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Dynamic Characteristics of a Periodically Forced Fluidized Bed Catalytic Reactor under Conventional PI Control

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Abstract. A two-phase model of a non-isothermal fluidized bed catalytic reactor with consecutive exothermic reactions $A \rightarrow B \rightarrow C$ is used to investigate the dynamic characteristics of this industrially important unit when it is periodically forced. The investigation concentrates on forcing a periodic regime of the autonomous system with a relatively high forcing frequency. It is shown that by periodic forcing of the feed temperature, chaotic regimes occur for small amplitudes of forcing. For constant frequency and with the change in the forcing amplitude the behavior of the unit alternates between periodic and chaotic regimes via period doubling and period adding mechanisms. When on the other hand the amplitude is fixed chaotic regimes also appear for small changes in the forcing frequency. The effects of these parametric perturbations on the yield of the desired intermediate product B is also investigated. It is shown that the yield can be improved by an appropriate selection of the forcing frequency.

Nomenclature

a	dimensionless two-phase parameter
A_C	cross-sectional area of the bed occupied by bubble phase (cm^2)
A_m	dimensionless forcing amplitude
G_c	volumetric gas flow rate in bubble phase (m^3s^{-1})
G_I	volumetric gas flow rate in dense phase (m^3s^{-1})
H	expanded bed height for the fluidized bed (cm)

K_c dimensionless proportional gain

K_i dimensionless integral mode gain

Le_i Lewis number of component i (heat capacity/mass capacity of component i)

Q_E mass exchange coefficient (s^{-1})

t normalized time

X_A dimensionless dense phase concentration of the reactant A

X_B dimensionless dense phase concentration of the product B

X_{if} dimensionless feed concentration of component i

X_{BH} dimensionless bubble phase concentration of B at the exit of the reactor

y yield of the intermediate product

Y dimensionless dense phase temperature

Y_f dimensionless feed temperature to the reactor (base value)

Y_m dimensionless set point of the controller

y_f dimensionless feed temperature

w_f dimensionless forcing frequency

w_o dimensionless natural frequency of the center of forcing chosen for the amplitude forcing.

w_p dimensionless natural frequency of the center of forcing chosen for the frequency forcing

Greek symbols

α_1 dimensionless pre-exponential factor for $A \rightarrow B$

α_2 dimensionless pre-exponential factor for $B \rightarrow C$

β_1 dimensionless overall exothermicity factor for $A \rightarrow B$

β_2 dimensionless overall exothermicity factor for $B \rightarrow C$

β reciprocal of the residence time of the bed

γ_1 dimensionless activation energy for $A \rightarrow B$

γ_2 dimensionless activation energy for $B \rightarrow C$

ϕ_i dimensionless effective mass capacity of the component i

Abbreviations

HB Hopf bifurcation point

PD period doubling point

Introduction

Chemically reactive systems are known to exhibit a variety of complex static and dynamic behavior depending on the values of system operating parameters. An autonomous model of a chemical reactor can exhibit simple or complex oscillations as well as chaotic behavior. The effect of feed changes on the dynamics of chemical reactors on the other hand can be of significant importance. In fact the interactions between the feed inputs and the nonlinearities of the reactor are quite relevant, since in real processes, feed conditions are generally time varying. The study of the dynamics of the non-autonomous system can, therefore yield significant results on the performances of the real physical process.

The periodically forced lumped parameter reactors have been extensively investigated in the last few years [1-7]. Mankin and Husdon [8], for instance, studied the case of the CSTR with a single irreversible reaction using the coolant temperature as the forcing variable. The authors showed the complex transition from quasi-periodic to chaotic behavior in the periodically forced reactor. The same system was studied in a more elaborate way by Kevrekidis *et al.* [1] using the two parameter (amplitude and frequency) excitations diagrams. Tambe and Kulkarni [9] studied a low frequency forcing of a chemical reactor and examined the resulting complex transition from periodicity to chaos through intermittency. Elnashaie and Abashar [7] on the other hand investigated the chaotic behavior of the forced fluidized bed reactor using a relatively simple two-phase model.

The results of these studies point out clearly to the complex dynamic behavior of the periodically forced chemical reactors and also to the unpredictability of the results associated with these parametric perturbations. Dramatic changes can be expected in the nature of the emerging dynamic attractors.

In this paper we investigate the periodic forcing of a three dimensional model of a non-isothermal fluidized bed reactor with consecutive catalytic reaction network $A \rightarrow B \rightarrow C$. The desired product is the intermediate component B. A schematic diagram of the reactor is shown in Fig.1. The autonomous system is known to exhibit steady state multiplicity as well as simple and complex oscillations [7]. By forcing a periodic regime of the autonomous system we will examine simultaneously the mechanisms of transition between periodic and chaotic regimes in the forced system as well as the effects of forcing on the yield of the intermediate desired product.

Previous work

The dynamic behavior of the autonomous model of a non-isothermal fluidized-bed reactor with the catalytic exothermic reaction $A \rightarrow B \rightarrow C$ was investigated by Elnashaie and co-workers [10,11]. The states used to describe the reactor model are the concentrations of the reactant A and product B, and the temperature of the reactor dense phase.

