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# Optimal Operation of Regeneratively Cooled Fixed Bed Reactors

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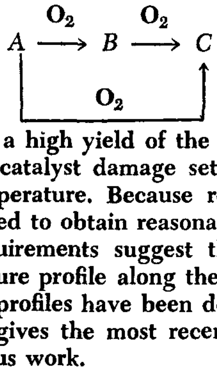
## SCOPE

Exothermic catalytic processes including hydrocarbon oxidations consist of multiple reactions such as



$$\begin{tikzcd} A \arrow[r, "O_2"] \arrow[rr, bend right, "O_2"'] & B \arrow[r, "O_2"] & C \end{tikzcd}$$

The production of a high yield of the intermediate $B$ and the prevention of catalyst damage set an upper limit on the allowable temperature. Because relatively high temperatures are needed to obtain reasonable conversions, the two opposing requirements suggest the existence of an optimum temperature profile along the reactor. Such optimum temperature profiles have been derived theoretically, and reference (*1*) gives the most recent results and summarizes the previous work.

The practical implementation of these profiles, however, would require rather elaborate heat transfer arrangements. Thus, the heat transfer methods used in industrial reactors maintain the temperature within allowable limits but do not attempt to impose any detailed temperature profile. Such arrangements include the tube- and shell-reactor heat exchanger, the dilution of the feed with an inert serving as a heat sink, the multistage reactor with intermediate cooling, and the fluidized bed reactor.

In a previous paper (*2*) we proposed regenerative cooling as another method for temperature control in exothermic catalytic reactions. Essentially, the heat of reaction is absorbed by the solid catalyst during one half of the cycle. During the other half, the catalyst is cooled by internal circulation of a gas that can be one of the reactants. This type of regenerative operation is distinct from the regeneration of a catalyst that has been deactivated by coking or a feed poison. Regenerative operation relative to heat transfer has so far been applied only to such endothermic processes as the production of ethylene and acetylene (*3*), and butene (*4*). As already mentioned, at steady state it is difficult to achieve the optimum yields because of difficulties in implementing the optimum temperature profiles. The present paper investigates the possibility of achieving yields close to the optimum steady state by optimizing the regenerative mode of operation. The implementation of the optimal policies by practical means is given special attention.

## SUMMARY

A regeneratively cooled fixed bed reactor, shown in Figure 1, consists of two sections alternating roles as reactor and heat regenerator by periodic flow reversals. The reactor can have any desired diameter because the cooling occurs strictly by regeneration. The system possesses four control variables that can be adjusted for optimal operation. The first is the temperature profile in the reacting section at the beginning of each cycle and is implemented during regeneration. The second is the temperature of the feed to the reacting section as a function of time. The third is the catalyst activity profile and can be implemented by mixing the catalyst with an inert solid. The last is the period of cycling, that is, the time interval between flow reversals. These four variables are not available in steady state designs.

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**The problem considered in this paper is the optimization of the desired reaction product by using the aforementioned four control variables. The necessary conditions of optimality are obtained by variational methods and have the form of two-point boundary value problems in the time and the position along the reactor. Numerical solutions are obtained for the oxidation of benzene to maleic anhydride in the presence of complete oxidation reactions.**

**The numerical calculations show that the temperature of the feed to the reacting section has a very small effect on the product when the cycling period is not very high. In practice, therefore, the hydrocarbon feed may be introduced at whatever temperature it is available. The importance of the catalyst activity profile was not possible to assess on the basis of the benzene oxidation example. Because the activation energies of the three competing reactions happen to be approximately equal, the product is insensitive to the activity distribution and, in fact, a uniform distribution gives the best results. In systems where the activation energies of the competing reactions are not equal, the catalyst activity profile may be a significant variable.**

**The two control variables that proved to be important are the temperature profile in the reacting section at the beginning of each cycle and the period of cycling. The optimal initial temperature profiles for various periods are shown in Figure 3. Generally they involve a dip near the reactor entrance followed by a slow rise. These profiles serve to prevent the creation of a hot spot that would occur with a uniform initial temperature. The initial profile can be implemented during the regenerative part of the cycle. This implementation requires air circulation with recycle, where the flow rate and recycle ratio must be time-programmed.**

