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OPTIMAL DESIGN OF SEQUENCE OF CONTINUOUS-FLOW STIRRED-TANK REACTOR WITH CROSS FEEDS

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The optimal design for carring out arbitrary reactions in a sequence of continuous-flow stirred-tank reactors with cross feeds is determined by using the discrete maximum principle.

Concise relations are developed by matrix operation which enable the reactor volume, temperature and cross-feed flow rate at each stage to be chosen such that the total profit is maximized.

A numerical example is solved by means of the iterative solution method by using Green's Tensor.

I. Introduction

Consider, for example, the following two reactions occurring simultaneously in a process.

$$A+B\rightarrow P \quad \text{, with} \quad R_P=k_1C_AC_B$$
$$A+A\rightarrow X \quad \text{, with} \quad R_X=k_2C^2{}_A \qquad (1)$$

P is the desired product and X a waste product, while A is an expensive reactant which cannot be easily removed from the product stream. It is therefore required to obtain a high relative degree of conversion of A together with a relatively high production, *i.e.* high yield of P.

The rate equation indicates that the latter may be achieved by keeping the concentration of A in the reactor system relatively low.

The problem of yield optimization about such a reaction system was illustrated by Denbigh[1] with regard to the production cyclonite from hexamine and nitric acid in the tank reactors as constant temperature by using the differential selectivity.

Similar discussions was made by Trambouze and Piret[2] and by Oden[3].

Van de Vusse and Voetter[4] pointed out that, carrying out such a reactions, a tank reactor gives a higher yield than a tubular reactor, but that even better results can be obtained if a distributed feed of reactant A is properly applied as an additional degree of freedom, and studied strategy of multiple injection of component A for obtaining a high yield of P.

One of these type of reactors is a sequence of continuous-flow stirred-tank reactor with cross-feeds, illustrated in Fig. 1. There are many practical parallel reactions which need some reactants to be injected by means of cross-feeds to maximize the yield of the desired product.

To maximize the total profit, however, reaction capacity must be considered together with the yield optimization, and the cross-feed flow rate, reactor volume and temperature at each stage should be optimized.

In this paper, it is discussed by using the discrete maximum principle to solve this problem with arbitrary complex reaction systems.

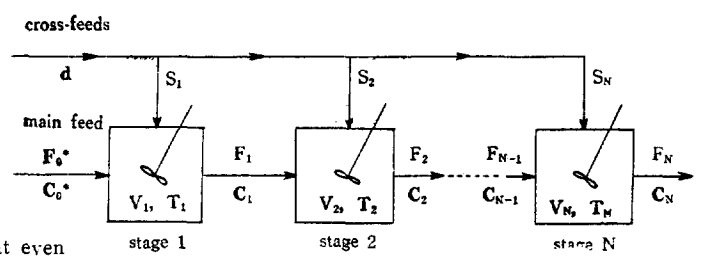

Fig. 1. A sequence of continuous-flow stirred-tank reactors with cross-feeds

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II. Developing Relations

We will denote by C_i and d_i the concentration of the i th substance in the reactor and in the cross-feed, respectively. Let T, V and $R(T, \boldsymbol{C})$ be the reactor temperature, volume and molar rate of production of the i-th substance per unit volume of reactor, and let F and S be the flow rates of main stream and of cross feeds, respectively

If suffix n denotes the values at the n-th stage in Fig. 1, a material balance for the i-th substance and amount of flow rate gives the following equations, supposing densities invariable during the reactions,

$$F_n C_{n,i} - F_{n-1} C_{n-1,i} - S_n d_i - V_n R_{n,i}(T_n, \boldsymbol{C}_n) = 0 \qquad i = 1, 2, \cdots\cdots s \quad (2)$$

$$F_n - F_{n-1} - S_n = 0 \quad (3)$$

where $C_{n,i}, F_n$ are the state variable, S_n, T_n and V_n the decision variable and d_i predetermined values, and s is the number of variables representing concentration, $C_{n,i}$. This system of eq. (2), (3) should satisfy the initial condition

$$C_{0,i} = C_0^*{}_{,i}, \quad F_0 = F_0^* \quad (4)$$

We want to maximize a total profit function of state variables in the last stage,