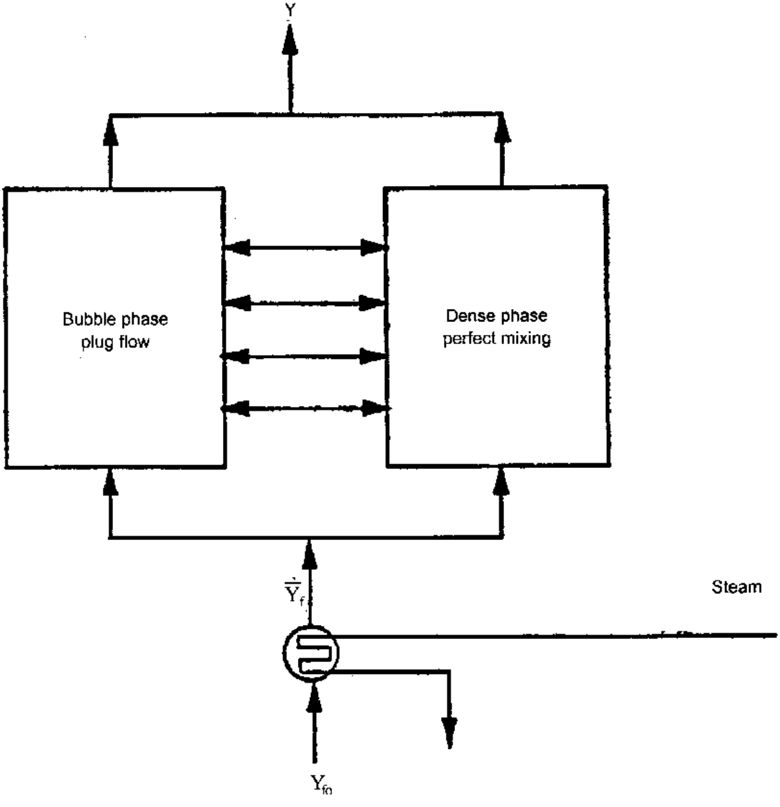

Fig. 1. Schematic diagram of the two-phase model of the fluidized bed reactor.

The autonomous i.e. unforced system is known to exhibit steady state multiplicity. The model investigated in this paper has three steady states, a low temperature (quenched) steady state, a high temperature (burn-out) steady state and a third steady state which is the desired operating point as it occurs at a physically realizable temperature and corresponds to the maximum yield. Since this desired steady state is unstable (saddle type) a feedback control system is needed for the operation of the reactor.

It is worth mentioning that this situation has practical industrial applications. Industrial fluid catalytic cracking (FCC) units are generally operated at the unstable steady state that gives maximum gasoline yield [12]. Other examples of the occurrence of this situation in petrochemical reactions are the partial oxidation of o-xylene to phthalic anhydride [13] and the oxidative dehydrogenation of ethylbenzene to styrene [14].

Elnashaie and Abashar [7] investigated the chaotic behavior of the periodically forced reactor with a simple proportional controller using a two dimensional model which neglects the fast dynamics of the intermediate product. The authors showed that period doubling and chaos occur for very small amplitudes of external forcing. Various mechanisms of transition between periodic windows and chaotic regimes were observed and analyzed.

In the present paper the original three dimensional system is investigated when it is under a conventional proportional-integral (PI) controller and when the feed temperature is periodically forced. The continuity diagram of the PI controlled reactor is shown in Fig. 2. It can be seen that the integral mode of the controller destroys the steady state multiplicity. The continuity diagram of the system shows one static branch that corresponds to the desired steady state. The integral action on the other hand induces interesting dynamic characteristics in the system including transition form simple to complex oscillatory behavior via period doubling bifurcation, intermittent chaos and period adding bifurcation [15].

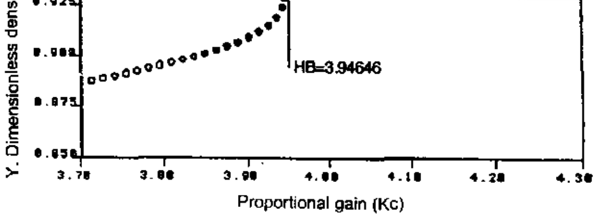

Fig. 2. Bifurcation diagram Y vs Kc when the reactor is under proportional-integral control. The integral mode gain is Ki = 0.05. Stable periodic attractors emanating form the Hopf point (HB) lose their stability through period doubling (PD) bifurcation.

A portion of the Poincare bifurcation diagram (intersection of the trajectories with a fixed hyperplane) is shown in Fig.3(a). The system alternates between chaotic regimes through period adding bifurcation i.e. the period of the system increases with the controller proportional gain.

The effects of forcing a periodic attractor of the autonomous system is investigated in this paper. The center of forcing is a period-8 attractor corresponding to Kc = 3.810 as shown in Fig. 3(b). The natural frequency of the system at the center of forcing is wo = 0.0210832235. High accuracy is required for the determination of the natural frequency because its value affects the location of forced subharmonics. A shooting method [16] was used for this purpose.

Before the results of the investigation are discussed we present briefly the model used for the reactor. The detailed assumptions and the mathematical derivation of the model were presented by Elnashaie *et al.* [10,11].

Mathematical Model of the Reactor

The bubble-phase mass and heat balances equations are assumed at pseudo steady state because of negligible mass and heat capacities. The equations for the dense phase materials and energy balances in dimensionless form are given by the following three nonlinear differential equations

$$\frac{1}{Le_A}\frac{dX_A}{dt} = \bar{\beta}(X_{Af} - X_A) - \alpha_1 \exp\left(-\frac{\gamma_1}{Y}\right)X_A, \tag{1}$$

$$\frac{1}{Le_B}\frac{dX_B}{dt} = \bar{\beta}(X_{Bf} - X_B) + \alpha_1 \exp\left(-\frac{\gamma_1}{Y}\right)X_A - \alpha_2 \exp\left(-\frac{\gamma_2}{Y}\right)X_B, \tag{2}$$

$$\frac{dY}{dt} = \bar{\beta}(\bar{Y}_f - Y) + \alpha_1\beta_1 \exp\left(-\frac{\gamma_1}{Y}\right)X_A + \alpha_2\beta_2 \exp\left(-\frac{\gamma_2}{Y}\right)X_B \tag{3}$$

