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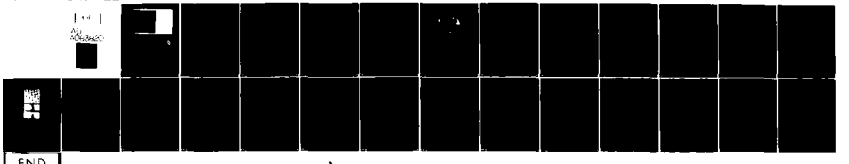

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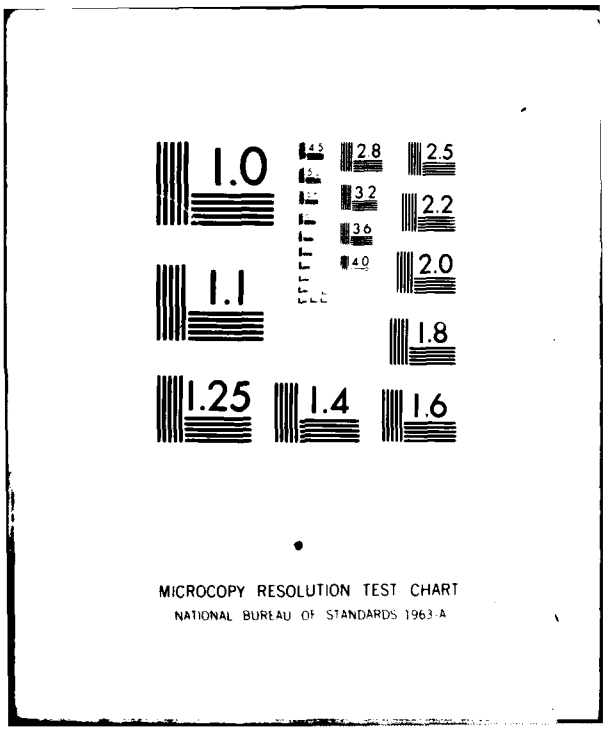
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# BIFURCATION PHENOMENA IN STIRRED TANKS AND CATALYTIC REACTORS

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# BIFURCATION PHENOMENA IN STIRRED TANKS AND CATALYTIC REACTORS

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## ABSTRACT

A review of the present state of understanding of continuous stirred tank reactors is presented along with certain new results on bifurcation phenomena for catalytic surfaces. A few examples are discussed which show repeated Hopf bifurcation, complex oscillations, and chaotic behaviour arising from a simple catalyst surface model. —> next page

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Key Words: Chemical systems, stability, stirred tank reactor, catalytic surfaces, limit cycles. Hopf bifurcation, complex oscillations, chaos.

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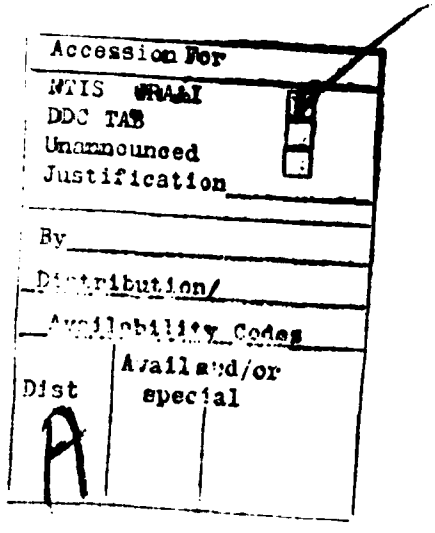
## SIGNIFICANCE AND EXPLANATION

This paper surveys the state of knowledge regarding the bifurcation behaviour of homogeneous stirred reactors and suggests a mechanism which can explain observed complex oscillations and apparent chaotic behaviour of catalytic reactors. The mathematical concepts brought to bear on this problem include static and Hopf bifurcation, multiple and secondary bifurcation, and singularity theory. The features arising in the problems discussed here are thought to be common to a wide range of chemically reacting systems.