**A very important control variable is the period of cycling. For very short periods the yield approaches the yield corresponding to the optimum steady state temperature profiles which, as discussed before, are difficult to implement. For very long periods the yield decreases rapidly because of unfavorable temperature profiles. A very short period, on the other hand, is not desirable because of mechanical difficulties and of material waste during the flow reversal. The calculations summarized in Figure 4 show that for intermediate periods, e.g., 50 to 100 resident times (or about a minute), the average yield is about 90% of the ideal optimum steady state yield. The period can be increased by diluting the catalysts with a solid inert if pressure drop is not an important consideration.**

**The main advantage of regenerative cooling is the achievement of near ideal yields with an operation that can be practically implemented. Its disadvantages are the additional piping and pumping, the larger amount of catalyst for a given throughput, and the varying composition of the product. If the varying composition poses problems in the separation equipment one must use surge tanks or more regenerative reactors in parallel. In any given process, pilot plant and economic studies are needed to determine whether the regenerative system is competitive.**

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## MATHEMATICAL FORMULATION

The material and energy balances for the reactor shown in Figure 1 are written in terms of a forward cell model with two additional assumptions:

1. Transport between bulk fluid and catalyst pellets is not rate limiting, that is, the concentrations and temperatures at the external pellet surface are the same as in the bulk of the fluid.

2. The concentrations are at steady state with respect to the temperature. This assumption is justified in reactions involving gases at ordinary to moderate pressures as the characteristic time for temperature change, $(\rho_s c_{ps}/\rho_g c_{pg})\theta$, is much larger than the residence time $\theta$.

The unsteady operation of the reactor is then described by

$$c_i(m,t) - c_i(m-1,t) = \frac{1-\epsilon}{\epsilon}\,\frac{\sigma(m)}{\overline{\sigma}}\,R_i\,[c(m,t),T(m,t)] \qquad (1)$$

$$\frac{dT(m,t)}{dt} = \frac{\epsilon}{(1-\epsilon)\theta}\,\frac{\rho_g\,c_{pg}}{\rho_s\,c_{ps}}\,[T(m-1,t)-T(m,t)] + \frac{1}{\rho_s\,c_{ps}}\,\frac{\sigma(m)}{\overline{\sigma}}\,R_T\,[c(m,t),\;T(m,t)] \qquad (2)$$

$$m = 1,\ldots,M$$

where $R_i$ is the rate of formation of the $i^{\text{th}}$ species per unit of pellet volume and $R_T$ is the corresponding rate of heat release. The initial conditions are

$$c_i(0,t) = c_{0i} \qquad (3)$$

$$T(0,t) = T_0(t) \qquad (4)$$

$$T(m,0) = T_s(m) \qquad (5)$$

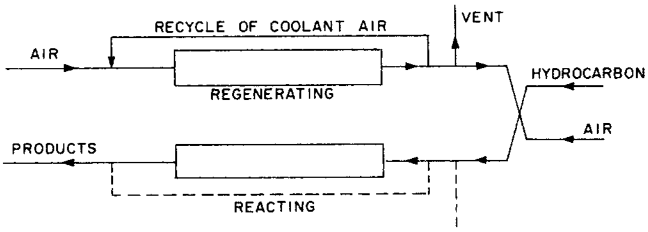


Fig. 1. A regeneratively cooled catalytic reactor system.

where the $c_{0i}$ are constant. The factors $\sigma(m)$ allow the possibility of varying the reactivity along the bed either by using catalyst of varying activity or by mixing the catalyst with an inert solid. By using the factor

$$\overline{\sigma} = \frac{1}{M}\sum_{m=1}^{M}\sigma(m) \qquad (6)$$

we automatically satisfy the constraint of fixed total reactivity.

The problem is to choose $T_s(m)$, $T_0(t)$, and $\sigma(m)$ to maximize the performance index

$$J = \int_0^{t_f} b^T\,c(M,t)\,dt \qquad (7)$$

which is simply a weighted sum of the reactor exit concentrations integrated over a period of operation. The control variables $T_0(t)$ and $\sigma(m)$ are subject to constraints

$$T_* \leq T_0(t) \leq T^* \qquad (8)$$

$$\sigma(m) \geq 0 \qquad m = 1,\ldots,M \qquad (9)$$

It is convenient to rewrite the problem in the notation

$$x(m,t) - x(m-1,t) = \frac{\sigma(m)}{\overline{\sigma}}\,g[x(m,t),\;y(m,t)] \qquad (10)$$

$$\frac{dy(m,t)}{dt} = \beta_0 [y(m-1,t) - y(m,t)] + \frac{\sigma(m)}{\bar{\sigma}} f[x(m,t),\, y(m,t)] \qquad (11)$$