X_A, X_B represent the dimensionless concentrations of components A and B, and Y is the dimensionless dense phase temperature. Y_m is the set point corresponding to the dimensionless dense phase temperature giving the maximum yield and Y_{fo} is the base value dimensionless feed temperature (when $Y = Y_m$). The PI controlled reactor is described by the equations above and the following control law where the feed temperature y_f, chosen as the manipulated variable, is related to the error (Y_m - Y) through a PI controller with proportional gain K_c and integral mode gain K_i

$$\bar{Y}_f = Y_f + K_c(Y_m - Y) + K_I\int_0^t (Y_m - Y)dt \tag{4}$$

When the reactor is periodically forced the control law (Eq. 4) takes the following form

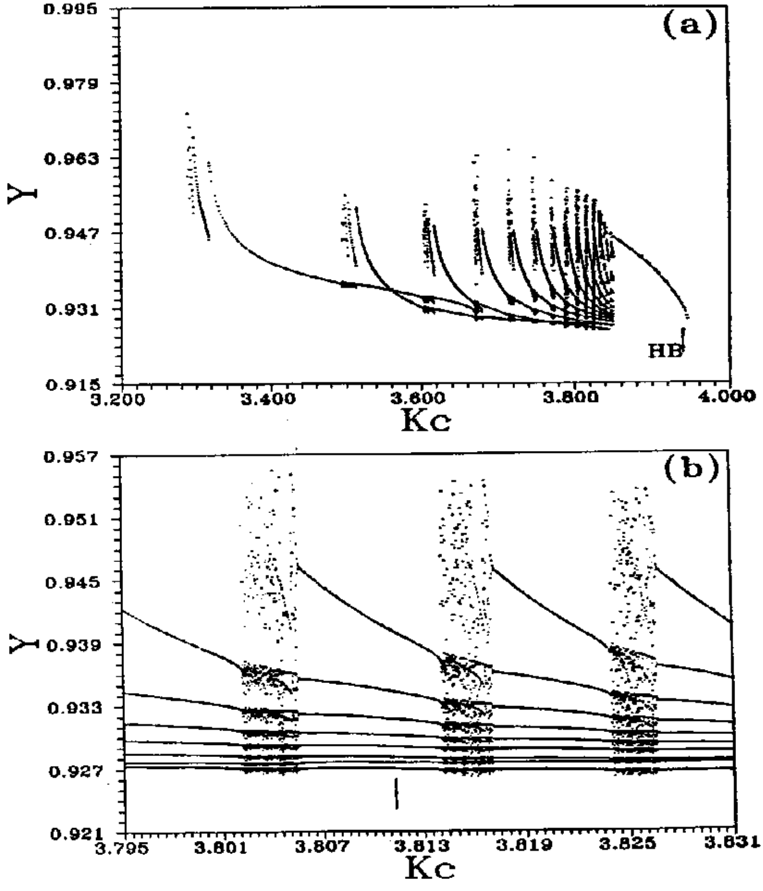

Fig. 3. (a) Poincare map (intersection of trajectories with the hyperplane XB = 0.6313) of the PI controlled reactor. The integram mode gain is Ki = 0.05. (b) Enlargement of Figure 3(a) showing the period-8 attractor (Kc = 3.810) chosen as the center of amplitude forcing.

$$\overline{Y}_f = (Y_f + A_m \sin wt) + K_c (Y_m - Y) + K_i \int_0^t (Y_m - Y)\,dt \qquad (5)$$

where A_m and w are respectively the forcing amplitude and frequency. The unforced system is obviously the limiting case of the forced system as the forcing amplitude (or frequency) goes to 0. The rest of the model parameters and the data used in the investigation are given in Table 1.

Table 1. Data used in the model

Normalized pre-exponent factor for the reaction $A \rightarrow B$, α_1	10^8
Normalized pre-exponent factor for the reaction $B \rightarrow C$, α_2	10^{11}
Dimensionless overall exothermicity factor for the reaction $A \rightarrow B$, β_1	0.4
Dimensionless overall exothermicity factor for the reaction $B \rightarrow C$, β_2	0.6
Dimensionless activation energy for the reaction $A \rightarrow B$, γ_1	18.0
Dimensionless activation energy for the reaction $A \rightarrow C$, γ_2	27.0
Lewis number of component A, Le_A	1.0
Lewis number of component B, Le_B	0.454545
Feed concentration of component A, X_{Af}	1.0
Feed concentration of component B, X_{Bf}	0.0
Dimensionless feed temperature to the reactor (basevalue), Y_f	0.55342072
Set point for the controller, Y_m	0.92955130
Reciprocal of the effective residence time of the bed, β	0.12543050
Dimensionless proportional gain, K_c	3.810
Dimensionless integral mode gain, K_i	0.05

The yield (y) of intermediate product in the reaction network $A \rightarrow B \rightarrow C$ is given by [10]

$$y = \frac{G_I X_B + G_C \overline{X}_{BH}}{(G_I + G_C) X_{Af}} \qquad (6)$$

where

$$\overline{X}_{BH} = X_B + (X_{Bf} - X_B) e^{-a} \qquad (7)$$

and

$$a = \frac{Q_E H A_C}{G_C} \qquad (8)$$

The two phase model used in this investigation has been used successfully to simulate type IV industrial fluid catalytic cracking units [17-19].

