Prausnitz, J. M., *Molecular Thermodynamics of Fluid-Phase Equilibria*, Prentice-Hall, Englewood Cliffs, N. J. (1969).
Prigogine, I., N. Trappeniers, and V. Mathot, "Statistical Thermodynamics of r-Mers and r-Mer Solutions," *Disc. Farad. Soc.*, **15**, 93 (1953); *J. Chem. Phys.*, **21**, 559 (1953).
Prigogine, I., *The Molecular Theory of Solutions*, North-Holland, Amsterdam (1957).
Rowlinson, J. S., *Liquids and Liquid Mixtures*, 2nd edit., pp. 34, 35, 53, Butterworth, London (1969).
Siow, K. S., G. Delmas, and D. Patterson, "Cloud-Point Curves in Polymer Solutions with Adjacent Upper and Lower Critical Solution Temperatures," *Macromolecules*, **5**, 79 (1972).
Timmermans, J., *Physicochemical Constants of Pure Organic Compounds*, Vol. 1 and 2, Elsevier, Amsterdam (1950, 1965).
van der Waals, J. H., and J. J. Hermans, "Thermodynamic Properties of Mixtures of Alkanes Differing in Chain Length. II. Vapour Pressures," *Rec. Trav. Chim. Pays-Bas*, **69**, 971 (1950).
Walker, E. E., "The Solvent Action of Organic Substances on Polyacrylonitrile," *J. Appl. Chem. (London)*, **2**, 470 (1952).
Wood, S. E., and J. A. Gray, "The Volume of Mixing and the Thermodynamic Functions of Binary Mixtures. II. Cyclohexane-Carbon Tetrachloride," *J. Am. Chem. Soc.*, **74**, 3729 (1952).
Wood, L. A., and G. M. Martin, "Compressibilities of Natural Rubber at Pressures Below 500 KG/CM²," *Rubber Chem. Tech.*, **37**, 850 (1964).

Manuscript received January 22, 1972; revision received April 11 and accepted April 12, 1973.

Optimization of Parallel Reactions in a Tubular Reactor by Feed Distribution

The system of isothermal, isobaric, perfect gas reactions with stoichiometric kinetics

$$A_1 + A_2 \underset{k_2}{\overset{k_1}{\rightleftharpoons}} A_3 + A_4;\ 2A_1 \overset{k_3}{\rightarrow} A_5$$

taking place in a tubular reactor is considered where A_3 is desired and A_5 undesired. A class of optimum selectivity problems is formulated, and distributed bypass of A_1 is investigated. It is shown that there are cases where the performance of the best plug flow reactor can be improved substantially (sometimes by more than 100%) by the bypass of A_1, but there are also cases where bypass is fruitless.

H. T. DELBRIDGE
and
D. C. DYSON

Department of Chemical Engineering
Rice University, Houston, Texas

SCOPE

It is well known that there are cases in which delaying the addition of a reactant to reactor (either in space or time) is accompanied by an improvement in performance. In fact, there are simple tests which can be used to determine whether a given reactor can be improved by infinitesimal bypass: Jackson and Senior (1968), Horn and Tsai (1967). It is not possible to say a priori however whether for a given physical system and objective function there will be significant benefit. The literature on feed distribution may be classified into three categories:

1. Variable volume reactors (for example Siebenthal-Aris (1964), and Graves (1968), who studied a semibatch stirred tank reactor in which the liquid level changes as the reaction proceeds).
2. Fixed volume reactors, for which it is assumed in the model that the volumetric flow rate of the main stream is unchanged by addition of the side stream (for example, the side stream consists of concentrated material which rapidly dissolves in the main stream with negligible volume change: for example, Jackson and Senior (1968); van de Vusse and Voetter (1961)).
3. Fixed volume reactors where it is assumed that the reacting mixture behaves as a perfect gas mixture: (for example, Dyson and Horn (1967), Dyson and Graves (1968)).

