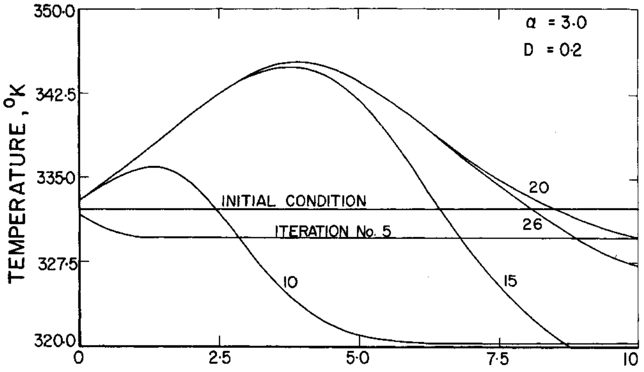

Fig. 1. Convergence of solution, temperature profile (D = 0.2, α = 3.0, u = 316.2°K., v_{30} = 332.2°K.)

is the method of nets (*23, 24*). We may view the state described by Equation (1) as the final steady state ($\partial v_i/\partial t = 0$) reached from some initial unsteady state (*15*). Then Equation (1) is replaced by

$$A_i \frac{\partial v_i}{\partial t} = D_i \frac{\partial^2 v_i}{\partial z^2} - \nu \frac{\partial v_i}{\partial z} + \frac{\delta_{in}}{\alpha}[u - v_n] + f_i(k_1, \ldots, k_r, v_1, \ldots v_n) \quad (4)$$

with the same boundary conditions in Equations (2) and (3) for $t > 0$. Asymmetric net equations can now be formulated for Equation (4) and alternating method and average method (*24*) can be used.

The consecutive first-order irreversible reaction scheme $A \xrightarrow{k_1} B \xrightarrow{k_2} C$ is now considered. The desired product is B. The reactions take place in a jacketed tubular reactor of length L. The governing differential equations with diffusion are

$$D_1 \frac{d^2 v_1}{dz^2} - \nu \frac{dv_1}{dz} - k_1 v_1 = 0$$

$$D_2 \frac{d^2 v_2}{dz^2} - \nu \frac{dv_2}{dz} + k_1 v_1 - k_2 v_2 = 0$$

$$D_3 \frac{d^2 v_3}{dz^2} - \nu \frac{dv_3}{dz} + \frac{1}{\alpha}[u - v_3] + b_1 k_1 v_1 + b_2 k_2 v_2 = 0 \quad (5)$$

with boundary conditions

$$\nu v_{i0} = \nu v_i - D_i \frac{dv_i}{dz} \quad \text{at} \quad z = 0 \quad (6)$$

and

$$\frac{dv_i}{dz} = 0 \quad \text{at} \quad z = L \quad (7)$$

$$(i = 1, 2, 3)$$

The results of optimization and optimal control are available for this reaction scheme in an ideal tubular reactor and a jacketed tubular reactor without diffusion (*1, 5, 6*). Consequently convenient comparisons can be made with regard to the effect of diffusion on optimal design. In view of this, we take numerical values:

k_{10} = 0.535×10^{11} min.$^{-1}$
k_{20} = 0.461×10^{18} min.$^{-1}$
E_1 = 0.180×10^5 cal./g.-mole
E_2 = 0.300×10^5 cal./g.-mole
R = 2.0
L = 10 units of length
ν = 1.0 unit of length/min.
b_1 = 100
b_2 = −50
v_{10} = 0.95 g.-mole/l.
v_{20} = 0.05 g.-mole/l.

We shall determine the optimal jacket temperature u, optimal feed temperature v_{30}, and optimal heat exchange parameter α such that the yield of the desired product B from the reactor exit is maximum. The cases studied are:

case 1 $D_1 = D_2 = D_3 = D = 0$ (unit of length)^2/min. (plug flow reactor)
case 2 $D_1 = D_2 = D_3 = D = 0.2$
case 3 $D_1 = D_2 = D_3 = D = 2.0$

Here D_i are assumed to be numerically the same for all i in view of the nature of D_i which reflects the dispersion by radial and longitudinal mixing of fluid rather than by true diffusion.