$$P(\boldsymbol{C}_N, F_N) \quad (5)$$

Now, we are to apply the discrete maximum principle to this system. Observing eq. (2) and (3), however, indicates that the system is somewhat more complicated in evaluating Katz's Hamiltonian function[5] of it, for the transformation equation is not an explicit relation(↗)

$$(\searrow) C_{n,i} = C_{n,i}(\boldsymbol{C}_{n-1}, F_{n-1}, T_n, V_n, S_n) \qquad i = 1, 2, \cdots\cdots s$$
$$F_n = F_n(\boldsymbol{C}_{n-1}, F_{n-1}, T_n, V_n, S_n) \quad (6)$$

, but an implicit one

$$f_{n,i}(\boldsymbol{C}_n, \boldsymbol{C}_{n-1}, F_n, F_{n-1}, T_n, V_n, S_n) = 0$$
$$i = 1, 2, \cdots s+1 \quad (7)$$

For the most cases, it is impossible to transform the former relation into the latter, unless all $R_{n,i}$ in eq. (2) are linear function of the concentrations $\boldsymbol{C}_n$, i.e. the first order reactions.

Difficulty is easily overcome by noting that we need only partial derivatives of H_n for all calculations.

Thus, taking partial derivatives of eq. (7) with respect to some variable q_n (say $C_{n-1,i}$, F_{n-1}, S_n, T_n or V_n) we obtain, by the chain rule of calculations.

$$\sum_{k=1}^{s} \frac{\partial f_{n,i}}{\partial C_{n,k}} \frac{\partial C_{n,k}}{\partial q_n} + \frac{\partial f_{n,i}}{\partial F_n} \frac{\partial F_n}{\partial q_n} + \frac{\partial f_{n,i}}{\partial q_n} = 0$$
$$i = 1, 2, \cdots\cdots s+1 \quad (8)$$

, which will be solved for $\frac{\partial C_{n,i}}{\partial q_n}$ and $\frac{\partial F_n}{\partial q_n}$ to obtain $\frac{\partial H_n}{\partial q_n}$. We then obtain not only Green's Vector[6,7,8] from $\frac{\partial H_n}{\partial C_{n-1,i}}$ and $\frac{\partial H_n}{\partial F_{n-1}}$, but optimal values of S_n, T_n and V_n by setting $\frac{\partial H_n}{\partial S_n}$, $\frac{\partial H_n}{\partial T_n}$ and $\frac{\partial H_n}{\partial V_n}$ equal to zero.

Substituting eq. (2) and (3) for $f_{n,i}$ into eq. (8) and representing the result with matrix notation gives

$$\begin{pmatrix} F_n - V_n \frac{\partial R_{n,1}}{\partial C_{n,1}} & -V_n \frac{R_{n,1}}{\partial C_{n,2}} \cdots\cdots & -V_n \frac{\partial R_{n,1}}{\partial C_{n,s}} & C_{n,1} \\ V_n \frac{\partial R_{n,2}}{\partial C_{n,1}} & F_n - V_n \frac{\partial R_{n,2}}{\partial C_{n,2}} \cdots\cdots & -V_n \frac{\partial R_{n,2}}{\partial C_{n,s}} & C_{n,2} \\ \cdots\cdots & \cdots\cdots & \cdots\cdots & \cdots\cdots \\ -V_n \frac{\partial R_{n,s}}{\partial C_{n,1}} & -V_n \frac{\partial R_{n,s}}{\partial C_{n,2}} \cdots\cdots & F_n - V_n \frac{\partial R_{n,s}}{\partial C_{n,s}} & C_{n,s} \\ 0 & 0 \cdots\cdots & 0 & 1 \end{pmatrix} \begin{pmatrix} \frac{\partial C_{n,1}}{\partial q_n} \\ \frac{\partial C_{n,2}}{\partial q_n} \\ \cdots\cdots \\ \frac{\partial C_{n,s}}{\partial q_n} \\ \frac{\partial F_n}{\partial q_n} \end{pmatrix} = - \begin{pmatrix} \frac{\partial f_{n,1}}{\partial q_n} \\ \frac{\partial f_{n,1}}{\partial q_n} \\ \cdots\cdots \\ \frac{\partial f_{n,s}}{\partial q_n} \\ \frac{\partial f_{n,s+1}}{\partial p_n} \end{pmatrix} \quad \cdots\cdots(9)$$