Reactors of the second class have probably received the most attention in the literature, and intuitive arguments have been developed to explain qualitatively effects which have been derived accurately by rigorous analysis. These intuitive arguments are of course applicable to all the above classes of reactors but do not lead to good numerical estimates of the magnitude of the benefit resulting from feed distribution any more than do the simple tests referred to above.

The published work on reactors of the third class has been limited to single reactions. In the present manuscript we describe the results of some calculations on the system:

$$\begin{gathered} A_1 + A_2 \underset{k_2}{\overset{k_1}{\rightleftharpoons}} A_3 + A_4 \\ 2A_1 \overset{k_3}{\rightarrow} A_5 \end{gathered} \quad (1)$$

carried out in a tubular reactor with main feed A_1 and subsidiary feed A_2 (see Figure 1). Isothermal, isobaric operation with a perfect gas mixture and stoichiometric kinetics was assumed. The substance A_3 is consided to be the desired product and A_5 an undesirable waste product. This system of reactions is interesting because there is competition for the A_1 molecules between other A_1 molecules and A_2 molecules so that it seems natural to bypass the feed of A_1 in order to suppress the undesired reaction. Also, the first reaction is reversible so that by increasing the reactor

H. T. Delbridge is with the Naval Nuclear Power School, Bainbridge, Maryland 21905.

volume in a conventional plug flow reactor without feed distribution, the yield of products would not necessarily be increased: to see this consider the case with a very large reactor volume, where any A_3 and A_4 formed would recombine to form A_2 and A_1, the latter being irreversibly degraded by the second reaction. (In this respect the system is similar to the system $A \rightarrow B \rightarrow C$ with B the desired product for which it is well known that yield considerations alone lead to an optimal residence time).

Now, it can be shown (for the system studied here) that in contrast to the plug flow reactor without feed distribution (which will be called the single inlet reactor) the yield of A_3 from an optimal distributed feed reactor increases monotonically with reactor volume. (What happens is that the optimal policy calls for more and more delay as the volume is increased so that in a reactor with a large volume practically no A_1 is added until almost the end of the reaction.) For this reason one has to be careful how one compares the distributed feed and single inlet reactors: if for example one made them each large enough, then the distributed reactor would turn out to be arbitrarily superior to the single inlet reactor whose product, by the above argument, would consist almost exclusively of the undesired species A_5.

A series of optimal selectivity problems were formulated for the two reactors: it is felt that considerable insight into the desirability of feed distribution may be gained from the solution to these problems.

CONCLUSIONS AND SIGNIFICANCE

We have reported the results of some optimization calculations aimed at evaluating the desirability of feed bypass of A_1 to an isothermal tubular reactor in which the reactions (1) take place simultaneously, with stoichiometric kinetics, and for which the reacting mixture may be taken to behave as a perfect gas mixture (Figure 1).

We have given in some detail an example indicating feed bypass to be very desirable but have quoted results of Delbridge (1970) in which other examples show feed bypass to be of little use. If one may generalize from somewhat scant information, it appears that the crucial point of distinction concerns the flow rate q_{2R} of A_2 to the reactor. If this can be increased substantially without incurring much increase in cost, then distribution of A_1 is not attractive. On the other hand, if it is expensive to separate the unconverted A_2 from the reactor effluent, feed distribution offers advantages.

BASIS FOR COMPARISON OF DISTRIBUTED FEED REACTOR WITH SINGLE INLET REACTOR

The reactor which is represented in Figure 1 is an isothermal plug flow tubular reactor with no axial dispersion but with perfect mixing in planes normal to the axis. The variable m represents the mass of catalyst to the left of the cross-sectional plane labeled m; m_f is the total mass of catalyst contained in the reactor. The feed stream of A_1 (whose molar flow rate is q_{1R}) may be distributed along the length of the reactor, but the feed stream of A_2 (whose molar flow rate is q_{2R}) may not.

