

$\beta(X)$ = function defined in Equation (28)
 Δt = $t_w - t_s$
 θ = $(T - 1)/(T_w - 1)$
 ν = kinematic viscosity
 μ = viscosity
 π = $C_w^2 D_w B^* / N_{Sc}$
 η_1 = similarity variable, Equation (15)
 η_2 = similarity variable, Equation (39)
 φ_1 = stretched variable, Equation (19)
 φ_2 = stretched variable, Equation (40)
 ϵ = local angle between vertical and normal to body in free convection
 ψ = stream function, see Equations (14) and (39)

Subscripts and Superscripts

1 = forced convection
 2 = free convection
 ∞ = free stream value
 w = surface value
 \bar{j} = denotes dimensional value of the quantity j

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Manuscript received October 5, 1966; revision received April 27, 1968; paper accepted August 29, 1968.

Stability Analysis of a Continuous Flow Stirred Tank Reactor with Consecutive Reactions

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The stability of a continuous flow stirred tank reactor with consecutive chemical reactions $A \rightarrow B \rightarrow C$ was investigated here for the first time. The system equations were integrated numerically, and parts of the three-dimensional phase space were generated for systems having one and three steady states, respectively.

The surface separatrix was determined by trajectory plotting after extensive computation. The system was also analyzed by Liapunov's second method. Krasovskii's form was used to generate Liapunov functions which proved to be satisfactory but extremely conservative in the estimation of the largest possible regions of asymptotic stability.

The stability of a stirred tank reactor in which a single reaction $A \rightarrow B$ occurs has been the subject of much research in the last 15 yr. The first definitive analysis of this nonlinear problem was reported by Bilous and Amundson (5) who showed the conditions for the existence of multiple steady states and investigated the transient behavior of typical systems. They demonstrated the utility of phase plane representations of the reactor dynamics and showed how the stability analysis could be carried out by linearization of the system equations.

Since then the literature on reactor stability has grown considerably, due in large part to the contributions of Amundson and co-workers. An overall review of this literature is not appropriate here. However, it should be noted that studies of this subject have evolved three general

approaches to the theoretical analysis of stability. These are linearization analysis, direct computation of reactor performance, and application of Liapunov's second method.

The goal of such analysis is to determine whether the operation of a reactor will be stable or unstable as well as the bounds of regions of stable behavior. A region of asymptotic stability (RAS) about a potential steady state point in the phase plane is defined as the set of initial conditions from which the reactor system trajectories will asymptotically approach that steady state. When multiple steady states exist, there will usually be such a region around each stable steady state point, and it is common to define a separatrix as that imaginary line in the phase plane which separates the largest possible RAS's. In this context, much of stability analysis may be thought of as an effort to measure these largest regions of asymptotic stability and thereby to locate the separatrix.

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Of the aforementioned approaches, Liapunov's second (direct) method provides a potential means for obtaining more complete information than the linearized analysis with significantly less calculation than direct computer simulation. The first application of this approach to the reactor stability question was made by Berger and Perlmutter (3, 4). They analyzed the case of a single reaction, under conditions where only one stable steady state point existed, and demonstrated the use of Krasovskii's theorem in the selection of an appropriate Liapunov function.

The present paper reports a stability study of a continuous flow stirred reactor in which consecutive reactions $A \rightarrow B \rightarrow C$ are taking place. Such a case has not been studied in any detail previously, although the work of Liu and Amundson (8) concerning a polymerization reactor and that of Schmitz and Amundson (11) concerning reactions in two-phase systems with interphase mass transfer have been involved with problems similar to those encountered here. Some of the data representations used by these workers have also been applied in the present case.

It should be noted that the consecutive reaction case is a much more complex problem than that of one reaction. Whereas the stability of the latter may be easily investigated by the direct mapping of a few selected trajectories in the two-dimensional concentration-temperature phase plane, the consecutive reaction system is characterized by a three dimensional phase space which may require extensive computation before a suitable mapping is possible. As a result, the conservative Liapunov method has greater potential utility for the complex consecutive system.

The balance of this paper will demonstrate the utility of both of these approaches for a specific sample case.

THE MATHEMATICAL MODEL

Consider a perfectly mixed reactor with a cooling coil in which the two consecutive irreversible, exothermic, first-order reactions $A \rightarrow B \rightarrow C$ are occurring. By assuming constant physical and thermochemical properties, constant coolant temperature, and a feed stream containing only component A, the reactor performance may be described by the thermal energy balance and the material balances for species A and B which follow:

$$\rho V_R C_p \frac{dT}{d\theta} = (-\Delta H_1) V_R k_1 C_A \exp(-E_1/RT) + (-\Delta H_2) V_R k_2 C_B \exp(-E_2/RT) - \rho q C_p (T - T_i) - U A_R (T - T_c) \quad (1)$$

$$V_R \frac{dC_A}{d\theta} = -V_R k_1 C_A \exp(-E_1/RT) - q (C_A - C_{Ai}) \quad (2)$$

$$V_R \frac{dC_B}{d\theta} = V_R k_1 C_A \exp(-E_1/RT) - V_R k_2 C_B \exp(-E_2/RT) - q C_B \quad (3)$$