The matrix convention by which partial derivative of a vector $\boldsymbol{X}$ with respect to another vector $\boldsymbol{Y}$ means matrix

$\left[\frac{\partial X_i}{\partial Y_j}\right]$, that is, $\frac{\partial \boldsymbol{X}}{\partial \boldsymbol{Y}} = \left[\frac{\partial X_i}{\partial Y_j}\right]$, will be used throughout.

Partitioned equation (9) and simplified by the above convention, it yields

$$\begin{pmatrix} F_n I - V_n \mathrm{R}_n \boldsymbol{C}_n \\ 0 \quad 1 \end{pmatrix} \begin{pmatrix} \frac{\partial \boldsymbol{C}_n}{\partial q_n} \\ \frac{\partial F_n}{\partial q_n} \end{pmatrix} = - \frac{\partial \boldsymbol{f}_n}{\partial q_n} \quad (10)$$

, where I is identity matrix and

$$\mathrm{R}_n = \frac{\partial \boldsymbol{R}_n}{\partial \boldsymbol{C}_n} \quad (11)$$

By equation (2) and (3), $\frac{\partial \boldsymbol{f}_n}{\partial q_n}$ yields C_{n-1}, F_{n-1}, S_n, T_n and V_n, separately,

$$-\frac{\partial \boldsymbol{f}_n}{\partial C_{n-1,\,i}}=\begin{bmatrix} F_{n-1} & \boldsymbol{e}_i \\ 0 & \end{bmatrix} \tag{12}$$

, where $\boldsymbol{e}_i$ is the i-th unit vector.

$$-\frac{\partial \boldsymbol{f}}{\partial F_{n-1}}=\begin{bmatrix} \boldsymbol{C}_{n-1} \\ 1 \end{bmatrix} \tag{13}$$

$$-\frac{\partial \boldsymbol{f}}{\partial S_n}=\begin{bmatrix} \boldsymbol{d} \\ 1 \end{bmatrix} \tag{14}$$

$$-\frac{\partial \boldsymbol{f}}{\partial T_n}=\begin{pmatrix} V_n\dfrac{\partial \boldsymbol{R}_n}{\partial T_n} \\ 0 \end{pmatrix} \tag{15}$$

$$-\frac{\partial \boldsymbol{f}}{\partial V_n}=\begin{bmatrix} \boldsymbol{R}_n \\ 0 \end{bmatrix} \tag{16}$$

Substituting eq. (12) into eq. (10), solving and summing up it from $i=1$ to s gives

$$\left.\begin{aligned} \frac{\partial \boldsymbol{C}_n}{\partial \boldsymbol{C}_{n-1}}&=F_{n-1}\,[F_nI-V_n\mathrm{R}_n]^{-1} \\ \frac{\partial F_n}{\partial \boldsymbol{C}_{n-1}}&=0 \end{aligned}\right\} \tag{17}$$

By similar method, substituting eq. (13), (14), (15), and (16) into eq. (10), respectively, we obtain following equations.

$$\left.\begin{aligned} \frac{\partial \boldsymbol{C}_n}{\partial F_{n-1}}&=[F_nI-\mathrm{V}_n\mathrm{R}_n]^{-1}\cdot\,[\boldsymbol{C}_{n-1}-\boldsymbol{C}_n] \\ \frac{\partial F_n}{\partial F_{n-1}}&=1 \end{aligned}\right\} \tag{18}$$

$$\left.\begin{aligned} \frac{\partial \boldsymbol{C}_n}{\partial S_n}&=[F_nI-V_n\mathrm{R}_n]^{-1}\cdot[\boldsymbol{d}-\boldsymbol{C}_n] \\ \frac{\partial F_n}{\partial S_n}&=1 \end{aligned}\right\} \tag{19}$$

$$\left.\begin{aligned} \frac{\partial \boldsymbol{C}_n}{\partial T_n}&=V_n[F_nI-V_n\mathrm{R}_n]^{-1}\frac{\partial \boldsymbol{R}_n}{\partial T_n} \\ \frac{\partial F_n}{\partial T_n}&=0 \end{aligned}\right\} \tag{20}$$

$$\left.\begin{aligned} \frac{\partial \boldsymbol{C}_n}{\partial V_n}&=[F_nI-V_n\mathrm{R}_n]^{-1}\,\boldsymbol{R}_n \\ \frac{\partial F_n}{\partial V_n}&=0 \end{aligned}\right\} \tag{21}$$

