Tangent Cone Analysis of Periodic Processes

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Recently considerable interest has been focused on the possibility of improving process performance by intentional unsteady state operation, especially periodic operation. A significant improvement in performance is possible only if time-average outputs can be obtained from the dynamic processes which are not realizable in conventional steady state operation. A more stringent requirement on the dynamic process is that it produces average outputs which are unobtainable from any number of parallel steady state processes.

In order to test a periodic process against these criteria, it is useful to restate them in precise mathematical terms. Then, various mathematical methods can be utilized to evaluate the potential advantages of unsteady operation. The objective of this communication is to report a new technique for this purpose and to compare it with already established procedures.

In several earlier papers, attainable sets have been utilized to compare proper (time-varying) periodic operation of a process with steady state process performance (*1*, *3*). It has been shown that in many instances any average objective vector in $co\ S_s$ may be attained by sufficiently slow cycling or by mixing the outputs of several processes operated in parallel at different steady states. Here the term "average objective vector" refers to the vector of time-average quantities (flows, concentrations, temperatures, etc.) which determine process performance. S_s is the steady state attainable set and $co\ S_s$ is its convex hull. The components of $\overline{y}$ span a space P, which will be called the objective space.

Consequently, it is of interest to determine when cyclic operation of the process yields points outside of $co\ S_s$. That is, conditions are sought which insure that $co\ S_s$ is a proper subset of S_p, the set attainable under all admissible periodic operations

$$co\ S_s \subset S_p \tag{1}$$

This condition has been verified in several previous studies by employing Pontryagin's Maximum Principle (*6*) and by investigating relaxed steady states of the process (*5*). The greater power of the latter approach is proven in (*2*). Both of these methods are useless, however, if the process equations are linear in the control. In this case, the method of tangent cone analysis can sometimes be utilized to prove the validity of condition (*1*).

TANGENT CONE ANALYSIS

Tangent cone analysis is similar to the relaxed steady state approach in that limiting cases corresponding to sequences of control policies are investigated in the P space. However, tangent cone analysis is concerned with limiting directions whereas limit points in the P space are located by relaxed steady state calculations. The tangent cone concept is intimately related to the theory of Maximum Principle (*7*).

In order to define the tangent cone, it is first necessary to introduce the notion of a directionally convergent sequence. Following Hestenes (*4*), a sequence $\{\overline{y}_j\}$ of points in P is said to converge directionally to $\overline{y}_0 \in P$ if

$$\overline{y}_j \neq \overline{y}_0 \quad j = 1, 2, \ldots \tag{2a}$$

$$\lim_{j\to\infty} |\overline{y}_j - \overline{y}_0| = 0 \tag{2b}$$

$$\lim_{j\to\infty} \frac{\overline{y}_j - \overline{y}_0}{|\overline{y}_j - \overline{y}_0|} = h \tag{2c}$$

where h is a unit vector. The orientation of h determines what will be called a limiting direction. Now consider a set $S \subseteq P$ with $\overline{y}_0 \in S$. The tangent cone of S at $\overline{y}_0$, which will be denoted by $C[\overline{y}_0, S]$ is the cone generated by the set of all unit vectors h which are the limits in the sense of (*2*) of directionally convergent sequences $\{\overline{y}_i\}$ of points in S. That is, if the unit vector $\hat{h}$ is the limit in the sense of (*2*) of a sequence of $\overline{y}_i \in S$, then $p\hat{h} \in C[\overline{y}_0, S]$ for all real scalars $p \geqq 0$. If no directionally convergent sequences of elements of S exist at $\overline{y}_0$, $C[\overline{y}_0, S]$ is the null cone.

Condition (*1*) is proven true if it can be shown that the limiting direction in P space corresponding to some sequence of dynamic periodic controls is not contained in the tangent cone of $co\ S_s$ at some $\overline{y} \in co\ S_s$; that is,

$$C[\overline{y}, S_p] \supset C[\overline{y}, co\ S_s] \Rightarrow S_p \supset co\ S_s\,. \tag{3}$$

This fact follows from the definition of the tangent cone. If some half-ray not belonging to $C[\overline{y}, co\ S_s]$ can be approached arbitrarily closely by realizable average rates, a rate not in $co\ S_s$ must be attainable. The use of limiting directions is especially convenient in the neighborhood of the origin of P space. Although all rates approach zero as the origin is approached, the ratios of the rates which define a direction in P space do not always approach zero. One example with dynamics linear in the control which illustrates this is presented in (*3*). Another one will be discussed in the next section.