These balances may be rewritten in more compact dimensionless form as shown in Equations (4), (5), and (6):

$$\frac{d\bar{T}}{d\theta} = \alpha_1 \beta_1 \bar{C}_A \exp(-\delta_1/\bar{T}) + \alpha_2 \beta_2 \bar{C}_B \exp(-\delta_2/\bar{T}) + \gamma \bar{T} + 1 \quad (4)$$

$$\frac{d\bar{C}_A}{d\theta} = -\alpha_1 \bar{C}_A \exp(-\delta_1/\bar{T}) - \bar{C}_A + 1 \quad (5)$$

$$\frac{d\bar{C}_B}{d\theta} = \alpha_1 \bar{C}_A \exp(-\delta_1/\bar{T}) - \alpha_2 \bar{C}_B \exp(-\delta_2/\bar{T}) - \bar{C}_B \quad (6)$$

STEADY STATE ANALYSIS

Equations (4), (5), and (6) describe the steady state behavior of the reactor when the left sides are set equal to zero. Concentrations $\bar{C}_{A_{ss}}$ and $\bar{C}_{B_{ss}}$ may be eliminated to yield

$$0 = \frac{\alpha_1 \beta_1 \exp(-\delta_1/\bar{T}_{ss})}{[\alpha_1 \exp(-\delta_1/\bar{T}_{ss}) + 1]} + \frac{\alpha_2 \beta_2 \exp(-\delta_2/\bar{T}_{ss})}{[\alpha_2 \exp(-\delta_2/\bar{T}_{ss}) + 1]} - \frac{\alpha_1 \exp(-\delta_1/\bar{T}_{ss})}{[\alpha_1 \exp(-\delta_1/\bar{T}_{ss}) + 1]} - (\gamma \bar{T}_{ss} - 1) \quad (7)$$

Equation (7) will admit one to five possible solutions and corresponding steady states. The existence of one, three, or five steady states is demonstrated schematically by the intersections of typical heat generation and removal curves in Figure 1. The existence of two or four steady states requires that the removal and generation curves be tangent at one or two points, a physically unlikely situation.

It was decided in this work to investigate cases in which one and three steady states exist. The appropriate parameters and steady state points are listed in Table 1. A five steady state case was not studied, since it is physically less likely to occur and the other cases demonstrate the essential features of the problem.

TRANSIENT ANALYSIS

The transient behavior of the system may be examined by numerical integration of Equations (4), (5), and (6) for a particular set of parameters and initial conditions. This yields a trajectory in the three-dimensional phase space of \bar{C}_A , \bar{C}_B , \bar{T} , which may be displayed by its projections on the three-coordinate planes. Since two such plots are sufficient to portray the trajectories, only the projections in the $\bar{T} = 0$ plane and the $\bar{C}_B = 0$ plane will be shown below. The phase space is thus represented in Figures 2 and 3 for the one steady state case and in Figure 4 and 5 for the case of three steady states. The numbered curves on these plots represent various trajectories. Similar representations have been used by others previously for different systems (8, 11).

Figures 2 and 3 demonstrate that the single steady state is stable, as may also be shown by linearization analysis. The apparent crossing of curves in these figures is not real but a result of the projection in two dimensions of the three-dimensional curves. Real crossing is impossible, since the solution of the equations is unique.

The set of curves for the three steady state case demonstrates that one of the steady states is unstable while the other two are stable, which again may be shown by linearization analysis. The determination of the separatrix from such trajectories is clearly not a simple task. One possible technique is to integrate the system equations in the negative time direction starting at or near the unstable steady state point. Although this approach may be used quite successfully in single reaction systems (10) where a few trajectories that bracket the separatrix closely may be easily found, it was not satisfactory for the three-dimensional phase space. Proper selection of representative trajectories was found to be very difficult, as was also observed by Schmitz (11) in his work. As a result, it was decided to follow a different but more systematic approach suggested by the projections of the curves on the $\bar{C}_B = 0$ plane in Figure 5. Comparison of this figure with those for a single-reaction phase plane, such as shown by Bilous

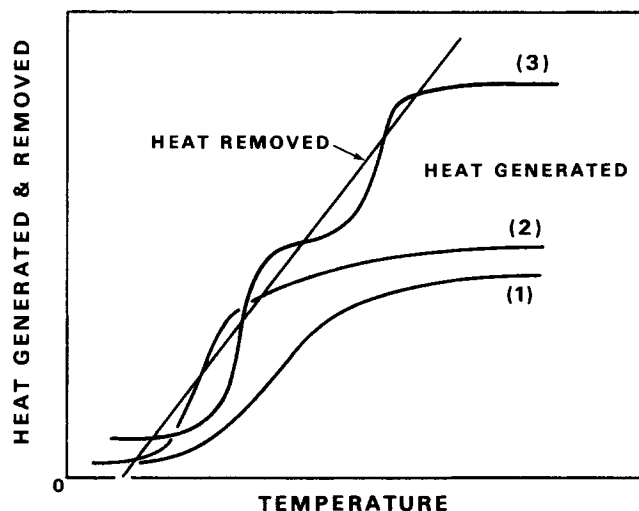


Fig. 1. Schematic heat generated and heat removed curves.

and Amundson (5), indicates a similarly shaped separatrix in that plane. In fact, the surface separatrix appeared essentially like a three-dimensional extension of the two-dimensional case.