We will denote Green's vector by

$$\begin{bmatrix} \omega_n \\ \phi_n \end{bmatrix} \tag{22}$$

F, in which ω_n corresponds to $\boldsymbol{C}_n$ and ϕ_n corresponds to F_n. Then, the adjoint system (9), (10), (11), (5) (6) can be represented, by matrix notation, as

$$\begin{aligned} \begin{bmatrix} \omega_{n-1} \\ \phi_{n-1} \end{bmatrix}&=-\frac{\partial[\boldsymbol{C}_n \quad F_n]^T}{\partial[\boldsymbol{C}_{n-1} \quad F_{n-1}]}\begin{bmatrix} \omega_n \\ \phi_n \end{bmatrix} \\ &=\begin{pmatrix} \dfrac{\partial \boldsymbol{C}_n}{\partial \boldsymbol{C}_{n-1}} & \dfrac{\partial \boldsymbol{C}_n}{\partial F_{n-1}} \\ \dfrac{\partial F_n}{\partial \boldsymbol{C}_{n-1}} & \dfrac{\partial F_n}{\partial F_{n-1}} \end{pmatrix}^T\cdot\begin{pmatrix} \omega_n \\ \phi_n \end{pmatrix} \end{aligned} \tag{23}$$

Substituting eq. (17) and (18) into eq. (23) and solving it gives

$$\omega_{n-1}=F_{n-1}[F_nI-V_n\mathrm{R}_n]^{-T}\omega_n \tag{24}$$

$$\phi_{n-1}=[\boldsymbol{C}_{n-1}-\boldsymbol{C}_n]^T[F_nI-V_n\mathrm{R}_n]^{-T}\,\omega_n+\phi_n \tag{25}$$

or, by eq. (24),

$$\phi_{n-1}=\frac{1}{F_{n-1}}[\boldsymbol{C}_{n-1}-\boldsymbol{C}_n]^T\omega_{n-1}+\phi_n \tag{26}$$

To maximize the profit function $P(\boldsymbol{C}_N,\ F_N)$, the boundary conditions[6] for ω_n and ϕ_n are

$$\left.\begin{aligned} \omega_N&=\frac{\partial P}{\partial \boldsymbol{C}_N} \\ \phi_N&=\frac{\partial P}{\partial F_N} \end{aligned}\right\} \tag{27}$$

From the definition of Hamiltonian function

$$H_n=\begin{pmatrix} \boldsymbol{C}_n \\ F_n \end{pmatrix}^T\cdot\begin{pmatrix} \omega_n \\ \phi_n \end{pmatrix} \tag{28}$$

, its partial derivatives with respect to q_n, say S_n, T_n or V_n, becomes

$$\frac{\partial H_n}{\partial q_n}=\begin{pmatrix} \dfrac{\partial \boldsymbol{C}_n}{\partial q_n} \\ \dfrac{\partial F_n}{\partial q_n} \end{pmatrix}\cdot\begin{pmatrix} \omega_n \\ \phi_n \end{pmatrix} \tag{29}$$

By substituting eq. (19), (20) and (21), separately, into eq. (29) and setting each derivative of H_n equal to zero, the following set of equations representing the optimal condition are obtained.