$$m = 1, \ldots, M$$

where $x^T = (x_1, \ldots, x_N)$, $g^T = (g_1, \ldots, g_N)$ are vectors and $y$, $f$ are scalars. The initial conditions are

$$x(0,t) = x_0 \qquad (12)$$

$$y(0,t) = y_0(t) \qquad (13)$$

$$y(m,0) = y_s(m) \qquad (14)$$

The problem now is to determine the control variables $y_0(t)$, $y_s(m)$, $\sigma(m)$ which maximize the quantity

$$J = \int_0^{t_f} b^T\, x(M,t)\, dt \qquad (15)$$

under the constraints

$$y_* \leqq y_0(t) \leqq y^* \qquad (16)$$

$$\sigma(m) \geqq 0 \qquad (17)$$

## CONDITIONS FOR OPTIMAL OPERATION

The necessary conditions for the maximization of $J$ are most conveniently derived by treating the system Equations (10) and (11) as constraints and defining a modified performance index

$$J_1 = \int_0^{t_f} \left[ H(x, y, \mu, \lambda, y_0, \sigma) - \sum_{m=1}^{M} \lambda(m)\, \dot{y}\,(m) \right] dt \qquad (18)$$

where the Hamiltonian $H$ is defined by

$$H(x, y, \mu, \lambda, y_0, \sigma) = b^T\, x(M) + \sum_{m=1}^{M} \left\{ \mu^T(m) \left[ \frac{\sigma(m)}{\bar{\sigma}} g[x(m),\, y(m)] - x(m) + x(m-1) \right] + \lambda(m) \left[ \frac{\sigma(m)}{\bar{\sigma}} f[x(m),\, y(m)] - \beta_0 y(m) + \beta_0 y(m-1) \right] \right\} \qquad (19)$$

Here we have used Lagrange multipliers $\mu(m,t)$ and $\lambda(m,t)$ to take care of the constraints (10) and (11). The quantities $\mu$ and $\lambda$ are also characterized as adjoint variables corresponding to $x$ and $y$.

By taking the first variation of $J_1$ with respect to $x$, $y$, $y_0$ and $\sigma$ and integrating by parts the term $\lambda(m)\, \dot{y}\,(m)$ there is obtained

$$\delta J_1 = \int_0^{t_f} \left\{ \sum_{m=1}^{M} [H_{x(m)} \delta x(m) + [H_{y(m)} + \dot{\lambda}(m)] \delta y(m) + H_{\sigma(m)}\, \delta\sigma(m)] + H_{x(0)}\, \delta x(0) + H_{y(0)}\, \delta y(0) \right\} dt \qquad (20)$$
$$- \sum_{m=1}^{M} [\lambda(m,t_f)\, \delta y(m,t_f) - \lambda(m,0)\, \delta y(m,0)]$$

As usual, we derive equations for the adjoint variables by setting the coefficients of $\delta x(m)$ and $\delta y(m)$ equal to zero as follows:

$$H_{x(m)} = 0 \qquad (21)$$

$$\dot{\lambda}(m) = -H_{y(m)} \qquad (22)$$

Because $x(0,t) \equiv x_0$ is fixed, $\delta x(0) = 0$. On the other hand, $y(m, t_f)$ is free, therefore $\lambda(m, t_f) = 0$. The variation $\delta J_1$ has now been simplified to

$$\delta J_1 = \int_0^{t_f} \left[ \sum_{m=1}^{M} H_{\sigma(m)}\, \delta\sigma(m) + H_{y(0)}\, \delta y(0,t) \right] dt + \sum_{m=1}^{M} \lambda(m,0)\, \delta y(m,0) \qquad (23)$$

The controls $y(m,0) = y_s(m)$ are unconstrained, therefore $\lambda(m,0) = 0$. Similarly, $H_{\sigma(m)} = 0$ for each $m$ where $\sigma(m) > 0$. The initial and boundary conditions for the variables $\mu$, $\lambda$ can now be summarized as

$$\mu(M+1,t) = b \qquad (24)$$

$$\lambda(M+1,t) = 0 \qquad (25)$$

$$\lambda(m,t_f) = 0 \qquad (26)$$

$$\lambda(m,0) = 0 \qquad (27)$$

As $H$ is linear in the control $y(0,t) = y_0(t)$, this control must be "bang-bang" by virtue of the constraints (16):

$$y_0(t) = \begin{cases} y^*, & \lambda(1,t) \geqq 0 \\ y_*, & \lambda(1,t) \leqq 0 \end{cases} \qquad (28)$$