In view of this, a series of trajectory calculations was started with initial conditions distributed appropriately in a plane of constant \bar{C}_B . The calculations from those points were then followed to determine where each starting point was situated relative to the separatrix surface. New initial conditions were then selected in the same plane and the procedure repeated until the intersection of the separatrix surface with that plane was sufficiently well determined. The entire operation was repeated with a new plane of constant \bar{C}_B , until the shape of the separatrix in three dimensions was well defined.

The results of these calculations are shown in Figure 6. The fact that the separatrix is in this case a surface similar in shape to the usual two-dimensional separatrix line is certainly a reflection of the set of parameters used in the present calculations. Totally different shapes can be expected for other values. However, the basic pattern searching demonstrated here should be useful for many situations.

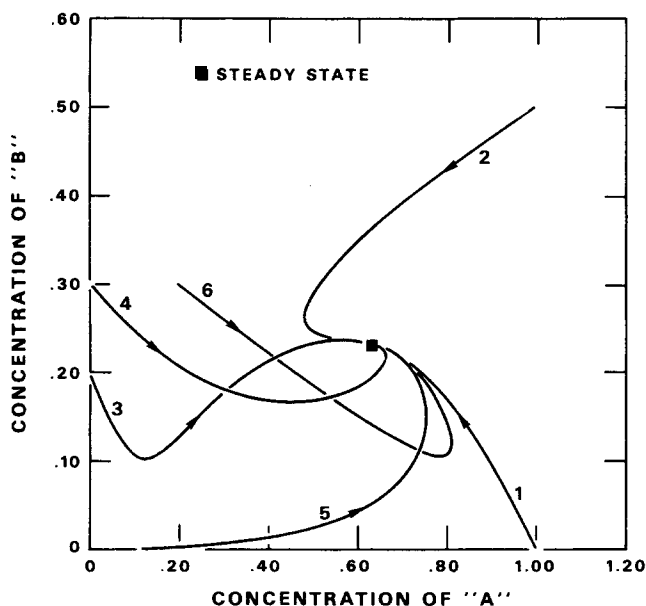


Fig. 2. Projections of system trajectories in $\bar{T} = 0$ plane, one steady state case.

A total of 1½ hr. of computer time (CDC 3400) was required to obtain the results of Figure 6. By contrast, only 10 min. of computation is required to determine the separatrix for a single-reaction system. This points up the potential value of an alternate method for determining the RAS's in the three-dimensional case.

DETERMINATION OF RAS BY LIAPUNOV'S SECOND METHOD

The well-known method of Liapunov has been presented (6) and summarized in the literature (3, 4) and will not be resummarized here. Suffice it to say that for a system described by the vector differential equation

$$\dot{\hat{x}} = \hat{f}(\hat{x}) \quad (8)$$

where $\hat{f}(0) = 0$, a region of asymptotic stability about $\hat{x} = 0$ will be defined by $V(\hat{x}) \leq K$, where V is scalar, positive definite function, with a negative derivative when $\hat{x} \neq 0$.

The use of this method which provides only a sufficient condition for stability becomes a search for a suitable Liapunov function. Although several possible functional forms were considered for this problem, it was finally decided to use Krasovskii's form (3). Thus

$$V(\hat{x}) = \hat{f}^T A \hat{f} \quad (9)$$

where A is positive definite matrix suitably chosen to provide a reasonable RAS.

Krasovskii's form was selected because of the success of Berger and Perlmutter (3, 4) with this approach in a two-dimensional case and the ease with which the contours of the Liapunov function may be oriented by manipulation of the matrix A . Berger and Perlmutter were able to achieve their best results in two dimensions when the Liapunov function contours were ellipselike curves straddling the steady state line (9) and more or less following the normal flow of the trajectories. Correspondingly, for a consecutive reaction system, the Krasovskii form may be chosen to yield contours essentially aligned with the steady state plane

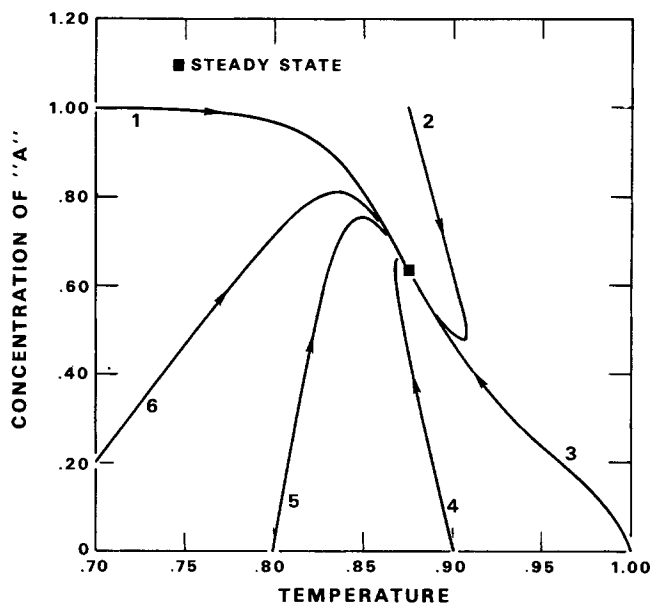


Fig. 3. Projections of system trajectories in $\bar{C}_B = 0$ plane, one steady state case.