The kinetics of the reaction system are such that the rate of formation of A_3 per unit mass of catalyst is

$$r_1(m) = k_1\gamma_1(m)\,\gamma_2(m) - k_2\gamma_3(m)\gamma_4(m) \qquad (2)$$

and the rate of formation of A_5 per unit mass of catalyst is given by

$$r_2(m) = k_3(\gamma_1(m))^2 \qquad (3)$$

In order to compare the performance of this distributed feed reactor with a single inlet plug-flow reactor we could consider each reactor to have the same mass of catalyst, reaction rate constants, and feed, that is, we could make the feed rates q_{1R} of A_1 and q_{2R} of A_2 fixed constants, the same for each reactor. The single inlet reactor would then be completely specified and we might choose the distribution of A_1 in the distributed feed reactor in such a way as to maximize

$$F = q_3(m_f) - \frac{1}{2}\phi\, q_5(m_f) \qquad (4)$$

where ϕ is a suitable positive constant. F is related to the gross rate of monetary gain M associated with the operation of the reactor as follows:

$$M = (a_3 + a_4 - a_1 - a_2)q_3 + (a_5 - 2a_1)q_5 \qquad (5)$$

and, noting that $a_3 + a_4 > a_1 + a_2$, otherwise the desired reaction would be unprofitable, and also that $a_5 - 2a_1 < 0$ since A_5 is undesired, we may put

$$\phi = (a_1 - 0.5a_5)/(a_3 + a_4 - a_1 - a_2) \qquad (6)$$

whence $\phi > 0$, as noted, and $M = \text{const.} \times F$.

One then would obtain values of F_{max} for reactors of the two types (for various values of ϕ) and compare them. Provided the reaction rate and other parameters which enter into the description of the reactors were not chosen in such a way as to ensure that increasing m_f would lead to a decrease in F for the single inlet reactor, this would appear to be a reasonable comparison. However, it turns out in many cases that the optimal policy for the distributed feed reactor meets the requirement that the total flow rate of A_1 to it, q_{1R}, should be a given constant by dumping a significant amount of this material in at the end of the reactor (where of course it cannot react). Obviously this is merely an indication that the reactor doesn't want this material so the comparison as formulated is unreasonable, and we modify it by requiring that the feed rate of A_1 to the reactor q_{1R} be bounded above by some fixed quantity Q, instead of being specified.

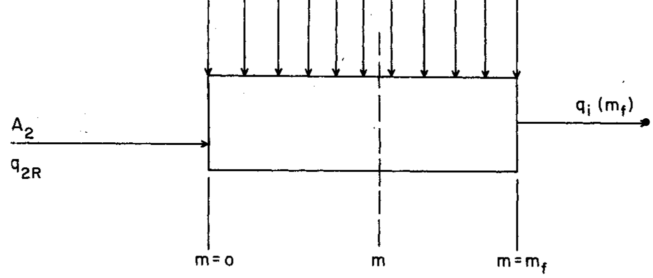

Fig. 1. Isothermal distributed feed reactor with feed of A_2 distributed.

Thus

$$q_{1R} \leqq Q \tag{7}$$

is an inequality constraint which must be satisfied by the distributed feed reactor. Q is some suitably chosen flow rate. We also relax the requirement that q_{1R} be fixed for the single inlet reactor, replacing it by (7).

DIMENSIONLESS DESCRIPTION OF REACTORS

It is convenient to make use of the fixed quantity Q described above as a basis for dimensionless quantities: we define $X_1(m)$, $X_2(m)$, $X_3(m)$ as follows:

$$X_1(m) = q_3(m)/Q; \quad X_2(m) = 2q_5(m)/Q \tag{8}$$

$$X_3(m) = (q_1(m) + q_3(m) + 2q_5(m))/Q \tag{9}$$

$$\theta = q_{2R}/Q, \quad t = k_1 m/Q, \quad \tau = k_1 m_f/Q, \quad \alpha = k_2/k_1, \quad \beta = k_3/k_1 \tag{10}$$