Finally, the necessary condition $H_{\sigma(m)} = 0$ can be written explicitly as

$$\xi(1) = \xi(2) = \ldots = \xi(M) \qquad (29)$$

where

$$\xi(m) = \int_0^{t_f} \{\lambda(m) f[x(m), y(m)] + \mu^T(m)\, g[x(m), y(m)]\}\, dt \qquad (30)$$

## NUMERICAL TECHNIQUE

The set of necessary conditions (10) to (14), (21), (22), and (24) to (29) can be regarded as a two-boundary value problem in the independent variables $m$, ($m = 1, \ldots, M$) and $t$, $0 \leqq t \leqq t_f$. The most economical numerical technique for their solution is the first order gradient method as discussed by Bryson and Ho (5). This consists of the following four steps:

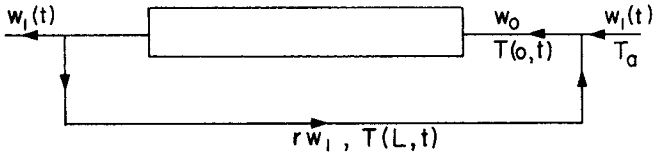
1. Choice of initial estimates for $y_0(t)$, $y_s(m)$, $\sigma(m)$
2. Integration of the state Equations (10) to (14) forward with respect to time and sequentially for $m = 1, 2, \ldots, M$. Integration of the adjoint Equations (21) (22), and (24) to (27) backwards with respect to time and sequentially for $m = M, M-1, \ldots, 1$.
3. Computation of improved controls based on the expression (23):

$$[y_s(m)]_{\text{new}} = y_s(m) + \kappa\, \lambda(m,0) \qquad (31)$$

$$[\sigma(m)]_{\text{new}} = \sigma(m) + \frac{\kappa}{\bar{\sigma}} \left[ \xi(m) - \frac{1}{M\bar{\sigma}} \sum_{k=1}^{M} \sigma(k)\, \xi(k) \right] \qquad (32)$$

with $[y_0(t)]_{\text{new}}$ computed from Equation (28).

4. The steps 1 to 3 are repeated with the new controls.



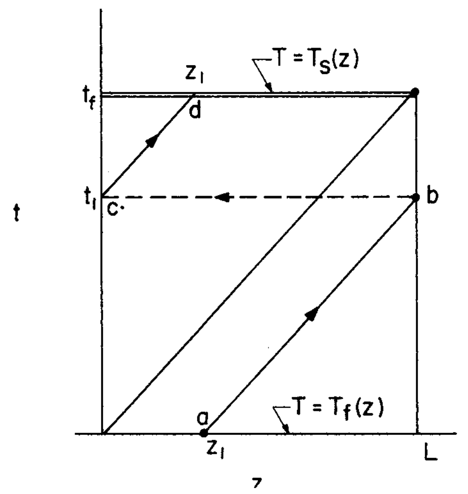


**Fig. 2. Implementation of temperature profile $T_S(z)$ during regeneration.**

The quantity $\kappa$, which determines the length of the step in the gradient direction, is chosen by physical considerations. For example, if $y$ is a dimensionless temperature, $\kappa$ may be chosen initially as

$$\kappa \max \lambda(m, 0) = 0.01 \tag{33}$$

which would give a maximum temperature change of a few degrees. As the iteration proceeds, $\kappa$ may be decreased. The iteration will be terminated when, for some chosen value of $\kappa$, e.g., $\kappa = 0.002$, there is no further improvement of $J$ with succeeding iterations.

## CONTROL IMPLEMENTATION

The control $\sigma(m)$ can be implemented by distributing a mixture of catalyst and inert or, in the case of a supported metal catalyst, by distributing pellets with the appropriate amount of metal.