ILLUSTRATIVE EXAMPLE INVOLVING CATALYST SELECTIVITY

Suppose that the reactions

$$R \underset{2}{\overset{1}{\rightleftarrows}} A_1 \tag{4a}$$

$$R + A_1 \overset{3}{\rightarrow} 2P_1 \tag{4b}$$

$$R \underset{5}{\overset{4}{\rightleftarrows}} A_2 \tag{4c}$$

$$R + A_2 \overset{6}{\rightarrow} 2P_3 \tag{4d}$$

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occur on an isothermal catalyst surface and that the concentration of R at this surface can be controlled. Here A_1 and A_2 denote intermediates absorbed on different active sites. It will be assumed that all of the above reactions are elementary so that the kinetics follow directly from Equations (4).

Admissible R concentrations will be those between zero and a maximum value m. A dimensionless R concentration, dimensionless time, and dimensionless kinetic parameters are defined as follows:

$$u = C_R/m, \quad t = t_{\text{real}}\, k_3 m \quad \beta = \frac{k_2}{k_3 m}$$
$$\xi = k_5/k_6 m \quad \chi = \frac{k_5}{k_3 m}, \tag{5}$$

while the following dimensionless time-average production rates of P_1 and P_2 are introduced

$$\bar{y}^1 = \frac{k_3}{k_1 m \tau} \int_0^\tau C_R(t)\, C_{A_1}(t)\, dt \tag{6a}$$

$$\bar{y}^2 = \frac{k_5}{k_4 m^2 \tau} \int_0^\tau C_R(t)\, C_{A_2}(t)\, dt \tag{6b}$$

It is easy to verify that the steady state attainable set S_s for this system is given by

$$S_s = y_s(U), \tag{7}$$

where

$$y_s(u) = \begin{bmatrix} \dfrac{u^2}{u+\beta} \\ \dfrac{u^2}{1+u/\xi} \end{bmatrix} \tag{8}$$

and

$$U = [0, 1] \tag{9}$$

The notion of a tangent cone will now be illustrated using S_s and its convex hull. If a set is a differentiable surface, the tangent cone of that set at a point in the surface is the tangent hyperplane at that point. Therefore, the tangent cone of S_s in Equation (7) at the origin $C\,[0, S_s]$ is the line given by

$$\bar{y}^2 = \begin{bmatrix} \dfrac{\dfrac{d\bar{y}_s^2}{du}}{\dfrac{d\bar{y}_s^1}{dy}} \end{bmatrix} \bar{y}^1 = \beta \bar{y}^1 \tag{10}$$

That the tangent cone is a more general concept than a tangent plane can be seen by considering $C\,[0, co\, S_s]$. Since $S_s \subseteq co\, S_s$, it is clear that the line $C\,[0, S_s]$ is contained in $C\,[0, co\, S_s]$. Also, supposing that $\{b_i\}$ is a sequence of positive scalars all less than unity which converge to zero, the sequence $\{b_i \bar{y}\}$ is a directionally converging sequence of points in $co\, S_s$ for any $\bar{y} \in S_s$. The elements of $C\,[0, co\, S]$ corresponding to the limiting direction determined by such a sequence are all points on the half-ray emanating from the origin through $\bar{y}$. The union of all such lines with $C\,[0, S_s]$ gives $C\,[0, co\, S_s]$, as can be seen in Figure 1.

Next a proper periodic control will be imposed. In particular, the average production rates obtained from the periodic bang-bang control

$$u(t) = \begin{cases} 1 & t \in [0, \gamma\tau) \\ 0 & t \in [\gamma\tau, \tau) \end{cases} \tag{11a}$$

$$u(t) = u(t+\tau); \quad t \geqq 0, \tag{11b}$$

will be examined. The system dynamical equations and rates resulting from such a control are given elsewhere (*1*). The dynamic equations are linear in u. The quantity of greatest interest here is their ratio, which is given by