and denote the image of composite mapping $t \to m \to X_i$ by $x_i(t)$, thus $x_i(t) = X_i(Qt/k_1)$, then it follows that the balances for A_3 and A_5:

$$\frac{dq_3}{dm} = r_1(m); \quad \frac{dq_5}{dm} = r_2(m) \tag{11}$$

become

$$\left.\begin{aligned} \frac{dx_1}{dt} &= f_1(x_1, x_2, \theta, h(t)) \\ &= \frac{(h(t) - x_1 - x_2)(\theta - x_1) - \alpha x_1^2}{(h(t) - 0.5x_2 + \theta)^2}; \quad x_1(0) = 0 \\ \frac{dx_2}{dt} &= f_2(x_1, x_2, \theta, h(t)) \\ &= \frac{2\beta(h(t) - x_1 - x_2)^2}{(h(t) - 0.5x_2 + \theta)^2}; \quad x_2(0) = 0 \end{aligned}\right\} \tag{12}$$

where we have replaced $x_3(t)$ by $h(t)$, since it is a control function and thus of a nature distinct from the state functions $x_1(t)$ and $x_2(t)$. (Note: from Equation (9) it can be seen that $QX_3(m)$ is the rate at which A_1 enters the reactor across boundaries to the left of the plane m in Figure 1; $h(t)$ thus determines the feed distribution).

OPTIMIZATION PROBLEMS

The family of the optimal selectivity problems mentioned above may now be precisely formulated.

For the distributed feed reactor we define a class of controls $H(t)$. This is the class of positive, nondecreasing, piecewise continuous functions which are smooth on the open intervals between jumps, whose domain is the interval $[0, \tau]$ and range the interval $[0, 1]$. Any element $h(t) \in H(t)$ will be termed an admissible control. If $x_1(\tau)$, $x_2(\tau)$ are the end values of the state functions which are solutions of (12) with some admissible control $h(t)$, then

$$F = x_1(\tau) - \phi x_2(\tau) \tag{13}$$

is a functional defined over $H(t)$ which we may denote $F\{h(t)\}$. Then, the problem is: given the reactor parameters α, β, τ, control parameter θ, and the economic parameter ϕ, find

$$F_{\max} = \max_{h(t) \in H(t)} F\{h(t)\} \tag{14}$$

and the corresponding control: $h_{\max}(t)$.

For the plug flow reactor the problem is the same but the control set is correspondingly restricted:

$$h(t) = K, \text{ a constant}; \quad K \in (0, 1] \tag{15}$$

This is the main problem we consider. Maximizing F in this problem is equivalent to maximizing M since $F =$ const. $\times M$. Thus in this problem the gross rate of monetary gain is maximized.

SOLUTION OF MAIN OPTIMIZATION PROBLEM

We consider here the case with

$$\alpha = 0.1, \quad \beta = 1, \quad \tau = 10, \quad \theta = 1. \tag{16}$$

Single Inlet Plug Flow Reactor

The Equations (12) were integrated numerically with $h(t) = K$ for values of K in increments of 0.1 to 1.0. Curve A in Figure 2 represents the case $K = 1.0$; the point O corresponds to $t = 0$, and t increases monotonically along A attaining the value $\tau = 10$ at B.

Trajectories for $K = 0.4$ and 0.7 are also shown; again t increases monotonically along these curves from zero at the point O up to their intersection with the Curve C which is the locus of the ends ($t = \tau$) of the trajectories. The later portions of the trajectories for $K = 0.1$, 0.2, 0.3, 0.5, 0.6, 0.8, and 0.9 are shown, the earlier portions having been omitted in order to prevent crowding near O. In each case t increases monotonically from left to right. From this curve C the optimal value of K may be determined for given ϕ, as follows. Consider the case $\phi = 0.9225$ (the reason for this choice will appear later). F then is $x_1 - 0.9225x_2$, so contours of F are lines of slope 1/0.9225 such as the lines G and J shown, and the maximum value