From $\frac{\partial H_n}{\partial S_n}=0$,

$$[\boldsymbol{d}-\boldsymbol{C}_n]^T[F_nI-V_n\mathrm{R}_n]^{-T}\,\omega_n+\phi_n=0 \tag{30}$$

or, $$\frac{1}{F_{n-1}}\,[\boldsymbol{d}-\boldsymbol{C}_n]^T\omega_{n-1}+\phi_n=0 \tag{31}$$

From $\frac{\partial H_n}{\partial T_n}=0$,

$$\frac{\partial \boldsymbol{R}_n^T}{\partial T_n}\,\omega_{n-1}=0 \tag{32}$$

From $\frac{\partial H_n}{\partial V_n}=0$,

$$\boldsymbol{R}_n^T\omega_{n-1}=0 \tag{33}$$

If able to solving the above eq. (31), (32), and (33) for S_n, T_n, and V_n simultaneously, we can obtain the optimal condition. But, there remains still a problem that, no one of equation (31), (32) and (33) containing decision variable S_n, they can not be solved. Disappearance of S_n results from that it affects the system only by additional form as eq. (2) and (3) shows.

Difficulty is overcome by taking F_n as decision variable in stead of S_n. Thus, after solving them for F_n, T_n and V_n, we obtain S_n by eq. (3).

Eliminating ϕ_n from eq. (31) will make the relations much more simplified and amount of calculation reduced.

Consider two successive stages, asy the n th and the $n-1$ th. If it is assumed that eq. (31) for the n th stage has already been satisfied, by substituting eq. (31) into eq. (26), it is reduced to

$$\phi_{n-1}=\frac{1}{F_{n-1}}\ [\boldsymbol{C}_{n-1}-\boldsymbol{d}]^T\ \omega_{n-1} \tag{34}$$

Substituting eq. (34) into eq. (30) for the $n-1$ th stage and replacing suffix $n-1$ with n gives

$$[\boldsymbol{d}-\boldsymbol{C}_n]^T\{F_n[F_n\mathrm{I}-V_n\mathrm{R}_n\}^{-T}-I]\omega_n=0$$
$$n=1, 2, \cdots\cdots N-1 \tag{35}$$

, which can be used, no longer, for the last stage. For the last stage, by eq. (27), eq. (31) yields

$$\frac{1}{F_{N-1}}\ [\boldsymbol{d}-\boldsymbol{C}_N]^T\omega_{N-1}+\frac{\partial P}{\partial F_N}=0 \tag{36}$$

Now, ϕ_n has been entirely eliminated from the relations, neccessary for calculation. Solving eq. (35) or (36) together with eq. (32) and (33) gives the optimal values of F_n, T_n and V_n.

III. Numerical Example

The following reaction system is employed, showing how to use the relations

$$A \xrightarrow{k_1} B \qquad R_A=-k_1C_A-k_2C^2_A$$
$$2A \xrightarrow{k_2} C \qquad R_B=k_1C_A \tag{37}$$

, with

This reaction is carried out isothermally in a sequence of three continuous-flow stirred-tank reactors of equal volume. C is regarded as a waste product and A as desired product. We wish maximize the reaction capacity as well as the amount of B produced for a given usage of A.

This reaction is one of the parallel reactions which need reactant A to be injected into each reactor by means of cross-feeds to maximize the yield of B, because the reaction $A\rightarrow B$ is of the first order and the reaction $2A\rightarrow C$ of the second order. The main(↗) feed into the first stage is an inert liquid stream, not containing the reactant A, and keeping concentration of it in reactors to be dilute, so as to restrain the reaction $2A\rightarrow C$. But, cross-feed contains the reactant A with concentration d_A.

If suffix A and B denote the values of substance A and B, the state variables at the n th stage are $C_{A,n}$, $C_{B,n}$ and F_n the decision variable is S_n.

The problem with unconstrained output may then be stated as follows: Given d_A and F_0, maximize the total profit function

$$p_BF_3C_{B,3}-p_A(F_3-F^*_0)\ d_A \tag{38}$$

by optimal choice of cross-feed flow rate at each stage, S_1, S_2, S_3, subject to

$$0\leq S_n \tag{39}$$

p_A and p_B are the unit prices of A and B per Kg-mole. Since d_A and F_0 are given and p_A and p_B constants, the maximum of eq. (38) is obtained when objective function