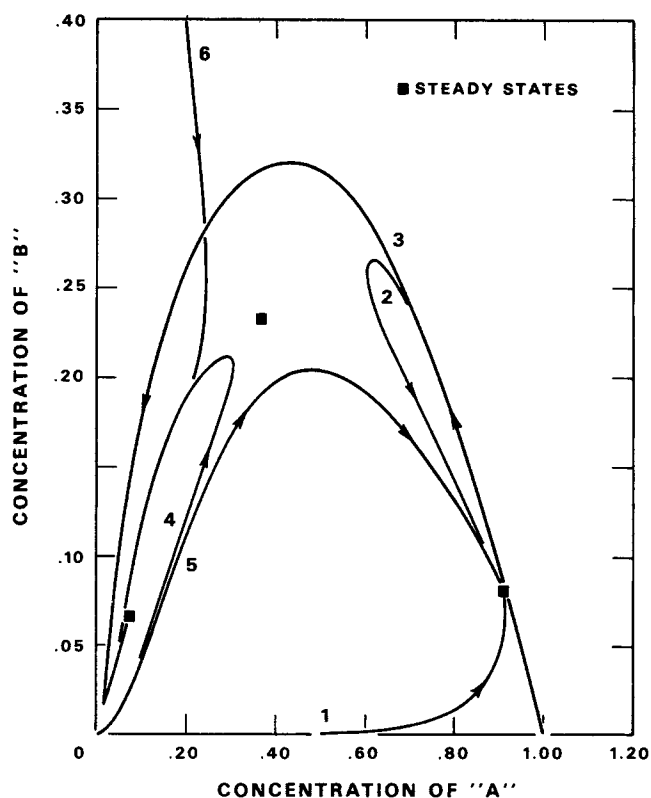


Fig. 4. Projections of system trajectories in $\bar{T} = 0$ plane, three steady state case.

$$\gamma\bar{T} + (\beta_1 + \beta_2)\bar{C}_A + \beta_2\bar{C}_B = 1 + \beta_1 + \beta_2 \quad (10)$$

FORMULATION OF THE LIAPUNOV FUNCTION

The symmetric, positive definite third-order matrix may be written in general as

$$A = \begin{bmatrix} 1 & s & p \\ s & t^2 & u \\ p & u & w^2 \end{bmatrix} \quad (11)$$

and must satisfy the following conditions:

$$\begin{vmatrix} 1 & s \\ s & t^2 \end{vmatrix} > 0 \quad (12)$$

$$|A| > 0 \quad (13)$$

When A is substituted into Equation (9), the latter may be expanded to yield

$$V = \hat{f}_1^2 + t^2 \hat{f}_2^2 + w^2 \hat{f}_3^2 + 2s \hat{f}_1 \hat{f}_2 + 2p \hat{f}_1 \hat{f}_3 + 2u \hat{f}_2 \hat{f}_3 \quad (14)$$

The question to be considered now is the choice of the constants p , s , t , u , and w to make V a Liapunov function and to orient it along the steady state plane. The second objective is accomplished by combining Equation (14) with the square of the plane equation, written in terms of the derivatives of the deviations from the steady state, and then by choosing constants p , s , and u so as to eliminate cross products of the derivative variables. The result is then (10)

$$V = [\gamma\hat{T} + (\beta_1 + \beta_2)\hat{C}_A + \hat{C}_B]^2 + [t^2 - (\beta_1 + \beta_2)^2]\hat{f}_2^2 + [w^2 - \beta_2^2]\hat{f}_3^2 \quad (15)$$

with A as follows:

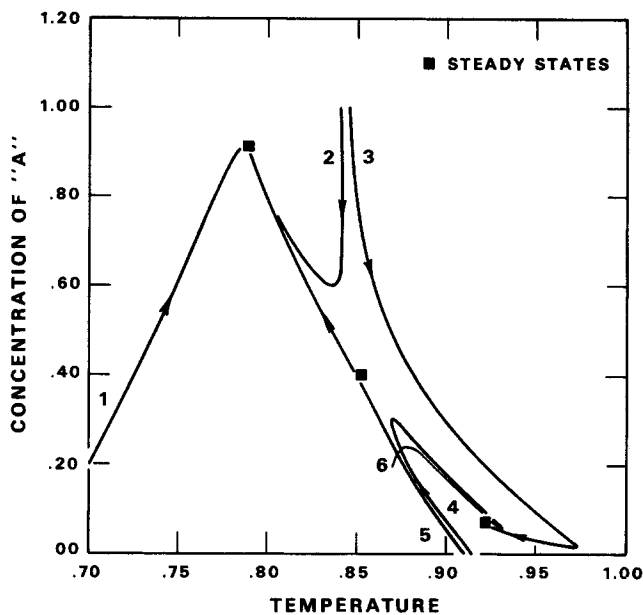


Fig. 5. Projections of system trajectories in $\bar{C}_B = 0$ plane, three steady state case.