The control $T_s(m) = T_r\, y_s(m)$ is implemented during the regenerative period which for convenience will be described by the plug flow model

$$\frac{\partial T}{\partial t} + \alpha \frac{\partial T}{\partial z} = 0 \tag{34}$$

where

$$\alpha = \frac{\epsilon\, \rho_g\, c_{pg}\, w_0}{(1-\epsilon)\rho_s\, c_{ps} + \epsilon \rho_g\, c_{pg}} \tag{35}$$

The regenerative cycle is described in Figure 2 where $T_s(z)$ is the desired control to be attained at $t = t_f$, and $T_f(z)$ is the temperature profile at $t = 0$ which is also the final temperature profile in the reactive cycle. The velocity $w_0$ is constant, but the velocity $w_1$ and the recycle ratio $r$ are functions of time which are determined as follows. We first fix $w_0$ by setting $\alpha = L/t_f$ so that the main diagonal in Figure 2 becomes a characteristic. This is not necessary but it results in simpler functions $r(t)$, $w_1(t)$. Following the path *abcd*, we see that the temperature at $b$ is $T_f(a)$, and the temperature at $c$ and $d$ is $[T_a + r\, T_f(z_1)]/(1+r)$, that is,

$$T_s(z_1) = \frac{T_a + r\, T_f(z_1)}{1+r} \tag{36}$$

which gives

$$r(t) = \frac{T_s(z) - T_a}{T_f(z) - T_s(z)} \tag{37}$$

$$w_1(t) = \frac{w_0}{1 + r(t)} \tag{38}$$

where

$$z = \left(1 - \frac{t}{t_f}\right) L \tag{39}$$

Finally, the control variable $T_0(t) = T_r\, y_0(t)$ can be implemented, as shown in Figure 1, by venting part of the air from the regenerator and introducing, if necessary, additional cold air.

### Example: The Oxidation of Benzene

The preceding theory will now be applied to the oxidation of benzene with air on a $V_2O_5$-on-silica catalyst with some data taken from (*6*). The oxidation reactions are all first order and have common activation energy:

$$\begin{array}{ccc} \text{benzene} & \xrightarrow{\quad k_1 \quad} & \text{maleic anhydride} \\ k_2 \searrow & & \swarrow k_3 \\ & CO_2 + H_2O & \end{array} \tag{40}$$

$$k_i = k_i^*\, e^{-E/R_gT} \tag{41}$$

Defining the dimensionless variables

$$x_1 = \frac{c_1}{c_{01}}, \quad x_2 = \frac{c_2}{c_{02}} \tag{42}$$

$$y = \frac{T}{T_r}, \quad a = \frac{E}{R_gT_r} \tag{43}$$

$$\gamma_i = \frac{\theta}{M} \frac{1-\epsilon}{\epsilon} k_i(T_r) \quad (i = 1, 2, 3) \tag{44}$$

$$\gamma = \gamma_1 + \gamma_2 \tag{45}$$

$$\beta_0 = \frac{M\, \rho_g\, c_{pg}\, T_r}{a\, c_{01}\, |\Delta H_1|} \tag{46}$$

$$\beta_1 = \frac{\theta(1-\epsilon)}{a\epsilon} k_1(T_r) \tag{47}$$

$$\beta_2 = \frac{\theta(1-\epsilon)}{a\epsilon} k_2(T_r) \left| \frac{\Delta H_2}{\Delta H_1} \right| \tag{48}$$

$$\beta_3 = \frac{\theta(1-\epsilon)}{a\epsilon} k_3(T_r) \left| \frac{\Delta H_3}{\Delta H_1} \right| \tag{49}$$

$$\beta = \beta_1 + \beta_2 \tag{50}$$

$$\tau = \frac{t}{t_r}, \quad \tau_f = \frac{t_f}{t_r} \tag{51}$$

$$t_r = \frac{(1-\epsilon)\theta}{\epsilon} \frac{\rho_s\, c_{ps}\, T_r}{c_{01}\, |\Delta H_1|\, a} \tag{52}$$