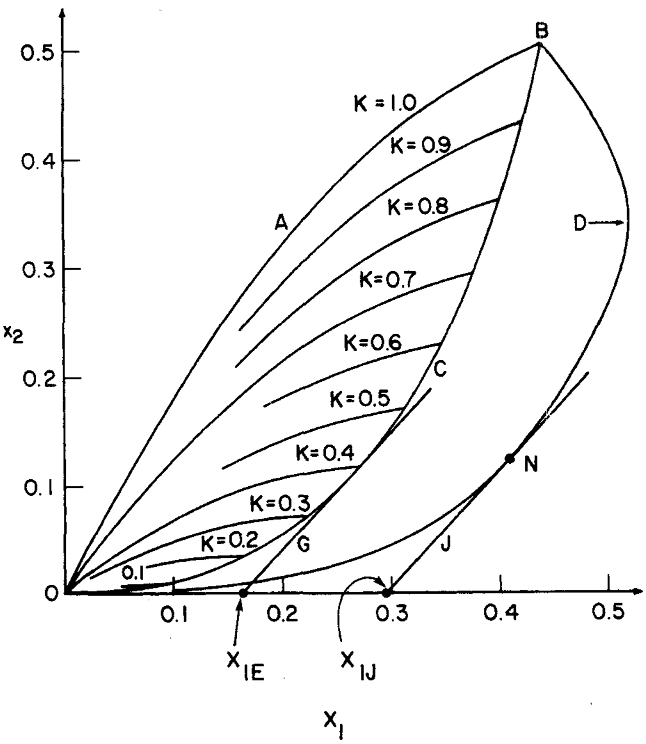

Fig. 2. Single inlet reactor trajectories in the x_1, x_2 plane for K from 0.1 to 1, with terminal locus C. Terminal locus for optimal distributed feed reactor (Curve D) for values of ϕ in the range (−0.772, ∞). All curves correspond to the parameter values given in Equation (16).

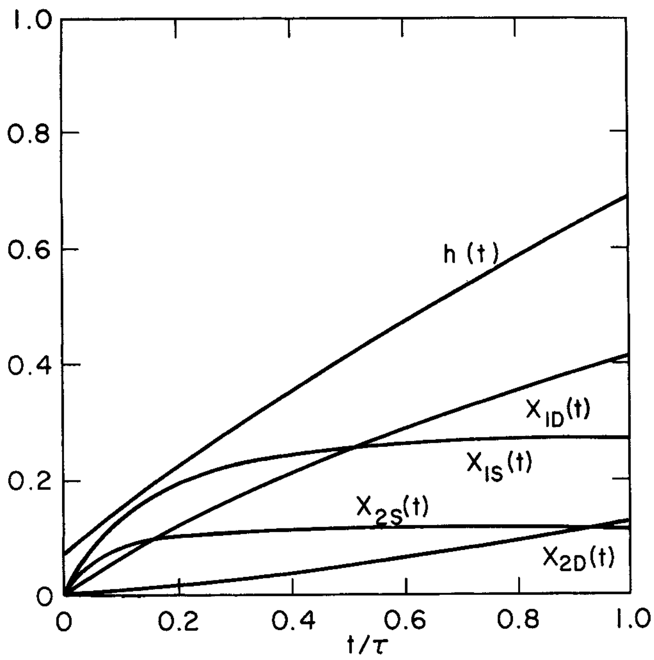

Fig. 3. Optimal trajectories as a function of t/τ for the case ϕ = 0.9225 and parameters given in Equation (16). Key: Suffix D—distributed feed reactor; suffix S—single inlet reactor.

of F is given by the intercept with the x_1 axis of that contour which while lying furthest to the right still intersects C; that is, the contour G. F_{max} then is X_{1E}, approximately 0.1635. By interpolation we find $K \approx 0.395$. The procedure for determining the optimal control for other values of ϕ is essentially the same. For $\phi = 0$ the optimal point is B, and the corresponding $F_{max} = 0.439$.