$$A = \begin{bmatrix} 1 & (\beta_1 + \beta_2) & \beta_2 \\ (\beta_1 + \beta_2) & t^2 & \beta_2(\beta_1 + \beta_2) \\ \beta_2 & \beta_2(\beta_1 + \beta_2) & w^2 \end{bmatrix} \quad (16)$$

The application of the criteria for positive definiteness [Equations (12) and (13)] to this matrix yields

$$t^* = [t^2 - (\beta_1 + \beta_2)^2] > 0 \quad (17)$$

$$w^* = [w^2 - \beta_2^2] > 0 \quad (18)$$

Thus, as long as t and w are selected so as to satisfy Equa-

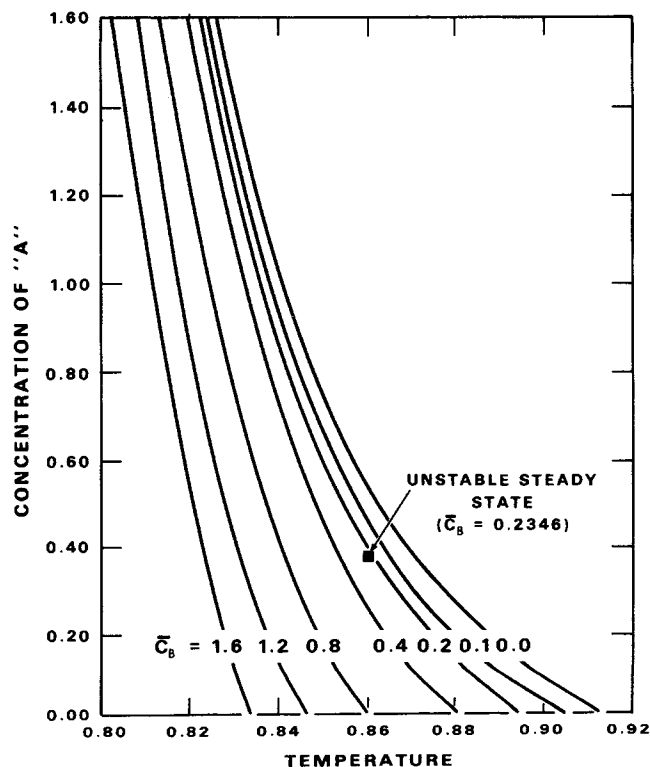


Fig. 6. Intersections of the separatrix and planes of constant \bar{C}_B .

TABLE 1. CONSECUTIVE REACTION SYSTEM PARAMETERS

	Physical parameters		Calculated parameters		
	(1)	(2)	(1)	(2)	
ρ	50	60	α_1	1.0×10^8	6.0×10^{13}
V_R	100	100	α_2	1.0×10^8	6.0×10^{13}
C_p	1.0	1.0	β_1	0.100	0.100
$-\Delta H_1$	5,000	9,885	β_2	0.100	0.100
$-\Delta H_2$	5,000	9,885	γ	1.20	1.28
k_1	1.0×10^8	1.08×10^{15}	δ_1	16.6	26.86
k_2	1.0×10^8	1.08×10^{15}	δ_2	16.6	26.86
E_1	21,240	45,000	\bar{T}_{ss1}	0.8748	0.7889
E_2	21,240	45,000	$\bar{C}_{A_{ss1}}$	0.6353	0.9108
U	5	60.57	$\bar{C}_{B_{ss1}}$	0.2318	0.08085
A_R	200	500	\bar{T}_{ss2}	—	0.8603
q	100	1,800	$\bar{C}_{A_{ss2}}$	—	0.3771
T_c	520	546	$\bar{C}_{B_{ss2}}$	—	0.2346
T_i	540	690	\bar{T}_{ss3}	—	0.9213
C_{Ai}	0.644	0.500	$\bar{C}_{A_{ss3}}$	—	0.07114
			$\bar{C}_{B_{ss3}}$	—	0.06572

(1) System with one steady state. (2) System with three steady states.