we obtain the following equations for the state variables

$$x_1(m) - x_1(m-1) = -\gamma\, x_1(m)\, e(m) \tag{53}$$

$$x_2(m) - x_2(m-1) = [\gamma_1\, x_1(m) - \gamma_3 x_2(m)]\, e(m) \tag{54}$$

$$x_1(0) = x_{01}; \quad x_2(0) = x_{02} \tag{55}$$

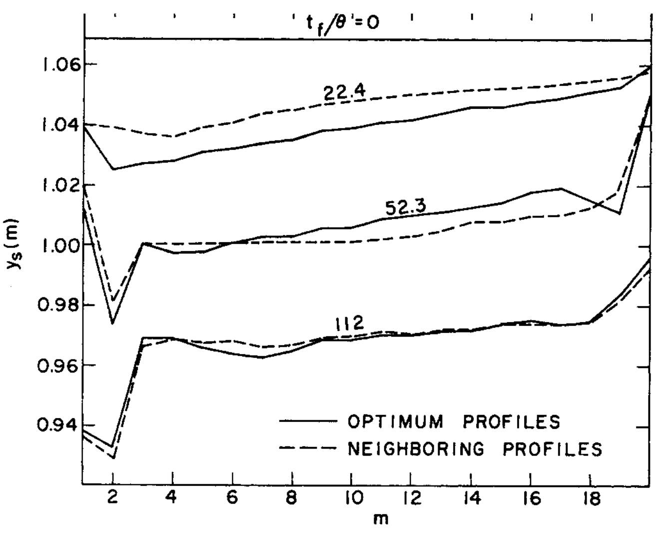


Fig. 3. Optimum initial temperature profiles.

$$\frac{dy(m)}{d\tau} = \beta_0 \left[ y(m-1) - y(m) \right] + \left[ \beta\, x_1(m) + \beta_3\, x_2(m) \right] e(m) \tag{56}$$

$$y(m, 0) = y_s(m) \tag{57}$$

where

$$e(m) = \exp\left[ a - a/y(m) \right] \tag{58}$$

The variables $\sigma(m)$ have been omitted because, as it turned out, the optimal policy was obtained for $\sigma(1) = \sigma(2) = \ldots = \sigma(M)$.

The adjoint equations are obtained by applying Equations (21), (22) as

$$\mu_2(m) = \frac{\mu_2(m+1) + \beta_3\, \lambda(m)\, e(m)}{1 + \gamma_3\, e(m)} \tag{59}$$

$$\mu_1(m) = \frac{\mu_1(m+1) + \gamma_1 \mu_2(m)\, e(m) + \beta\, \lambda(m)\, e(m)}{1 + \gamma\, e(m)} \tag{60}$$

$$\mu_1(M+1) = 0; \quad \mu_2(M+1) = 1 \tag{61}$$

$$\frac{d\lambda(m)}{d\tau} = \beta_0 \left[ \lambda(m) - \lambda(m+1) \right] - \frac{a\, e(m)}{y^2(m)} \{ \lambda(m) \left[ \beta\, x_1(m) + \beta_3 x_2(m) \right] - \gamma\, \mu_1(m)\, x_1(m) + \mu_2(m) \left[ \gamma_1\, x_1(m) - \gamma_3\, x_2(m) \right] \} \tag{62}$$

$$\lambda(m, \tau_f) = 0 \tag{63}$$

$$\lambda(M+1, \tau) \equiv 0 \tag{64}$$

$$\lambda(m, 0) = 0 \tag{65}$$

The following values of physical constants and parameters were used:

$E = 28{,}000$ cal./g.-mole °K.
$\Delta H_1 = 432{,}800$ cal.g.-mole
$\Delta H_2 = 758{,}900$ cal./g.-mole
$\Delta H_3 = 326{,}100$ cal./g.-mole
$\rho_g = 1.472 \times 10^{-3}$ g./cm.$^3$
$c_{pg} = 0.258$ cal./g. °K.
$\epsilon = 0.4$
$\rho_s = 1.0$ g./cm.$^3$
$c_{ps} = 0.2$ cal./g. °K.
$k_1^* = 2.702$ sec.$^{-1}$
$k_2^* = 1.040$ sec.$^{-1}$
$k_3^* = 1.497$ sec.$^{-1}$
$c_{01} = 1.106 \times 10^6$ g.-mole cm.$^{-3}$
$T_r = 648$ °K.
$\theta = 1.00$ sec.
$M = 20$

These result in the following values for the dimensionless groups: $a = 21.746$, $\beta_0 = 0.47282$, $\beta_1 = 0.01195$, $\beta_2 = 0.00807$, $\beta_3 = 0.00499$, $\gamma_1 = 0.01300$, $\gamma_2 = 0.00500$, $\gamma_3 = 0.00720$.