$$P(\boldsymbol{C}_3, F_3)=F_3C_{B,3}-\lambda F_3 \tag{40}$$

is maximized. λ is a constant as following

$$\lambda=\frac{p_A}{p_B}d_A \tag{41}$$

The material balance at the n th stage gives the following equations,

$$\left.\begin{aligned}
&F_nC_{A,n}-F_{n-1}C_{n,n-1}-S_nd_A-V_n(-k_1C_{A,n}\\
&\quad -k_2C^2_{A,n})=0\\
&F_nC_{B,n}-F_{n-1}C_{B,n-1}-V_nk_1C_{A,n}=0\\
&F_n-F_{n-1}-S_n=0 \qquad ,n=1,2,3\\
&C_{A,0}=0, \qquad C_{B,0}=0, \qquad F_0=F_0{}^*
\end{aligned}\right\} \tag{42}$$

These are implicit functions of the form of eq. (7) or eq. (2) and (3).

This reaction system is characterized by matrix

$$\mathrm{R}_n=\begin{pmatrix} -k_1-2k_2C_{A,n} & 0\\ k_1 & 0\end{pmatrix} \tag{43}$$

, where k_1 and k_2 are rate constants indicated eq(37).

Substituting eq. (43) into eq. (24) gives the adjoint system,

$$\begin{pmatrix} W_{A,n-1}\\ W_{A,n-1}\end{pmatrix}=\begin{pmatrix} \dfrac{F_{n-1}}{F_n+V_nk_1+2V_nk_2C_{A,n}} & \dfrac{F_{n-1}V_nk_1}{(F_n+V_nk_1+2V_nk_2C_{A,n})F_n}\\ 0 & \dfrac{F_{n-1}}{F_n}\end{pmatrix}\cdot\begin{pmatrix} W_{A,n}\\ W_{B,n}\end{pmatrix} \tag{44}$$

, which can be simplified as following relation,

$$\frac{W_{B,n-1}}{W_{A,n-1}}=\frac{F_n+V_nk_1+2V_nk_2C_{A,n}}{F_n\left(\dfrac{W_{A,n}}{W_{B,n}}\right)+V_nk_1} \tag{45}$$

and the boundary condition of Green's vector is given, by eq. (27) and (40),

$$W_{A,B}=0, \quad W_{B,3}=F_3 \tag{46}$$

By substituting eq. (43) into eq. (35), we obtain optimal condition for the first stage and the second,

$$C_{A,n}=-\frac{k_1}{2k_2}\left(\frac{W_{B,n}}{W_{A,n}}-1\right), \quad n=1,2 \tag{47}$$

, and for the last stage, by substituting eq. (43) and (40) into eq. (36),

$$F_3=\frac{1}{\lambda}\{V_nk_1(d_A-\lambda)-(V_nk_1+2\lambda V_nk_2)C_{A,3}\} \tag{48}$$

For a given set of numerical values of k_1, k_2, V_n, d_A, F_0^* and λ, solving eq. (42), (45), (46), (47) and (48) simultaneously for F_1, F_2 and F_3, gives us optimal values of S_1, S_2 and S_3. But, this is much complicated problem and can not be solved by straight-run method. Various iterative methods[6), 11), 12)] were employed to solve such a problem. The one chosen here is the iterative solution method by using Green's Tensor[6), 7), 8)], which is used to improve the assumed last stage state variables for the next-iteration.

Green's Tensor of this problem is given by multiplying the transformation matrix of eq. (44) for $n=3$, by the matrix for $n=2$ and by the matrix for $n=1$.

In general cases, by using Green's Tensor, an improved values of $\boldsymbol{C}_N$ is given by

$$\boldsymbol{C}_N=\bar{\boldsymbol{C}}_N+rG\cdot[\boldsymbol{C}_0^*-\bar{\boldsymbol{C}}_0] \tag{49}$$

, where Green's Tensor is

$$G=F_0F_1\cdots F_{N-1}[F_1I-V_1\mathrm{R}_1]^{-T}\ [F_2I-V_2\mathrm{R}_2]^{-T}\cdots\cdots [F_NI-V_N\mathrm{R}_N]^{-T} \tag{50}$$

, $\bar{\boldsymbol{C}}_N$ is the previously assumed values of $\boldsymbol{C}_N$