tions (17) and (18), V will be positive definite. To insure that V is also < 0 , it is necessary that the matrix \hat{F}^* be positive definite, where

$$\hat{F}^* = -[A\hat{F} + \hat{F}^T A] \quad (19)$$

and

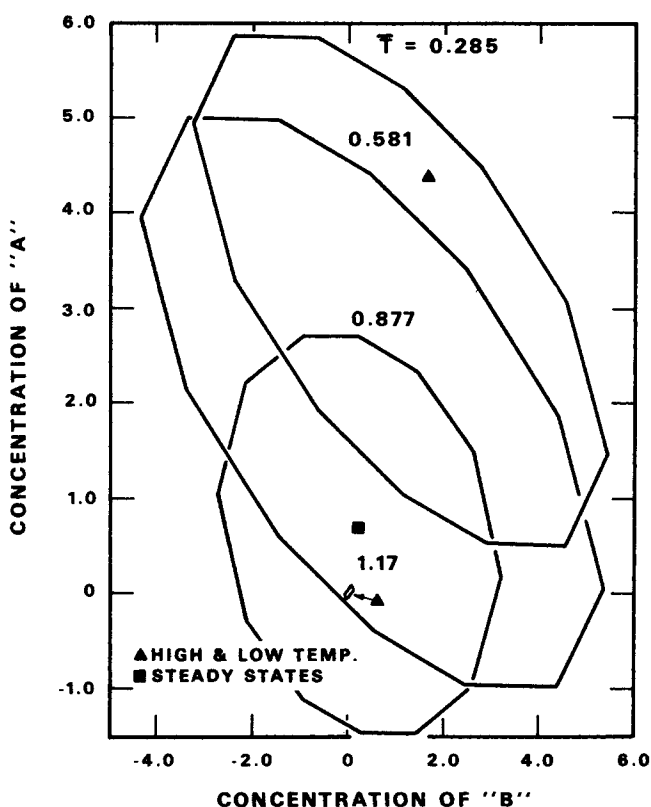


Fig. 7. Liapunov function, one steady state system.

$$\hat{F} = \left[\frac{\partial f_i}{\partial x_j} \right] \quad (20)$$

Now, suitable t^* and w^* may be found in principle by application of Sylvester's conditions, as done in the two-dimensional case by Berger and Perlmutter (3, 4). This requires the evaluation of $|\hat{F}^*|$, which in the present case involves an expression in terms of the variables \hat{T} , \hat{C}_A , and \hat{C}_B containing approximately 150 terms. Furthermore, the three-dimensional surface to which this determinant corresponds must be located on the proper side of the

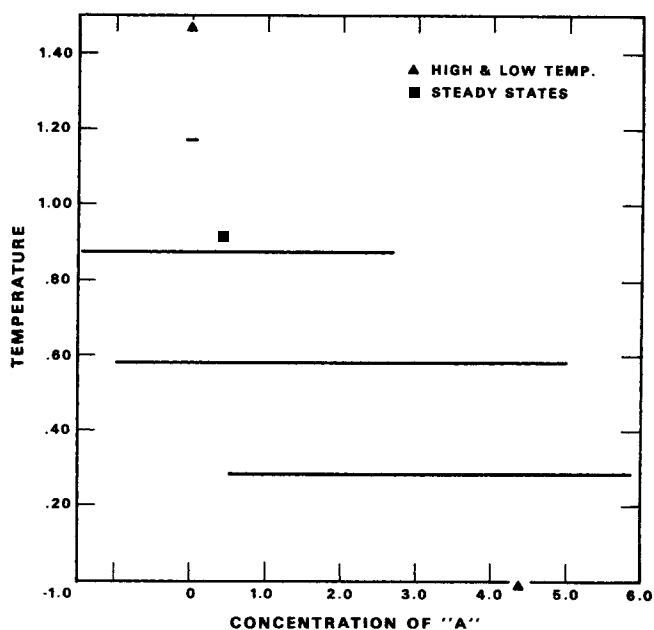


Fig. 8. Liapunov function, one steady state system.

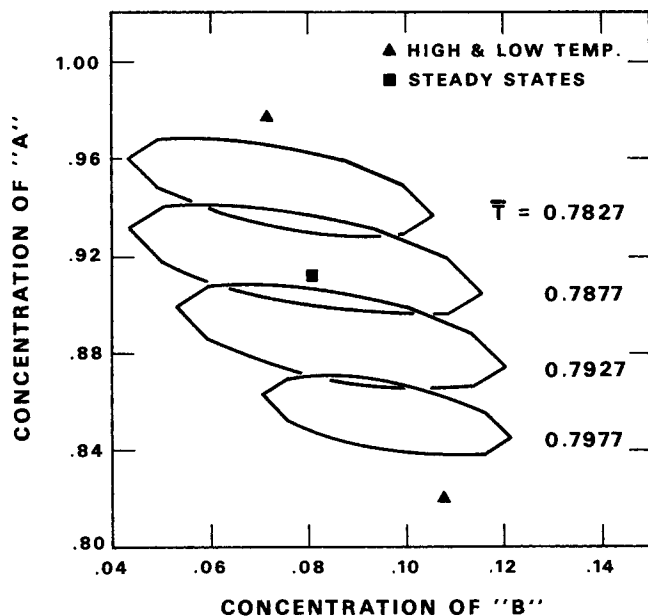


Fig. 9. Liapunov function, three steady states system. Steady state No. 1.