$$s(\gamma, \tau) \equiv \frac{\bar{y}^2(\gamma,\tau)}{\bar{y}^1(\gamma,\tau)}$$
$$= \frac{\xi(1+\beta)}{(1+\xi)} \left\{ \frac{\gamma - \dfrac{\xi\eta^*(\gamma,\tau)}{(1+\xi)\chi\tau}\left[1 - e^{-\left(\frac{1+\xi}{\xi}\right)\gamma\chi\tau}\right]}{\gamma - \dfrac{\eta(\gamma,\tau)}{(1+\beta)\tau}\,[1 - e^{-(1+\beta)\gamma\tau}]} \right\} \tag{12a}$$

where

$$\eta = \frac{1 - e^{-(1-\gamma)\beta\tau}}{1 - e^{-(\beta+\gamma)\tau}} \tag{12b}$$

and

$$\eta^* = \frac{1 - e^{-(1-\gamma)\chi\tau}}{1 - e^{-\left(\frac{\xi+\gamma}{\xi}\right)\chi\tau}} \tag{12c}$$

In order to define a limiting direction, a sequence of periodic controls of the form (11) will be defined. The period τ is fixed at a positive value, and a sequence of γ values converging to zero from above is considered. Both $\bar{y}^1$ and $\bar{y}^2$ approach zero as γ does, but their ratio s has a finite limiting value. The resulting limiting direction is obtained by two successive applications of L'Hospital's rule to Equations (12), which yields

$$s_0(\tau) = \chi \frac{\tanh(\beta\tau/2)}{\tanh(\chi\tau/2)} \tag{13}$$

where

$$s_0(\tau) \equiv \lim_{\gamma \to 0^+} s(\gamma, \tau) \tag{14}$$

Since s defines a limiting direction for a particular sequence of periodic controls, the cone C^* with elements

$$\begin{pmatrix} \bar{y}^1 \\ \bar{y}^2 \end{pmatrix} = \begin{pmatrix} \alpha \\ \alpha s_0(\tau) \end{pmatrix} \quad ; \quad \alpha \geqq 0, \ \tau > 0 \tag{15}$$

is a subset of the tangent cone of S_p at the origin

$$C^* \subseteq C\,[0, S_p] \tag{16}$$

Now, if it can be shown that the set $C^* - C\,[0, co\, S_s]$ is not empty, then the existence of average objective vectors not belonging to the convex hull of the steady state attainable set is proven.

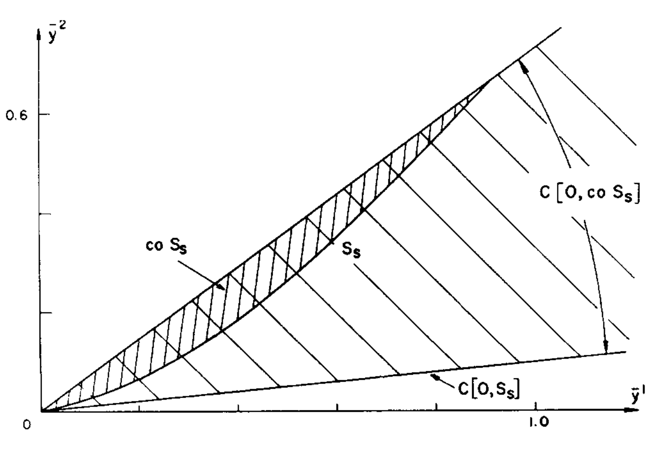

Fig. 1. Tangent cones in P space ($\xi = 2$, $\beta = 0.1$)

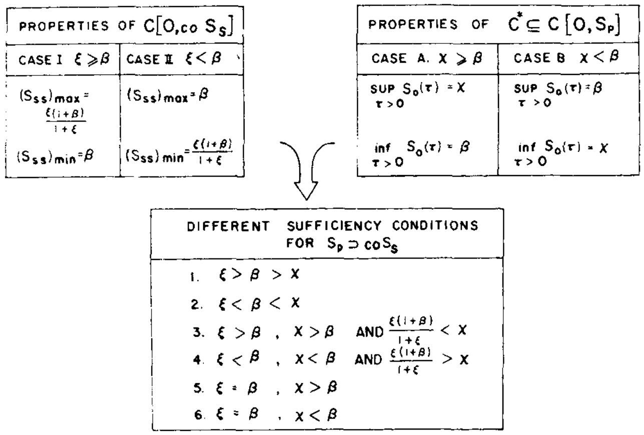

Fig. 2. Application of tangent cone analysis to example.