, $\bar{\boldsymbol{C}}_0$ is the calculated $\boldsymbol{C}_0$ started with $\bar{\boldsymbol{C}}_N$

and r is a parameter controlling convergence. In this example eq. (49) yields, to give an improved value of $C_{A,3}$,

$$C_{A,3}=\bar{C}_{A,B}+r(C^*_{A,0}-\bar{C}_{A,0})\cdot \prod_{n=1}^{3}\frac{F_{n-1}}{F_n+V_nk_1+2Vk_2C_{A,n}} \tag{51}$$

Because of the irreversibility of this reaction system, we may assume only $C_{A,3}$, not $C_{B,3}$. Thus, value of $C_{A,3}$ is first assumed. The corresponding value of F_3 is then calculated by eq. (48).

From this value, F_3, together with the assumed value, $C_{A,3}$, $W_{B,2}/W_{A,2}$ is calculated using eq. (45). The corresponding value of $C_{A,2}$ is then computed using eq. (47) and then F_2 using eq. (42), by similar method, $W_{B,1}/W_{A,1}$, $C_{A,1}$ and F_1 are computed. Then, $C_{A,0}$ is calculated by using the initial condition $F_0=F_0^*$ and compared with the initial condition-value, $C^*_{A,0}=0$. When the calculated $C_{A,0}$ differs from the initial condition-value, using Green's Tensor gives an improved value of $C_{A,3}$ for the next iteration. This procedure is repeated until the calculated value of $C_{A,0}$ agrees with the initial condition-value, $C^*_{A,0}=0$, within allowable error limit.

It should be pointed out that as S_n's are nonnegative values, eq. (39), F_n's must satisfy the following restriction

$$F_0^*\leq F_1\leq F_2\leq F_3 \tag{52}$$

Therefore, the optimal value of F_n is given by

$$F_n=\begin{cases} F_{n+1}, & \text{if } F_n\geq F_{n+1} \\ F_n, & \text{if } F_0^*\leq F_n\leq F_{n+1} \\ F_0^*, & \text{if } F_0^*\geq F \end{cases} \tag{53}$$

which gives the optimal S_n by eq. (3).

The following numerical values were taken in the computation;

$V_nk_1=10$ $\quad M^3\mathrm{min}^{-1}$

$V_nk_2=10$ $\quad M^6\mathrm{min}^{-1}\ \mathrm{Kg\text{-}mole}^{-1}$

$F_0^*=1$ $\quad M^3\mathrm{min}^{-1}$

$d_A=5$ $\quad M^{-3}\mathrm{Kg\text{-}mole}$

$\lambda=3$ $\quad M^{-3}\mathrm{Kg\text{-}mole}$

During computation, the value of r was chosen to be 1 for iteration 1 to 10, to be 2 for iteration 11 and 12, and to be 4 for iteration 13 to 18, to control convergence. After twenty iterations, optimal condition was obtained as following;

$S_1=0.9230$ $\quad S_2=0.8572$ $\quad S_3=0.2804$

Table 1. Calculation assuming $C_{A,3}=0.1$

No. of iteration	$C_{A,3}$	S_1	S_2	S_3	$C_{A,0}$	r
1	0.1000	1.2900	1.0443	0.0000	−5.5000	1
2	0.1109	2.1101	0.8835	0.0854	−5.4292	1
3	0.1214	1.9251	0.8772	0.1318	−4.5982	1
4	0.1300	1.5888	0.8720	0.1725	−3.0492	1
5	0.1340	1.4798	0.8695	0.1907	−2.5510	1
6	0.1389	1.3723	0.8661	0.2083	−2.0610	1
7	0.1410	1.2900	0.8653	0.2215	−1.6820	1
8	0.1431	1.2330	0.8640	0.2307	−1.4210	1
9	0.1450	1.1814	0.8629	0.2390	−1.1850	1
10	0.1464	1.1495	0.8620	0.2452	−1.0432	1
11	0.1490	1.0731	0.8604	0.2565	−0.6892	2
12	0.1505	1.0392	0.8595	0.2562	−0.5238	2
13	0.1530	0.9700	0.8525	0.2742	−0.2173	4
14	0.1540	0.9378	0.8574	0.2781	−0.0699	4
15	0.1542	0.9324	0.8569	0.2790	−0.0450	4
16	0.1544	0.9270	0.8571	0.2798	−0.0202	4
17	0.1545	0.9243	0.8571	0.2802	−0.0078	4
18	0.15454	0.92324	0.85708	0.28041	−0.00283	4
19	0.15455	0.92297	0.85708	0.28046	0.00160	—
20	0.15456	0.92237	0.85746	0.28044	0.00309	