We note that τ in this example has not been chosen unreasonably large, since, from the figure, it is obvious that by increasing τ slightly an end point locus lying to the right of C everywhere (except of course at the origin O) would have been obtained. The functions $x_1(t)$, $x_2(t)$, corresponding to $K \approx 0.395$ are plotted in Figure 3.

Distributed Feed Reactor

The optimal controls were synthesized by application of an algorithm based on Pontryagin's maximum principle (Pontryagin et al., 1964): the procedure which we will describe later yielded optimal terminal values $x_1(\tau)$, $x_2(\tau)$ together with a value ϕ for which the control is optimal. By adjustment of a parameter in the algorithm different sets of $x_1(\tau)$, $x_2(\tau)$, ϕ were obtained. The optimal terminal values obtained were plotted and a smooth curve drawn through them (curve D in Figure 2). It is clear from the above discussion that the ϕ obtained must correspond to dx_1/dx_2. This is illustrated in Figure 2 by the point N, for which $x_1 = 0.4108$, $x_2 = 0.1245$ and $\phi = 0.9225$. Thus the curve D must be drawn through the point N in such a way that its tangent has slope 1/0.9225. The complete functions $x_1(t)$ and $x_2(t)$ together with the optimal control $h(t)$ corresponding to the point N in Figure 2 are shown in Figure 3.

The portion OD of the curve D in Figure 2 provides us with F_{max} for any value of ϕ in the interval $(0, \infty)$, by merely drawing a tangent to it of slope $1/\phi$ and reading the x_1 axis intercept, as discussed above. The line J which is tangent to D at the point N has intercept 0.2959 (c.f. 0.1635 for the single inlet reactor) indicating an 80% improvement over the best single inlet reactor in this case. For $\phi = 0$, the point D is optimal, and $F \approx 0.52$ showing about 18% improvement over the single inlet reactor. The section DB of the curve is optimal for negative values of ϕ (these are not of much interest but are included to complete the picture). Concerning the controls corresponding to points on the curve D, we have found that for $\phi >> 1$, $h(t)$ is everywhere small (it is better to have no reaction than an unprofitable one); for $\phi = 0.9225$, $h(t)$ is shown in Figure 3. Note $h(\tau) \approx 0.685$. For ϕ approximately zero $h(t)$ is shown in Figure 4. In this case $h(t)$ attains the value 1 after $\approx 60\%$ of the reactor has been traversed. As one moves further along the D curve toward the point B (that is, as one decreases ϕ), the optimal $h(t)$ function moves toward the bound $h(t) \equiv 1$, attaining it for $\phi = -0.772$, corresponding to the point B.

For $\phi > 0.9225$ the improvement in F_{max} on going from single inlet to distributed feed increases steadily: for $\phi = 1.5$ it exceeds 100%.

SYNTHESIS OF OPTIMAL CONTROL FOR DISTRIBUTED FEED REACTOR

There is only one point of difficulty: the requirement that $h(t)$ be nondecreasing. If one ignores this requirement, one is free to apply the Maximum Principle with $h(t)$ considered as the control function: it then usually turns out that the optimal control has the required monotonic property. The maximum principle procedure is as follows:

To Equations (12) are adjoined:

$$\left.\begin{aligned} \dot{\lambda}_1 &= -\lambda_1 \frac{\partial f_1}{\partial x_1} - \lambda_2 \frac{\partial f_2}{\partial x_1} \\ \dot{\lambda}_2 &= -\lambda_1 \frac{\partial f_1}{\partial x_2} - \lambda_2 \frac{\partial f_2}{\partial x_2} \end{aligned}\right\} \quad (17)$$

with boundary condition

$$\lambda_1(0) = 1 \quad (18)$$

and

$$\mathcal{H} = \lambda_1 f_1 + \lambda_2 f_2 \quad (19)$$

$\lambda_2(0)$ is then specified arbitrarily and (12) and (17)