Calculations were performed with $M = 50$ and $M = 20$ to check the dependence of the results on the number of cells in the reactor model. The difference is very small and only the results with $M = 20$ are reported. First, calculations were performed with the initial temperature profile $y_s(m)$ as the only control variable, that is, setting $y_0(\tau) \equiv 1.0$, $\sigma(m)/\bar{\sigma} \equiv 1.0$. The optimal temperature profiles and the optimal yields as functions of the period are shown in Figures 3 and 4. The optimal initial temperature profiles $y_s(m)$ have a dip near the entrance of the reactor followed by a slow rise which is accelerated near the exit. The profiles often exhibit also a secondary dip near the exit. The section of the profile near the exit is much less important with respect to the final yield than the section near the entrance. Thus, the slight dip and the sudden rise near the exit may be an artifact of the numerical method and, in any case, they have a small effect on the product. The dip at the entrance, however, has an important effect.

Figure 5 shows the initial and final temperature profiles corresponding to two policies. The solid line corresponds to the optimal policies described in previous sections, that is, using $y_s(m)$, $m = 1, \ldots, N$ as control variables. The dotted line corresponds to a policy using $y_s(1) = \ldots = y_s(N) = \bar{y}_s$ and optimizing with respect to $\bar{y}_s$. In this

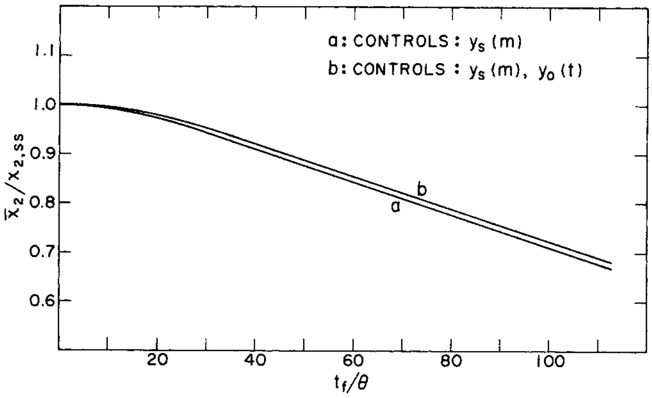


Fig. 4. Optimum yields vs. period.

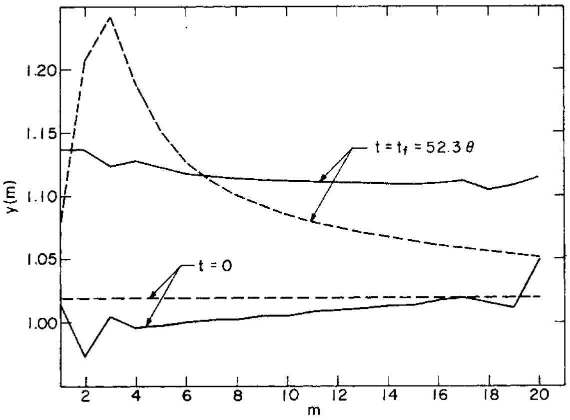


Fig. 5. Final temperatures corresponding to uniform and nonuniform optimum initial temperatures.

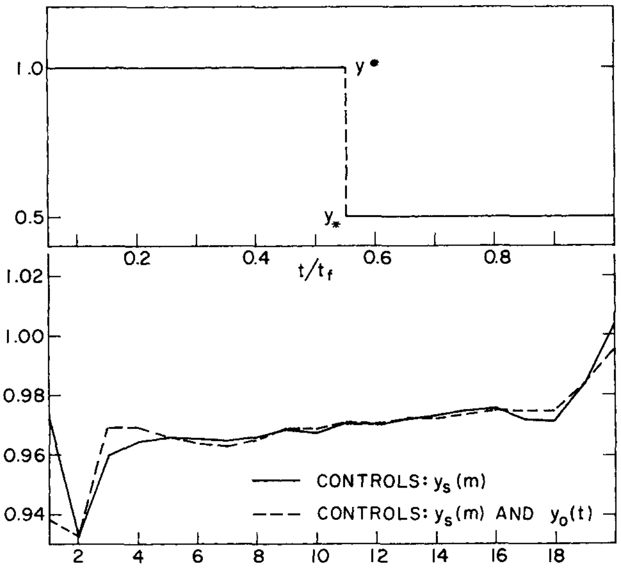


**Fig. 6. Optimum temperature profiles in simultaneous optimization of $y_s(m)$, $y_0(t)$ for $t_f = 52.3$.**

case, the final temperature profile has a strong maximum near the entrance followed by a gradual decrease in temperature. The dip in the optimal policy with nonuniform $y_s(m)$ attempts to cancel the "hot point" and thereby increase the yield. When $t_f/\theta = 52.3$, the use of a constant $y_s$ results in a 5% decrease in $\bar{x}_2$. When $t_f/\theta = 112$, the decrease is 26%.