To locate the boundary of C^*, the supremum and infinum of s_0 must be determined. This can be accomplished by first writing s_0 in the form

$$s_0(\tau) = \chi \frac{\sinh\left(\frac{\beta+\chi}{2}\tau\right) + \sinh\left(\frac{\beta-\chi}{2}\tau\right)}{\sinh\left(\frac{\beta+\chi}{2}\tau\right) - \sinh\left(\frac{\beta-\chi}{2}\tau\right)} \quad (17)$$

Differentiation of this expression gives

$$\frac{ds_0(\tau)}{d\tau} = \frac{2\chi \sinh\left(\frac{\chi+\beta}{2}\tau\right)\sinh\left(\frac{\chi-\beta}{2}\tau\right)}{\left[\sinh\left(\frac{\chi+\beta}{2}\tau\right) + \sinh\left(\frac{\chi-\beta}{2}\tau\right)\right]^2} \quad (18)$$

Since χ, β, and τ are all positive parameters and sinh is an odd function, it follows that

$$sgn \frac{ds_0(\tau)}{d\tau} = sgn\ (\chi - \beta) \quad (19)$$

Equation (19) indicates that for given unequal values of χ and β, s_0 is either monotone increasing or decreasing in τ. (The case $\chi = \beta$ is clearly of little interest, since Equations (10) and (13) reveal that $C^* = C\ [0, S_s]$ in this instance.) The extreme values of s_0 may therefore be obtained by considering the limiting cases $\tau \rightarrow 0$ and $\tau \rightarrow \infty$. From the earlier discussion of $C\ [0, co\ S_s]$ and Equation (8), the boundaries of the tangent cone of $co\ S_s$ at the origin follow in a like manner.

The limiting directions which determine these boundaries are summarized in Figure 2 for all cases. From these results several different possible sufficient conditions for the validity of Condition (1) for this example can be obtained immediately. These conditions are also listed in Figure 2. Although Condition (1) could have been verified directly here by simply evaluating Equation (12) for various values of γ, τ, β, ξ and χ, tangent cone analysis gives simple sufficient conditions in terms of the system parameters alone.

DISCUSSION

The example discussed above and an earlier one (*3*) reveal that tangent cone analysis can be useful even when the system dynamics are linear in the control. In this sense it is a stronger technique than relaxed steady state analysis, which in turn is stronger than Maximum Principle analysis (*2*), for verifying relation (1). However, a systematic procedure for conducting tangent cone analysis is difficult to formulate, and special techniques may be required in each new application. Also, only movement of small magnitude away from $co\ S_s$ under cycling can be guaranteed by tangent cone analysis. This limitation exists because in the usual application of tangent cone analysis, a directionally convergent sequence of elements is considered which also converges to a point in $co\ S_s$. On the other hand, the attainability of average objective vectors far away from $co\ S_s$ can sometimes be demonstrated using relaxed steady state analysis (*1, 5*).

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NOTATION

A_1, A_2 = adsorbed species
$C_R, C_{A1}, \ldots$ = concentrations
h = unit vector
$k_1, k_2, \ldots$ = rate constants
m = maximum reactant concentration
P = objective space
P_1, P_2 = products of catalytic reaction
R = reactant
s = defined in Equation (12a)
s_0 = s for limiting case $\gamma \rightarrow 0^+$
S = arbitrary set
S_p = periodic attainable set
S_s = steady state attainable set
t = dimensionless time
t_{real} = real time
u = dimensionless R concentration
U = admissible control set
$\overline{y}, \overline{y}_0, \overline{y}_i, \ldots$ = average objective vectors
$\overline{y}^1, \overline{y}^2$ = different components of an average objective vector

Greek Letters

α = nonnegative scalar
β = dimensionless kinetic parameter
γ = bang-bang control parameter
η, η^* = defined in Equation (12b) and (12c), respectively
ξ = dimensionless kinetic parameter
τ = period
χ = dimensionless kinetic parameter

MATHEMATICAL SYMBOLS

co = convex hull
$\subset$ = "is a proper subset of"
$\subseteq$ = "is a subset of"

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