For a given value of D_i, this is a three dimensional search problem, a sequence of iterations being performed at each searching stage. A direct search was carried out within the range 200°K. ~ 400°K. for u and v_{30} and $0 \leq \alpha \leq 10$. These ranges were chosen from the knowledge of previous analysis and synthesis of the problem in plug flow reactors and also from the feasibility consideration. The numerical computations have been carried out using the net equations formulated. The time and length intervals were adjusted beginning from fine and coarse intervals, respectively, (Δt = 0.05 min., Δz = 0.4) in the early stage to coarse and fine intervals (Δt = 0.1 min., Δz = 0.1) in the later stage. The initial conditions were arbitrarily set at $v_i = 0$ ($i = 1, 2$) and $v_3 = v_{30}$. The convergence of the solution was assumed when the iterated concentration difference was less than 10^{-5} and the temperature difference was less than 5×10^{-2} at every mesh point. A typical example of convergence of solution is shown in Figure 1 for temperature profile. On IBM 360 system 75, each iteration takes about 0.53 sec. of execution time or approximately 14 sec. for 26 iterations.

RESULTS

For the three cases, the optimal v_{30} and u are plotted as functions of α in Figure 2. In each case, the optimal feed temperature drops sharply to an asymptotic value in the low range of α and stays there in the high α range whereas the optimal jacket temperature decreases in a straight line fashion with α. There is no appreciable difference between case 1 (D = 0) and case 2 (D = 0.2). However, for case 3 (D = 2.0) the effect of diffusion is reflected mainly on the optimal jacket temperature which is now higher than those in case 1 or 2. The straight line behavior is still retained. The corresponding optimal yields are plotted in Figure 3. It is clear that the yield curves

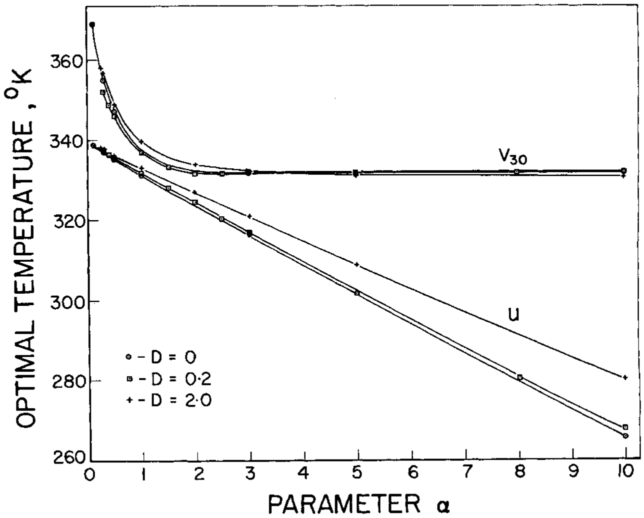

Fig. 2. α vs. optimal v_{30} and u (D = 0, 0.2, 2.0)

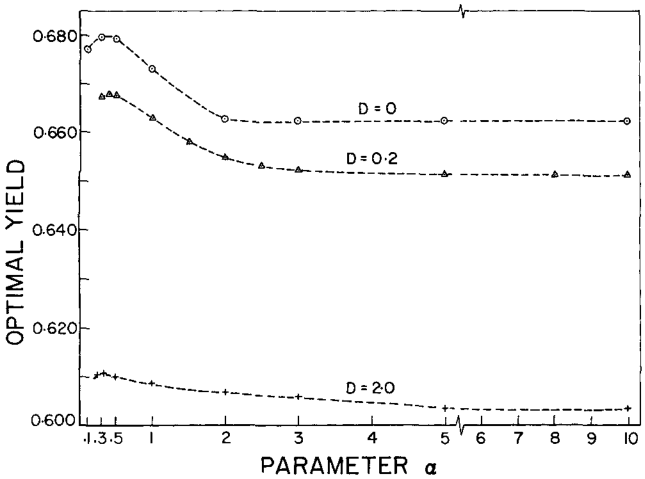

Fig. 3. α vs. optimal yield ($D = 0, 0.2, 2.0$)