As the period $t_f$ tends to zero, the yield approaches the optimal steady state yield $x_{2,ss} = 0.4108$. The smooth operation of the pumping system sets a lower limit on the period $t_f$. Moreover, $t_f$ must be much larger than the residence time $\theta$ to keep the gas wasted during the flow reversal at a minimum. In the present example, the value $t_f = 60\theta = 1$ min. appears a reasonable choice. The period can be increased by using a longer bed of a less active catalyst at the cost of increasing the pressure drop.

The period $t_f$ has also an important effect on the sensitivity of the system to small changes in the control variable $y_s(m)$. The broken lines in Figure 3 are profiles which produce a yield about 3% lower than the yield of the neighboring optimum profiles. It is seen that the sensitivity of the yield with respect to $y_s(m)$ decreases with decreasing $t_f$. This is one more advantage of using short periods $t_f$ because the implementation of the control $y_s(m)$ will always involve some error.

By using $y_s(m)$ and $y_0(t)$ simultaneously as control variables, we obtain a small increase in the yield, Figure 4. The control $y_0(t)$ is "bang-bang", and typical profiles $y_s(m)$ and $y_0(t)$ are shown in Figure 6.

Calculations were also performed using $y_s(m)$ and $\sigma(m)$ simultaneously as control variables. The best yield was obtained for uniform reactivity $\sigma(1) = \ldots = \sigma(M)$. Moreover, the yield has been found very insensitive to this control variable. This is probably a result of having used equal activation energies. A related result is that at steady state there is an infinite number of temperature profiles that produce the same yield. In cases where the activation energies of the competing reactions differ, $\sigma(m)$ is expected to be a useful control variable.

## NOTATION

$a$ = constant defined by Equation (43)
$b$ = constant
$c_{0i}$ = concentration of $i^{\text{th}}$ species in feed
$c_i(m, t)$ = concentration of $i^{\text{th}}$ species in $m^{\text{th}}$ cell
$c_{pg}$ = heat capacity of gas
$c_{ps}$ = heat capacity of solid
$E$ = activation energy
$e(m)$ = function defined by Equation (58)
$f(x, y)$ = dimensionless rate function
$g(x, y)$ = dimensionless rate function
$H$ = Hamiltonian function
$I$ = the identity matrix
$J$ = performance index
$k_i$ = rate constant
$k_i^*$ = frequency factor of $k_i$
$L$ = length of fixed bed
$M$ = total number of cells in reactor model
$m$ = cell number
$R_g$ = gas constant
$r(t)$ = recycle ratio
$T(m, t)$ = temperature in $m^{\text{th}}$ cell
$T_a$ = temperature of coolant air
$T_0(t)$ = feed temperature
$T_f(z)$ = final temperature profile in reactor
$T_s(m)$ = initial temperature profile in reactor
$T_r$ = reference temperature
$t$ = time
$t_f$ = duration of a cycle, period
$w_0$ = gas velocity in regenerator
$w_1(t)$ = gas velocity in outer regenerator loop
$x_{0i}$ = dimensionless $c_{0i}$
$x_i(m, t)$ = dimensionless $c_i(m, t)$
$\bar{x}_2$ = average value of $x_2(M, t)$
$x_{2,ss}$ = optimum steady state value of $x_2(M)$
$y(m, t)$ = dimensionless $T(m, t)$
$y_0(t)$ = dimensionless $T_0(t)$
$y_s(m)$ = dimensionless $T_s(m)$
$z$ = position along fixed bed

### Greek Letters

$\alpha$ = constant defined by Equation (35)
$\beta_0, \beta_1, \beta_2, \beta_3$ = constants defined by Equations (46) to (50)
$\gamma_1, \gamma_2, \gamma_3, \gamma$ = constants defined by Equations (44) and (45)
$\epsilon$ = void fraction in fixed bed
$\theta$ = residence time in reactor
$\kappa$ = step size in gradient search
$\lambda(m, t)$ = adjoint variable
$\mu_i(m, t)$ = adjoint variable
$\rho_g$ = density of gas
$\rho_s$ = density of solid
$\sigma(m)$ = relative reactivity in $m^{\text{th}}$ cell
$\bar{\sigma}$ = $[\Sigma\, \sigma(m)]/M$
$\tau$ = dimensionless time
$\tau_f$ = dimensionless $t_f$

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