Results and Discussion

The forcing frequency used in this investigation is relatively high. The ratio w_f/w_o of the forcing frequency to the system natural frequency is taken to be a rational value equal to 5. Therefore at low amplitude of forcing we expect the phase trajectories of the system to lie on a three dimensional torus and an entrainement region to prevail. It is known that the possible attractors for periodically forced systems are periodic, quasi-periodic and chaotic attractors. When the forced system is periodic its period is an integer multiple of the forcing period. The investigation of the periodically forced system is suitably carried out using a Poincare map. The phase projection of the trajectories are inspected at specific times t_s which are multiples of the forcing period i.e. $t_s = \dot{m}(2\pi/w_f)$. This is also called stroboscopic technique for obvious reasons. Transient motions appear as scattered dots on the map while the emergence of a periodic attractor of order n (subharmonic) would be seen as jumps between n fixed points.

A complete one-parameter stroboscopic bifurcation diagram for the reactor model is shown in Fig. 4(a). The Y-axis represents the dimensionless dense phase temperature. On the scale of this figure the diagram looks like an alternation of periodic regimes interrupted by chaotic-like strips via period adding bifurcation. At large forcing amplitude i.e. beyond the value $A_m = 0.0420$ the system is fully entrained and emerges as a period-one attractor.

The effects of the amplitude forcing on the yield of the intermediate product in the reaction network $A \rightarrow B \rightarrow C$ can be seen in Fig. 4(b) showing the variations of the average yield with the forcing amplitude. Starting with a yield 0.5644 corresponding to the autonomous system ($A_m = 0.0$) it can be seen that the yield trend is generally a decreasing one. Occasional bursts can be observed but in no instance is the yield greater than the value corresponding to the autonomous system. It is therefore not possible to improve the yield by increasing the forcing amplitude. This however is not the case when the frequency is chosen as the bifurcation parameter as it will be shown in later sections.

The behavior of the system as shown in Fig. 4(a) is in fact more complicated and an enlargement of some regions of the diagram is needed in order to analyze the finer structure of the system behavior.

Region R_1, $A_m = 0.00$ to 0.0270

The limiting case of this region is the autonomous oscillatory system ($A_m = 0.00$). Five branches emerge on the stroboscopic diagram of Fig. 5(a). Frequency locking occurs therefore at relatively small amplitudes, as five sub-harmonic saddles and five nodes are born on the surface of a three dimensional torus and the system is entrained by the forcing frequency. The resonant (entrained) trajectory has obviously periodicity five times the forcing period (frequency locking). It can be seen that through out region R_1 the system

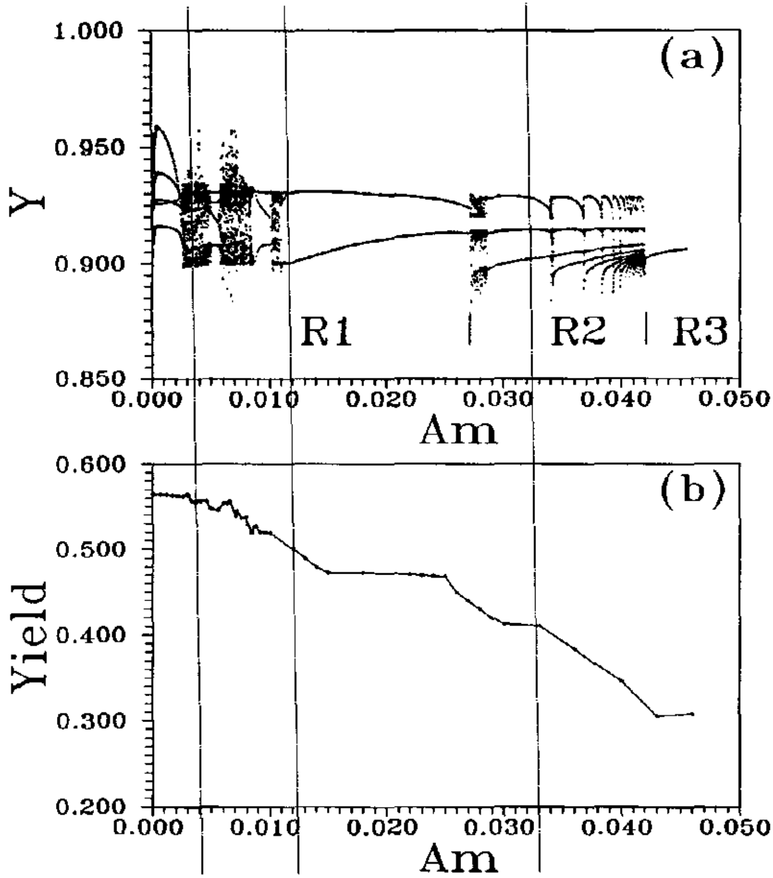

Fig. 4. (a) One-parameter stroboscopic Poincare bifurcation diagram for the amplitude forcing of the reactor. (b) The variations of the yield with the forcing amplitude.

alternates between periodic and chaotic-like regimes by reverse period adding i.e. the period of the windows decrease as the forcing amplitude increases. At the end of region R_1 the system emerges as a period-2 attractor.

Periodic windows and intermittency

To analyze the mechanism of transition from chaos to periodic regime in region R_1 of Fig. 5(a) we consider the region R_4 enlarged in Fig. 5(b) covering the parameter range $A_m \in [0.0015, 0.0055]$. The system starts its transformation from periodic to a chaotic-like behavior with a region of Period-5 attractor starting at $A_m = 0.0020$. As the forcing amplitude increases the periodic atttractor starts to undergo period doubling sequence at $A_m = 0.0026$ giving a region of Period-10 attractor which bifurcates again to chaos through period doubling and so on. Through out this region the system goes through an alternation of periodic windows and chaos and emerges as a Period-4 attractor starting at A_m = 0.0049.