are all far apart showing lower yields for higher D. In each case, the optimal peak yield occurs in the range $0.2 \leq \alpha \leq 0.4$. The peak yields are 0.679 g.-mole/liter for $D = 0$, 0.667 g.-mole/liter for $D = 0.2$ and 0.610 g.-mole/liter for $D = 2.0$. For case 1 ($D = 0$), the peak is sharp and decreases rapidly with α to the asymptotic value of 0.662 g.-mole/liter. For case 2 ($D = 0.2$), the asymptotic value is 0.651 g.-mole/liter. For case 3 ($D = 2.0$), the yield curve is flat and the peak is not as sharp as in the other cases. The asymptotic value is 0.603 g.-mole/liter.

It is apparent from the results that the optimal design parameter α, the optimal feed temperature v_{30}, and the optimal jacket temperature u are not appreciably influenced by the value of diffusion coefficient and that the optimal yield, on the other hand, is adversely affected. This may allow one to design the reactor based on a plug flow assumption. If the scale deposit during the operation reduces the reactor performance due to the corresponding change in α value, one would simply switch the operating conditions v_{30} and u to the new ones. In doing this, if the new α is less than 3.0 one may have to adjust both v_{30} and u but if $\alpha > 3.0$ then adjust u only.

CONCLUSION

Optimal design of nonlinear jacketed tubular reactor with Taylor diffusion has been carried out by means of stable asymmetric nets applied to the nonlinear partial differential equations. The results of numerical computation for the consecutive chemical reactions reveal the effect of Taylor diffusion upon the optimal design parameter and operating conditions and upon the resulting optimal yield.

For a wide range of parameter α, the optimal feed temperature v_{30} is not influenced to any appreciable degree by the magnitude of diffusion. The optimal jacket temperature u is slightly affected by the diffusion but in a straight line fashion with α. This implies that as α increases due to fouling during a prolonged operation, a simple linear adjustment of u is still sufficient to ensure the maximum yield. Consequently, a reactor optimally designed based on a plug flow assumption can be made almost optimal by simply changing the operating conditions even in the presence of a significant amount of diffusion.

However, the adverse effect of diffusion upon optimal yield is significant. As the diffusion coefficient becomes large, the yield decreases drastically and the yield curve becomes flat losing the sharp peak which existed for a truly plug flow reactor. One then can not expect to have a better yield with diffusion than without diffusion. The adverse effect of D upon the yield is due to the flattening effect upon the temperature and concentration profiles in the reactor.

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NOTATION

A_i = weighting coefficient
b_i = $(-\Delta H_i/C_p \cdot \rho)$ $(i = 1, 2)$
C_p = heat capacity
D, D_i = Taylor diffusion coefficients
E_j = activation energy $(j = 1, 2)$
f_i = function
$(-\Delta H_i)$ = heat of reaction $(i = 1, 2)$
k_j = $k_{j0} \exp(-E_j/R \cdot v_n)$ $(j = 1, 2, \ldots, r)$
k_{j0} = pre-exponential factor
L = reactor length
R = gas constant
r = reactor radius
t = time
U = overall heat transfer coefficient
u = jacket fluid temperature
v_i = concentration $(i = 1, 2, \ldots, n - 1)$
v_n = tube fluid temperature
v_{i0} = feed concentration $(i = 1, 2, \ldots, n - 1)$
v_{n0} = feed temperature
z = distance

Greek Letters

α = $r \cdot C_p \cdot \rho / 2 \cdot U$
δ_{in} = Kronecker delta
ν = fluid velocity
ρ = density

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