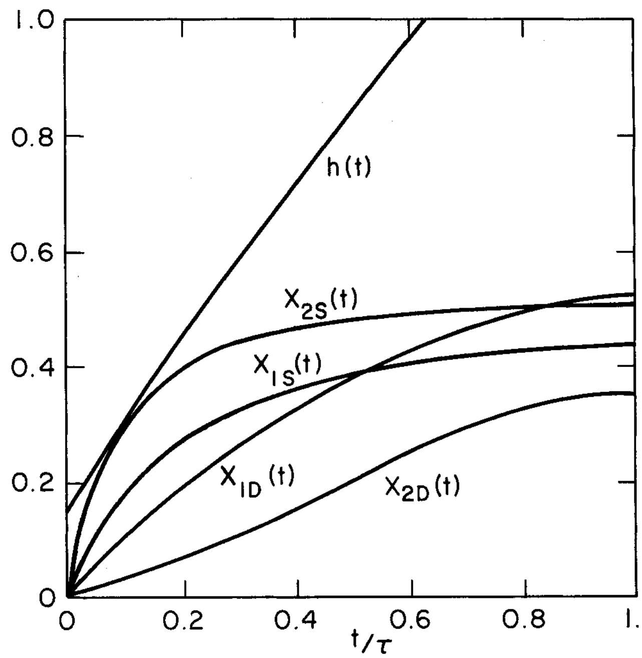

Fig. 4. Optimal trajectories as a function of t/τ for the case ϕ = −0.0594 and parameters given in Equation (16). Key: Suffix D—distributed feed reactor; suffix S—single inlet reactor.

are integrated, choosing $h(t)$ to maximize the Hamiltonian $\mathcal{H}$ at each point.

Then ϕ is given by

$$\phi = -\frac{\lambda_2(\tau)}{\lambda_1(\tau)} \qquad (20)$$

By choosing different values of $\lambda_2(0)$ one obtains trajectories optimal for different values of ϕ. It has been our computational experience that the controls $h(t)$ obtained in this way have an initial increasing portion (which may in fact extend over the whole interval $(0, \tau)$ (for example, Figure 3), followed by a constant portion $h(t) = 1$ (for example, Figure 4) followed sometimes by a downward jump. The maximum principle procedure was modified to overcome this inadmissible downward jump by insisting that if for some value t' of t, the corresponding $h(t)$ took on the value 1, then the control $h(t) \equiv 1$ be applied on $t \epsilon [t', \tau]$. This modified procedure was the algorithm we employed. Other tests were applied to see whether controls generated by the algorithm, (in the cases where the maximum principle procedure called for inadmissible controls) were optimal over the class of admissible controls. (That the controls were not optimal over the class of controls which would allow $h(t)$ to decrease over some sub interval is of course not important.) For these tests, the derivative $v = \frac{dh}{dt}$ was taken as the control and the maximum principle applied. There then arises a singular trajectory which corresponds precisely to the initial increasing portion of the control $h(t)$ described above and departures from this trajectory have to be examined for optimality. Analysis of optimal departure points from singular trajectories is by now quite routine. We found no evidence that the modified maximum principle algorithm described above gave inferior control functions.

SUBSIDIARY OPTIMIZATION PROBLEMS: VARIATION OF θ

We have given an instance of substantial improvement in performance attributable to feed bypass. This was found for a given set of parameters, but all features of the problem depend smoothly on these parameters so this improvement is obviously not isolated.

Some effort was spent in investigating whether feed distribution was worthwhile in a much broader context. [Delbridge (1970) gives details.] The situation will be described briefly. For given α, β, τ, and ϕ, the best single inlet reactor (with $h \equiv 1$) was found by maximizing F with respect to θ. Then the best distributed feed reactor was found (F was maximized simultaneously with respect to the parameter θ and the function $h(t)$). A set of problem parameters corresponding to points in the rectangle in α, β, τ space defined by the ranges [0, 2.5] of α, [2, 100] of β and [1, 5] of τ were selected as problem parameters. The results were uniformly disappointing: the case $\phi = 0$ is typical. In this case the improvement in F in going from best single inlet to best distributed feed reactor was merely 4.13%.