$C_{A,3}=0.15455$ $C_{B,3}=2.51495$

The convergence and numerical solution were shown in Table 1 and Fig. 2. If r had been chosen to be 4 through all iterations, the optimal condition would have been obtained more swiftly.

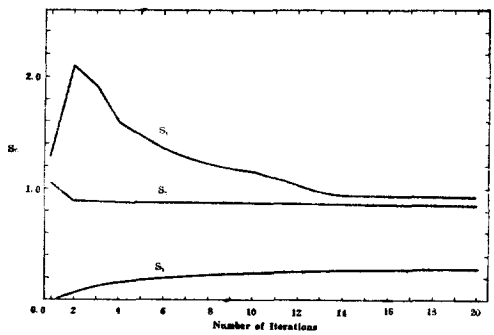

Fig. 2. Interaction Tranjactories

Conclusions

The discrete maximum principle may be used to determine the optimal design for carrying out any arbitrary reaction in a sequence of continuous-flow stirred-tank reactors with cross-feeds.

The method allows the design variables (reaction temperature, reactor volume and cross-feed flow rate of each stage) to be chosen such that the maximum profit is obtained for specified feed flow rate and feed concentration. This method may be applied to optimal design of the sequence of reactors without cross feeds, only putting $S_n=0$ in relations, and to the problem of tubular reactors, looking upon the continuous case as the limit of the discrete when the number of stages approaches infinity, whether with cross-feeds or not.

This paper illustrates how the relations are concisely obtained by matrix operation, and how Green's Tensor technique may be employed to obtain numerical solution of the difference equations which result when the discrete maximum principle is employed.

Nomenclature

$\boldsymbol{C}_n$: concentrations in stage n (state vector):component, $C_{n,i}$

$\boldsymbol{C}_0^*$: feed concentrations (constant vector): component, $C_{0,i}^*$

$\bar{\boldsymbol{C}}_N$: assumed values of $\boldsymbol{C}_N$, component $\bar{C}^*_{N,i}$

$\bar{\boldsymbol{C}}_0$: calculated values of $\boldsymbol{C}_0$ assuming $\boldsymbol{C}_N=\bar{\boldsymbol{C}}_N$:component, $\bar{C}_{0,i}$

$\boldsymbol{d}$: cross-feed concentrations (constant vector): component, d_i

$\boldsymbol{e}_i$: the i th unit vector

F_n : main flow rate from stage n(state variable)

F_0^*: main feed flow rate (constant)

$\boldsymbol{f}_n$: implicit transformation function

G : Green's Tensor, defined in eq. (50)

H_n : Hamiltonian function for stage n

I : identity matrix

k_i : reaction rate constant of the i th substance

P : total profit function (objective function)

p_i : unit price per mole of the i th substance

q_n : some variable

$\boldsymbol{R}_n$: reaction rate (vector function of $\boldsymbol{C}_n$, T_n);component, $R_{n,i}$

R_n : characteristic matrix for stage n, defined in eq. (11)

r : parameter in eq. (49)

S_n : cross-feed flow rate into stage n (decision variable)

s : number of state variables, $C_{n,i}$

T_n : temperature of stage n (decision variable)

V_n : reactor volume of stage n (decision variable)

ω_n: Green's vector to $\boldsymbol{C}_n$: component, $W_{n,i}$

$\boldsymbol{X}$: some vector

$\boldsymbol{Y}$: some vector

λ : constant in eq. (40)

ϕ_n : one component Green's vector to F_n

suffix

A : substance A

B : substance B

i : the i th substance

n : stage n

o : feed

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