The characteristics of the chaotic attractor are displayed by choosing the point A_1 ($A_m = 0.0040$) of Fig. 5(b). Dynamic simulations are shown in Fig. 6. Time trace and stroboscopic histogram are presented. The points on this histogram are clearly mixed indicating at least an ergodic behavior. The chaotic nature of the attractor is confirmed by computing Lyapunov exponents. A chaotic attractor has at least one positive Lyapunov exponent. These exponents can be computed efficiently using the technique and algorithm of Wolf *et al.* [20] and are found to be $\lambda_1 = 0.00245$, $\lambda_2 = -0.01269$, $\lambda_3 = -0.554951$ and $\lambda_4 = -0.403501$. The largest component is positive giving proof of the chaotic nature of the attractor.

The mechanism of bifurcation from the chaotic attractor to a Period-4 attractor is investigated by considering the point A_2 of Fig. 5(b) corresponding to a forcing amplitude $A_m = 0.0048$. Dynamic simulations at this point are shown in Fig. 7. The forth iterate of the dimensionless dense phase temperature $Y(n+4)$ is plotted against $Y(n)$. Figure 7 shows that the curve approaches the diagonal and almost becomes tangent at four points. The chaotic attractor is thus destroyed by the mechanism of intermittency as shown by Pomeau and Manneville [21]. The term intermittency refers to oscillations that are periodic for certain intervals (laminar phase) interrupted by intermittent erratic bursts of periodic oscillations of finite duration.

Region R_2: Period adding of the second kind

Beyond the forcing period corresponding to $A_m = 0.0270$ (end of region R_1) the system alternates between chaotic behavior and periodic windows until the system is fully entrained at high amplitudes (region R_3). The dominant mechanism here is period adding with strips of chaos. This behavior is termed period adding of the second kind and was first studied by Holden and Fan [22] in their analysis of the behavior of the autonomous three-dimensionless Rose-Hindnarch model for neural activity.

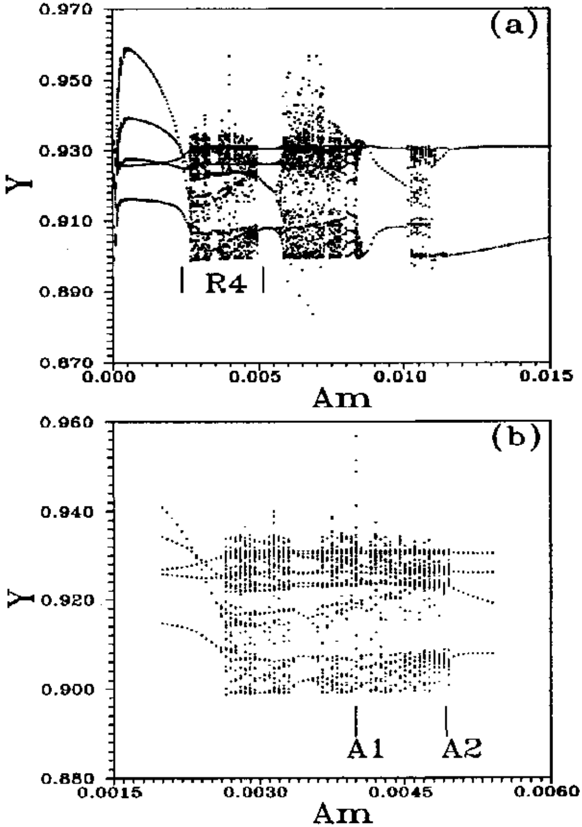

Fig. 5. (a) Enlargement of region R1 of Fig. 4(a) for the range Am ∈ [0.0,0.015]; (b) Enlargement of region R4 for the range Am ∈ [0.0015, 0.0055]. Period-5 attractor goes through an alternation of period doubling and chaos and emerges as a period-4 attractor.

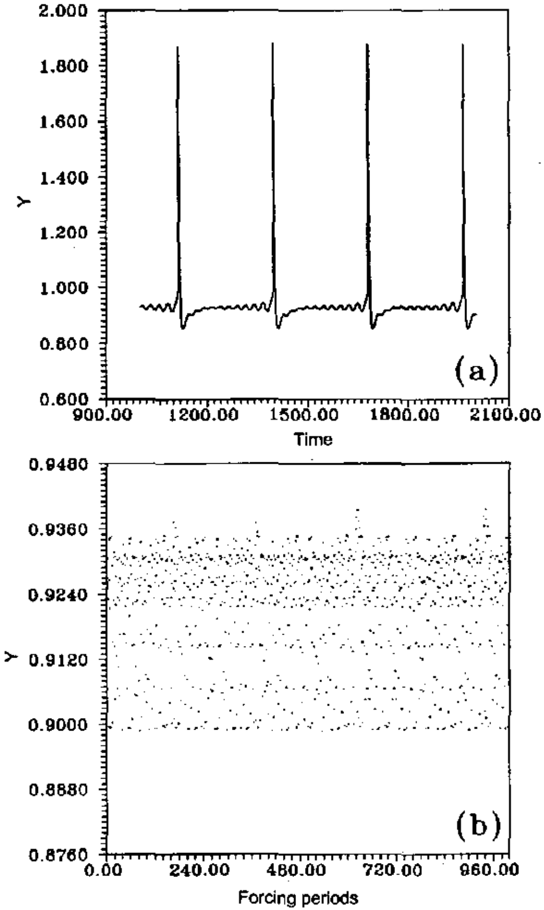

Fig. 6. Dynamics of the chaotic attractor at the point A_1 ($A_m = 0.040$) of Fig. 5(b). (a) Time trace; (b) Stroboscopic points histogram.