In this study, it was invariably the case, however, that the optimal θ for the single inlet reactor was substantially higher than for the distributed feed reactor, a factor which could lead to offsetting expenditures in a subsequent separation unit.

ACKNOWLEDGMENTS

This work was supported by the following NSF grants: GU 1153 (Systems grant to Rice University), and GK 12522. Supplementary support was provided by NASA.

The authors are grateful to a reviewer, R. R. Hughes, for suggestions which led to improvements in the clarity of this paper.

NOTATION

A_1-A_5 = chemical species 1 to 5
a_i (for $i = 1, 2 \ldots 5$) = cost per mole of the chemical A_i
F = objective function, see Equation (13)
$\bar{F}$ = objective function, see Equation (4)
f_1, f_2 = see Equation (12)
$H(t)$ = set of admissible controls, see text above Equation (13)
$\mathcal{H}$ = Hamiltonian function, see Equation (19)
$h(t)$ = control function, see text below Equation (12)
K = control parameter for single inlet reactor, see Equation (15)
k_1, k_2, k_3 = rate constants
M = gross rate of monetary gain associated with the operation of the reactor
m = see text above Equation (2)
Q = bound on q_{1R}, see Equation (7)
$q_1(m)$, $q_2(m)$, etc. = molal flow rate of species A_1, A_2 etc. at section m of reactor
q_{1R}, q_{2R} = feed rates of A_1 and A_2 to reactor
$r_1(m)$, $r_2(m)$ = rates of formation of A_3 and A_5 respectively; moles/time mass of catalyst
t see Equation (10)
t' see text below Equation (20)
v = alternative control variable, see text below Equation (20)
$X_1(m)$, $X_2(m)$, etc. = see Equation (8)
$x_1(t)$, $x_2(t)$, etc. = see text below Equation (10)

Greek Letters

α see Equation (10)
β see Equation (10)
γ_1, γ_2 etc. = mole fractions of species A_1, A_2, etc.
θ see Equation (10)
λ_1 see Equations (17) and (18)
λ_2 see Equation (17) and text below Equation (19)
τ see Equation (10)
ϕ = economic parameter, see, for example, Equation (6)

LITERATURE CITED

Delbridge, H. T., "Optimization of Chemical Reactors with Feed Distribution," Ph.D. thesis, Rice Univ., Texas (1970).

Dyson, D. C., and J. R. Graves, "Optimum Distributed Feed Reactor with Temperature Constraints," *Chem. Eng. Sci.*, **23**, 435 (1968).

Dyson, D. C., and F. J. M. Horn, "Optimum Distributed Feed Reactors for Exothermic Reversible Reactions," *J. Optim. Theory Appl.*, **1**, 40 (1967).

Graves, J. R., "Optimal Control of Distributed Feed Reactor," Ph.D. thesis, Rice Univ., Texas (1968).

Horn, F. J. M., and M. J. Tsai, "The Use of the Adjoint Variables in the Development of Improvement Criteria for Chemical Reactors," *J. Optim. Theory Appl.*, **1**, 131 (1967).

Jackson, R., and M. G. Senior, "The Optimization of Chemical Reactors by the Delayed Addition of Reactant," *Chem. Eng. Sci.*, **23**, 971 (1968).

Pontryagin, L. S., V. G. Boltyanskii, R. V. Gamkrelidge, and E. F. Mischenko, *The Mathematical Theory of Optimal Processes*, MacMillan, New York (1964).

Siebenthal, C. D., and R. Aris, "The Application of Pontryagin's Methods to the Control of Batch and Tubular Reactors," *Chem. Eng. Sci.*, **19**, 747 (1964).

van de Vusse, J. G., and H. Voetter, "Optimum Pressure and Concentration Gradients in Tubular Reactors," *ibid.*, **14**, 90 (1961).

Manuscript received March 21, 1972; revision received March 20 and accepted April 5, 1973.