Liapunov function surfaces. Because of these complexities, this approach was discarded in favor of a numerical procedure for testing specific Liapunov functions. Values were selected for t^* and w^* , and the derivatives of the corresponding Liapunov function were calculated at a grid of discrete points on the function. If the derivatives were negative at all points of the grid, the function was considered a suitable candidate. Its value was then decreased and derivatives checked to verify that it applied very closely to the steady state point in question. The value was next increased to find the largest possible V value and the correspondingly largest possible RAS.

Although the above procedure is less elegant than that

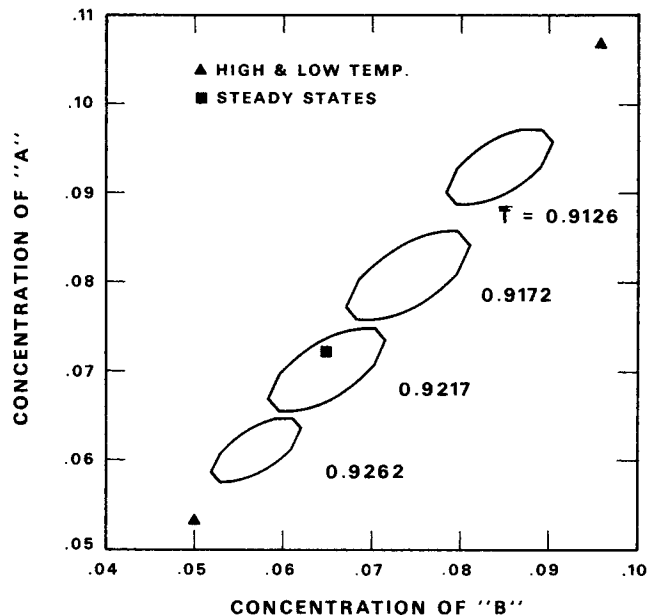


Fig. 11. Liapunov function, three steady states system. Steady state No. 3.

based on Sylvester's conditions, it permits manageable calculations. In addition, it may be superior, since as shown by Luecke and McGuire (7), Sylvester's method tends to produce a conservative estimate of the RAS. Recently, Berger and Lapidus (2) have described an alternate procedure for applying Krasovskii's theorem in multidimensional cases. Their computer algorithm is designed to minimize $-V$ at constant values of V in order to find the largest value which produces negative time derivatives. They report good results with this approach which uses a second-order minimization technique with penalty functions. Although this method may be superior to the present one for systems with many dimensional phase spaces, it is not clearly so for the system under consideration here.

From the search method described above, the largest regions of asymptotic stability were found for the stable steady states listed in Table 1 when

$$t^* = w^* = 0.01 \quad (21)$$

The resulting RAS for the case of one steady state is shown in Figures 7 and 8. These are projections on the $\bar{T} = 0$ and $\bar{C}_B = 0$ coordinate planes of the intersection curves of the Liapunov function and four different planes of constant \bar{T} . The actual functions represent sausage-shaped volumes extended along the steady state plane with the major axis leaning generally toward the \bar{T} axis. The cross sections in the $\bar{C}_A - \bar{C}_B$ plane form ellipses or circles but are shown as polygons in the figures which were computer drawn.

For this case, the RAS shows that the system is stable for deviations from steady state as large as 500°F. and 3.0 lb./cu.ft. Of course, the entire positive octant of the phase space is stable in this case, which, therefore, does not present a stringent test of the approach. It should be noted that the Liapunov function extends into negative portions of the phase space, indicating the trajectories starting at those points will pass into the positive octant and terminate at the steady state. Physically, such trajectories only become meaningful when they enter the positive octant.

RAS's found for the three steady state systems are shown in Figures 9 through 12. These regions are similar in gen-

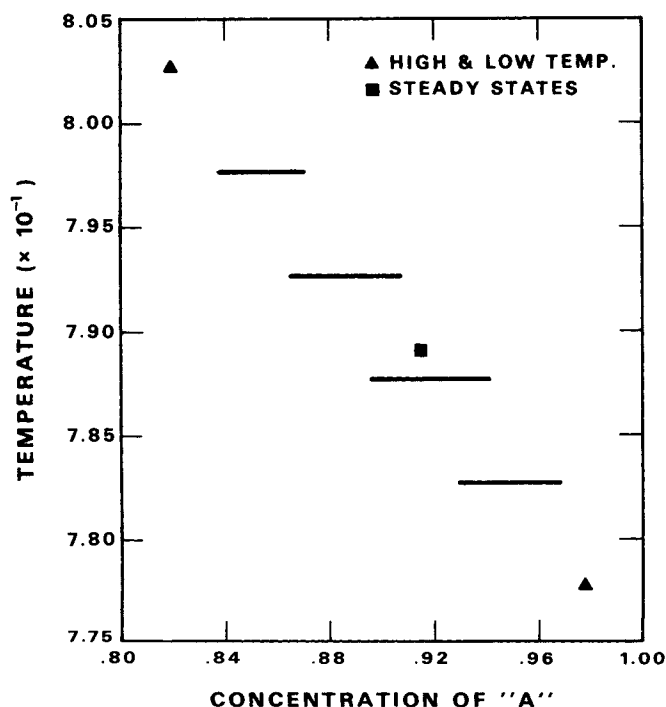


Fig. 10. Liapunov function, three steady states system. Steady state No. 1.