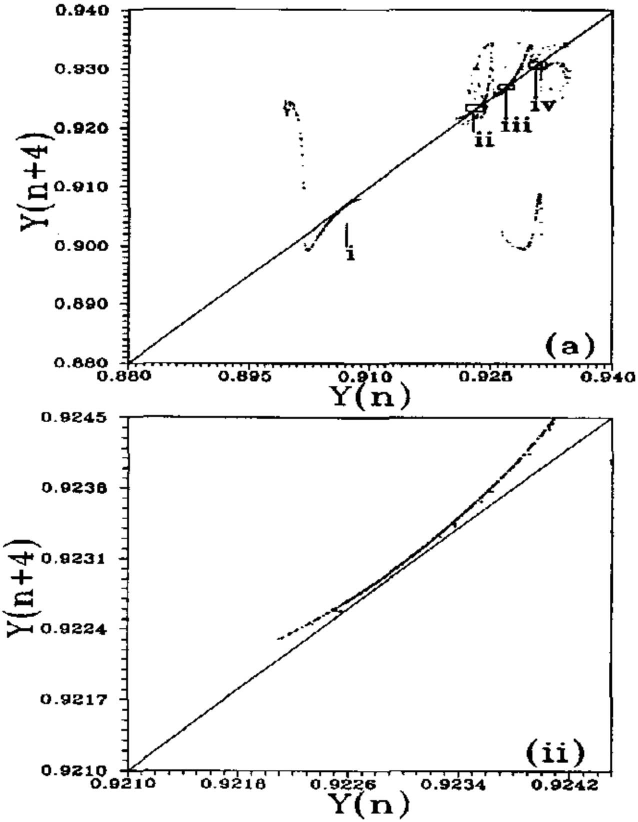

Fig. 7. Characteristics of the intermittent chaos at point A2 (Am = 0.0048) of Fig. 5(b). (a) Forth-iterate map. The trajectory is tangent at the diagonal in four points. (ii), (iii) and (iv) are enlargements of the corresponding boxes in (a).

The final bifurcation to the harmonic trajectory (fully entrained) is investigated by considering the point A_3 in region R_3 enlarged in Fig. 8(a) and corresponding to A_m = 0.0419. Dynamic simulations for this point are shown in Fig. 8(b-c). The maximum Lyapunov exponent is zero confirming further the quasi-periodic nature of the attractor.

Frequency Forcing of a Periodic Attractor

The second part of this investigation focuses on studying the effects of varying the forcing frequency while the amplitude is fixed. The center of forcing corresponds to a period-2 attractor at the constant amplitude $A_m = 0.02$ (Fig. 4(a)). The natural frequency w_p of the attractor is therefore half the forcing frequency w_f.

A one-parameter stroboscopic bifurcation diagram is shown in fig. 9. Figure 9(a) covers the parameter range $w / w_p \geq 1$ while the complementary range $w / w_p \leq 1$ is shown in Fig. 9(b).

On the scale of Fig. 9(a) it can be shown that starting form the periodic attractor ($w/w_p = 1$) the system starts its bifurcation with a periodic regime that persist through out region S_1. At the end of this region at $w / w_p = 1.401$ the periodic-like oscillations terminate and a chaotic-like regime appears. The chaotic-like regime persists through out region S_2. At the end of region S_2 at $w / w_p = 1.905$ a periodic attractor emerges and so on. The system goes then through an alternation of periodic and chaotic-like regimes. Similar behavior can be observed for the region $w / w_p < 1$ in Fig. 9(b). The system starts with a periodic regime and as the forcing frequency decreases the system goes through an alternation of chaotic-like and periodic regimes.

In order to analyze the finer structure of this mechanism the Lyapunov exponents spectrum is computed for a region covering the frequency range $w / w_p \in [3.50, 6.00]$ as shown in Fig. 10(a). The region corresponds to the end of region S_3 and the whole region S_4 of Fig. 9(a). By analyzing the Lyapunov spectrum shown in Fig. 10(b) it can be seen that its maximum starts with negative values, indicating a periodic regime then it becomes positive for some values of the forcing frequency, indicating a chaotic behavior and then alternates between positive and negative values.

Effects on the yield

The effects of frequency forcing on the yield of the desired product in the reaction network can be seen in Fig. 11 showing the variations of the average yield with the forcing frequency. The investigation covers the parameter range $w / w_p \in [0.0, 2.0]$. Starting from the value of the yield (0.5644) corresponding to the autonomous case ($w = 0$) the yield jumps to higher values for some range of the forcing frequency and then decreases. It can be seen that for a large range of forcing frequency ($w / w_p \in [0.0, 1.05]$) the yield is higher than the value 0.474830 corresponding to the center of forcing ($A_m = 0.020$).

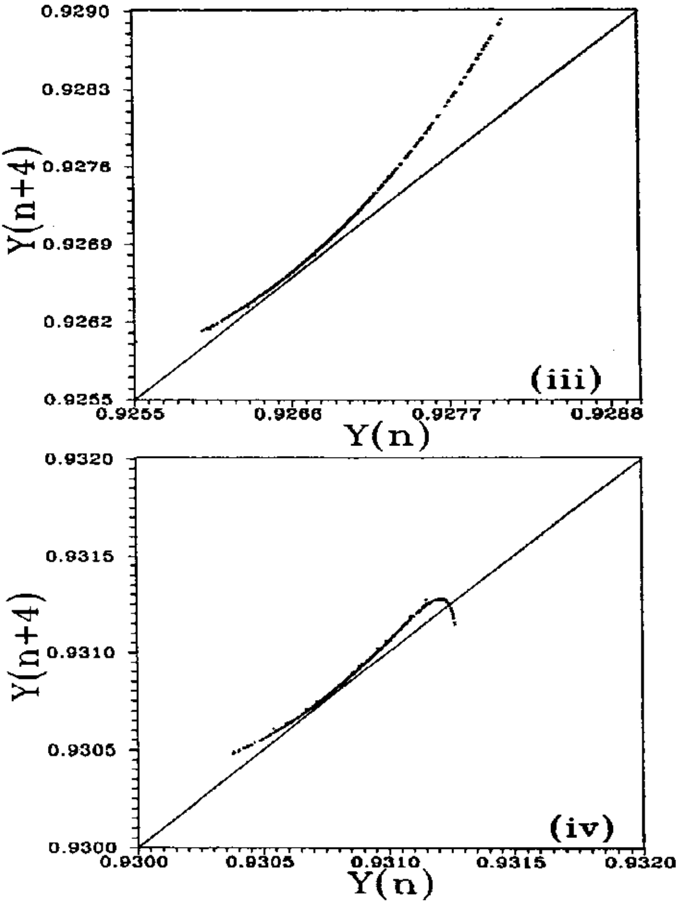

Fig. 8. (a) Enlargement of region R3 of Fig. 4(a) for the range Am $\in$ [0.033, 0.044]. The system alternates between periodic regimes with strips of chaos. (b) Stroboscopic points histogram.

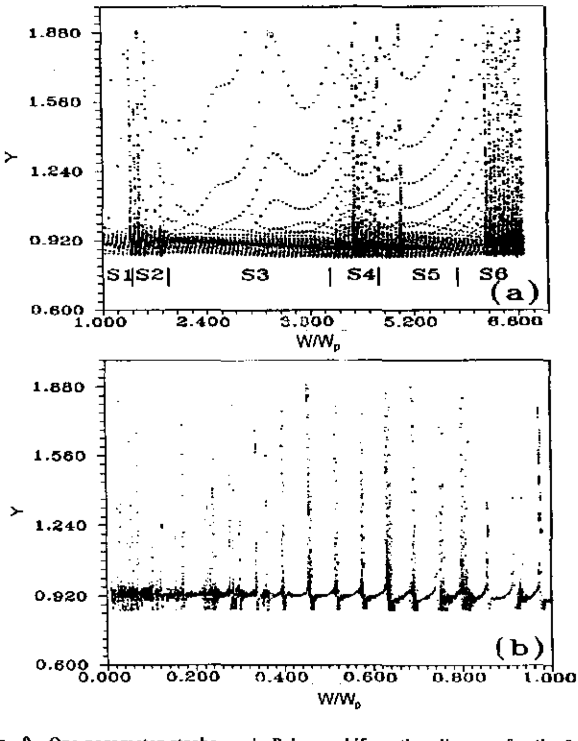

Fig. 9. One-parameter stroboscopic Poincare bifurcation diagram for the frequency forcing. The center of forcing corresponds to period-2 attractor at Am = 0.02 in Fig. 4(a). (a) Diagram for the range $w/w_p > 1$. (b) Diagram for the range $w/w_p < 1$..

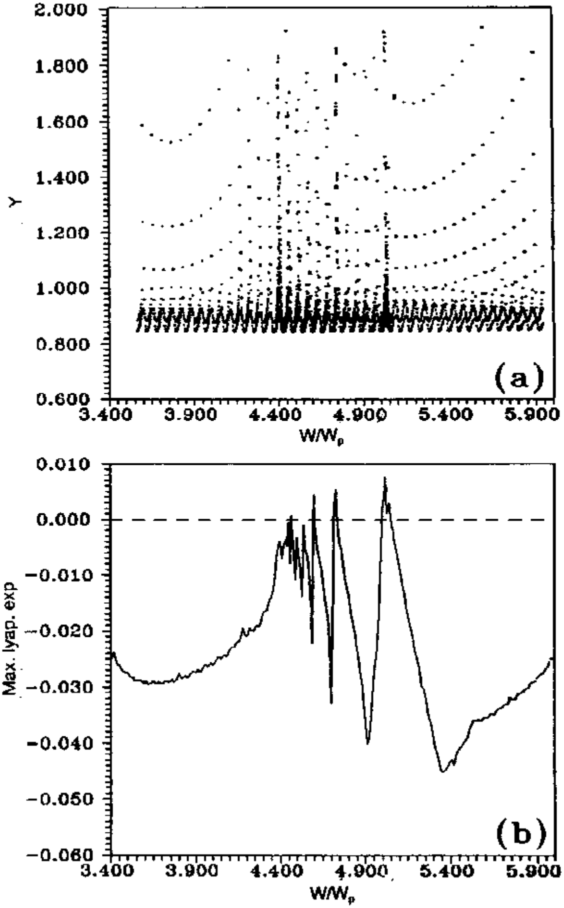

Fig. 10. (a) Enlargement of regions S3 and S4 of Figure (a) for the parameter range w/w$_p$ $\in$ [3.50, 6.00]. (b) Corresponding Lyapunov exponents specturm.

The maximum value reached by the yield (0.5765) is even higher than the yield (0.5644) corresponding to the autonomous system. Figure 12(a) shows an enlargement of a portion of Fig. 11(a) for the parameter range $w/w_p \in [0.185, 0.210]$. The maximum value of the yield (0.5765) occurs at the forcing frequency of $w/w_p = 0.2004$. Figure 11(b) shows the stroboscopic map for the region in question while the Lyapunov exponent spectrum is shown in Fig. 11(c). It can be observed that the yield increases through out the region of high periodicity and reaches its maximum in a region of chaotic behavior.