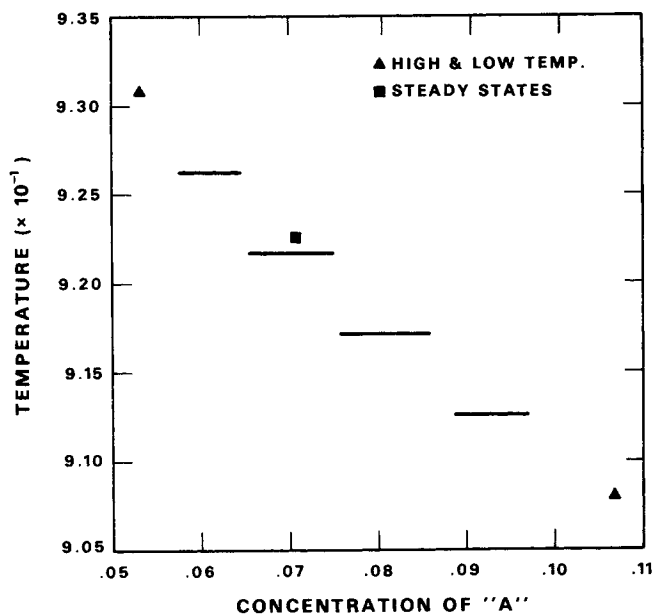


Fig. 12. Liapunov function, three steady states system. Steady state No. 3.

eral shape yet substantially smaller than that found for the one steady state case and are, at best, a conservative estimate of the RAS. One reason for this conservative behavior is the closeness of the steady states. This causes the system trajectories to change direction rapidly near the steady states, thus limiting the applicability of any Liapunov function. In addition, the type of function used in this work forces the shape to be more or less symmetric in the temperature direction. The RAS can, in general, be no longer than twice the temperature difference between the stable steady state under consideration and the unstable point. The best cases reported here were only 20% of this difference. What is clearly needed is a Liapunov function which can extend further on one side of the steady state than the other. However, no suitable form is apparent at present.

SUMMARY

This stability study has shown some of the characteristics of the three-dimensional phase space for a specific consecutive reaction system. Determination of the system separatrix by exhaustive calculation has been demonstrated, as well as analysis by Liapunov's second method. Although Krasovskii's form has been successfully used to find suitable Liapunov functions, the results are so conservative in multiple steady state cases that direct integration of system equations is recommended for analysis of such a problem at present.

NOTATION

A = positive definite matrix used in Liapunov functions
 A_R = heat transfer surface area, sq.ft.
 C = concentration, lb._m/cu.ft.
 C_i = inlet concentration, lb._m/cu.ft.
 \bar{C} = dimensionless concentration = C/C_{Ai}
 C_p = specific heat of reaction mixture, B.t.u./lb._m (°R.)
 E = activation energy of reaction, B.t.u./lb. mole
 \hat{f} = vector of dimensionless state derivative space, see Equation (8)

F = Jacobian matrix of \hat{f} , see Equation (20)
 F^* = positive definite matrix, see Equation (19)
 $-\Delta H$ = exothermic heat of reaction, B.t.u./lb._m
 k = frequency factor for reaction, sec.⁻¹
 K = Liapunov function value
 p = element of matrix A , see Equation (11)
 q = volumetric flow rate, cu.ft./sec.
 R = gas constant = 1.987 B.t.u./lb. mole (°R.)
 s = element of matrix A , see Equation (11)
 t = element of matrix A , see Equation (11)
 T = reactor temperature, °R.
 T_c = coolant temperature, °R.
 T_i = inlet temperature, °R.
 T^* = characteristic temperature = $\left(\frac{UA_R T_c}{\rho q C_p} \right) + T_i$ °R.
 \bar{T} = dimensionless temperature = T/T^*
 u = element of matrix A , see Equation (11)
 U = overall heat transfer coefficient, B.t.u./sq.ft. (sec.) (°R.)
 V_R = reactor volume, cu.ft.
 V = Liapunov function, dimensionless
 w = element of matrix A , see Equation (11)
 \hat{x} = dimensionless state space vector

Greek Letters

α = kV_R/q , dimensionless
 β = $\frac{(-\Delta H) C_{Ai}}{\rho C_p T^*}$, dimensionless
 γ = $UA_R/\rho q C_p + 1$, dimensionless
 δ = E/RT^*
 ρ = density of reaction mixture, lb._m/cu.ft.
 θ = time, sec.
 $\bar{\theta}$ = dimensionless time = $V_R \theta / q$

Subscripts

A, B = species A, B
 $1, 2$ = the first or second of the consecutive reactions
 ss = steady state condition

Superscripts

\wedge = deviation from steady state
 $—$ = dimensionless variable

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Manuscript received February 13, 1968; revision received August 9, 1968; paper accepted August 12, 1968.