Numerical investigations have revealed then that yield of the autonomous periodic regime can be improved by appropriate choice of the forcing frequency.

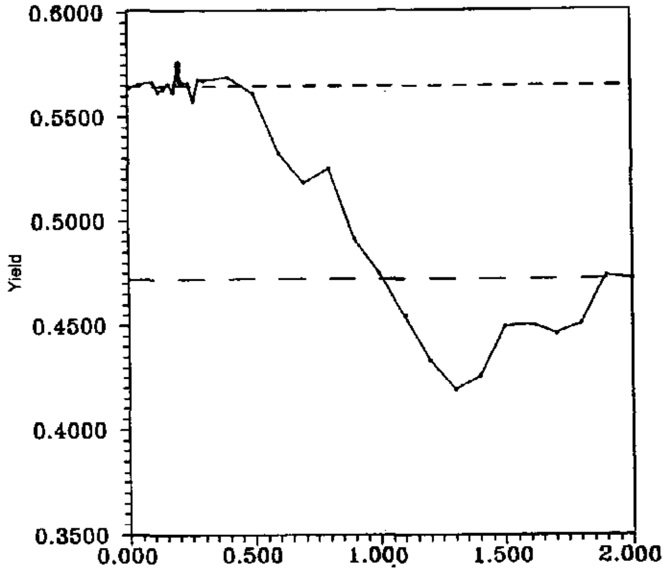

Fig. 11. Variations of the yield with the forcing frequency; dashed (-) yield for the autonomous system ($A_m = 0.0$); dashed (-) yield for the center of forcing ($A_m = 0.020$).

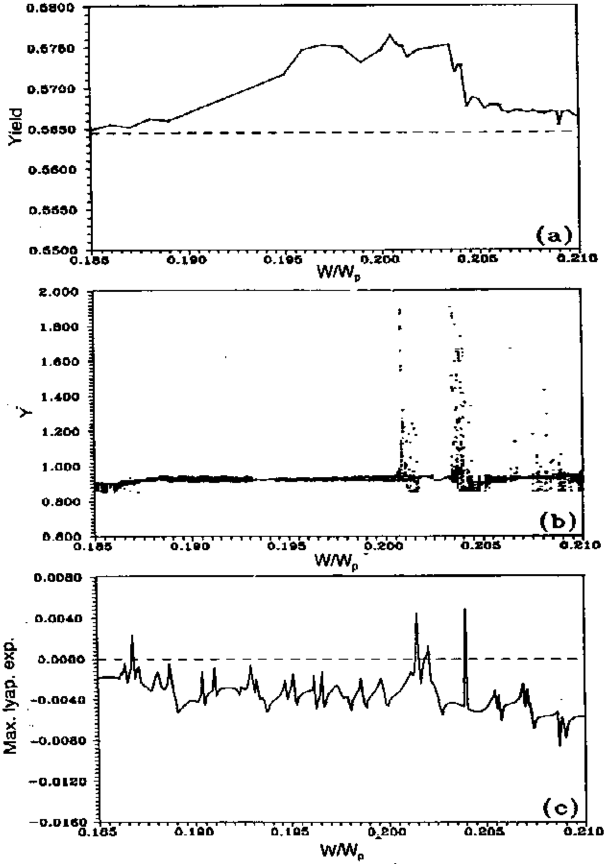

Fig. 12. (a) Enlargement of the yield variations for the parameter range w/wp $\in$ [0.185, 0.210]. The yield is higher than the yield of the autonomous system. (b) Corresponding stroboscopic diagram; (c) Corresponding Lyapunov exponent spectrum. The maximum yield occurs in a chaotic region.

Conclusions

An investigation of the chaotic behavior of a non-isothermal fluidized bed reactor when the feed temperature is periodically forced has been carried out in this paper. The investigation revealed the richness of the dynamic behavior of this system. Chaotic regimes emerge form the initially periodic system even for small amplitudes. Different mechanisms for the transition between chaotic and periodic regions has been analyzed, including period adding and saddle-node bifurcation. Numerical investigation has shown that the yield while decreasing with the change in the amplitude can be improved by the selection of appropriate forcing frequency.

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الخصائص الديناميكية لمفاعل محفّز، مميّع المهد ومستحث بطريقة دورية باستعمال التحكم النسبي التكاملي الكلاسيكي

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ملخص البحث. تم اختيار نموذج ثنائي الطور لمفاعل محفّز ومميّع المهد بوجود التفاعلات المتتابعة الطاردة للحرارة لدراسة الخصائص الديناميكية لهذه الوحدة المهمة صناعيا عندما تكون مستحثة بطريقة دورية. ويركّز البحث على دراسة تأثير حث النظام التلقائي بطريقة دورية ذات درجة عالية من الذبذبة.

تظهر نتائج البحث أنه بالحث الدوري لدرجة الحرارة المغذية فإن أنظمة من الفوضى تظهر عند نطاقات صغيرة من الحث. ففي حالة ثبات التذبذب وتغير مدى الحث فإن سلوك الوحدة يتراوح بين أنظمة دورية وفوضوية. وفي المقابل عند ثبات النطاق فإن أنظمة من الفوضى تظهر أيضا بتغير صغير لذبذبة الحث.

كما أن البحث يتضمن دراسة لتأثير هذه التغيرات على العائد من المنتج المرغوب. وقد أوضحت النتائج إمكانية زيادة العائد عن طريق الاختيار الملائم لذبذبة الحث.