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# BIFURCATION PHENOMENA IN STIRRED TANKS AND CATALYTIC REACTORS

W. H. Ray* and K. F. Jensen*

1. Introduction. The existence of bifurcation phenomena in common types of chemical reactors has been known for more than 150 years (e.g.; [1 - 3]) and yet new experimental and mathematical results are being discovered and reported even today. A rather good history of the these developments may be found in recent review articles (e.g. [4 - 6]). Probably the simplest type of chemical reactor which exhibits interesting bifurcation behaviour is the continuous stirred tank reactor (CSTR). Equally interesting are catalytic surfaces and catalytic reactors which are only slightly more complex systems but which provide a wide variety of bifurcation phenomena (e.g.; multiple steady states, both simple and complex non-linear oscillations, chaotic behaviour, standing and travelling waves, etc.) [4 - 7]. In this paper we plan to briefly discuss new results which have been reported for the CSTR and then move on to show some of the intriguing bifurcation phenomena which arise from a new model for catalytic surfaces.

*Mathematics Research Center and Department of Chemical Engineering, University of Wisconsin-Madison. This work was sponsored in part by the United States Army under Contract Number DAAG29-75-C-0024 and the National Science Foundation. Acknowledgement is made to the donors of the Petroleum Research Fund administered by the American Chemical Society for the partial support of this research. The authors are indebted to Tunde Ogunnaike for contributing his artistic talents to Figure 1.

2. The Continuous Stirred Tank Reactor. The continuous stirred tank reactor (CSTR) is a stirred pot (Figure 1) into which chemicals of a certain recipe flow continuously and are stirred and heated or cooled while undergoing chemical reaction. These reactors often occur in connected multiples which can have ever increasing static and dynamic complexity. The bifurcation behaviour of the CSTR has been studied for more than 25 years [6, 8-15] and yet qualitatively new results seem to continually appear. To begin the discussion let us consider the simplext case: a CSTR in which a single irreversible first order exothermic reactor is being carried out. The modelling equations take the form

$$\frac{dx_1}{dt} = -x_1 + Da(1-x_1)\exp\{\frac{x_2}{1+x_2/\gamma}\} \tag{1}$$

$$Le\,\frac{dx_2}{dt} = -x_2 + BDa(1-x_1)\exp\{\frac{x_2}{1+x_2/\gamma}\} - \beta(x_2-x_{2c}) \tag{2}$$

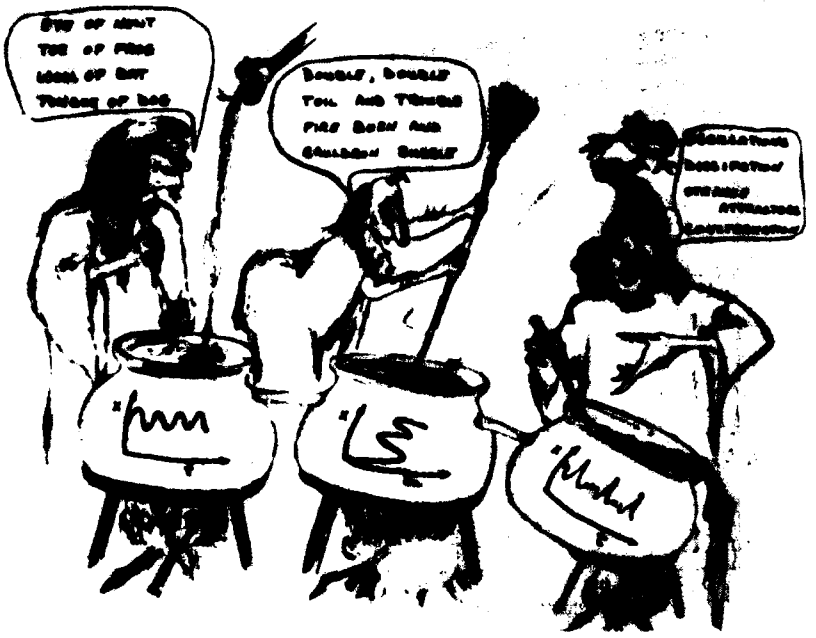

Figure 1. The Three Sisters

where $x_1$ denotes the conversion of reactant and $x_2$ a dimensionless temperature. The system parameters are B, a dimensionless heat of reaction, $\gamma$ a dimensionless activation energy, $x_{2c}$ a dimensionless coolant temperature, Le a ratio of characteristic times for transport, $\beta$ a dimensionless heat transfer coefficient, and Da a dimensionless ratio of reactor residence time to reaction time. If desired, these latter two parameters can be further parameterized in terms of the reactor residence time, $\tau$ ,

$$Da = Da_0\tau \tag{3}$$

$$\beta = \beta_0\tau \tag{4}$$

where $Da_0, \beta_0$ are reference parameters.

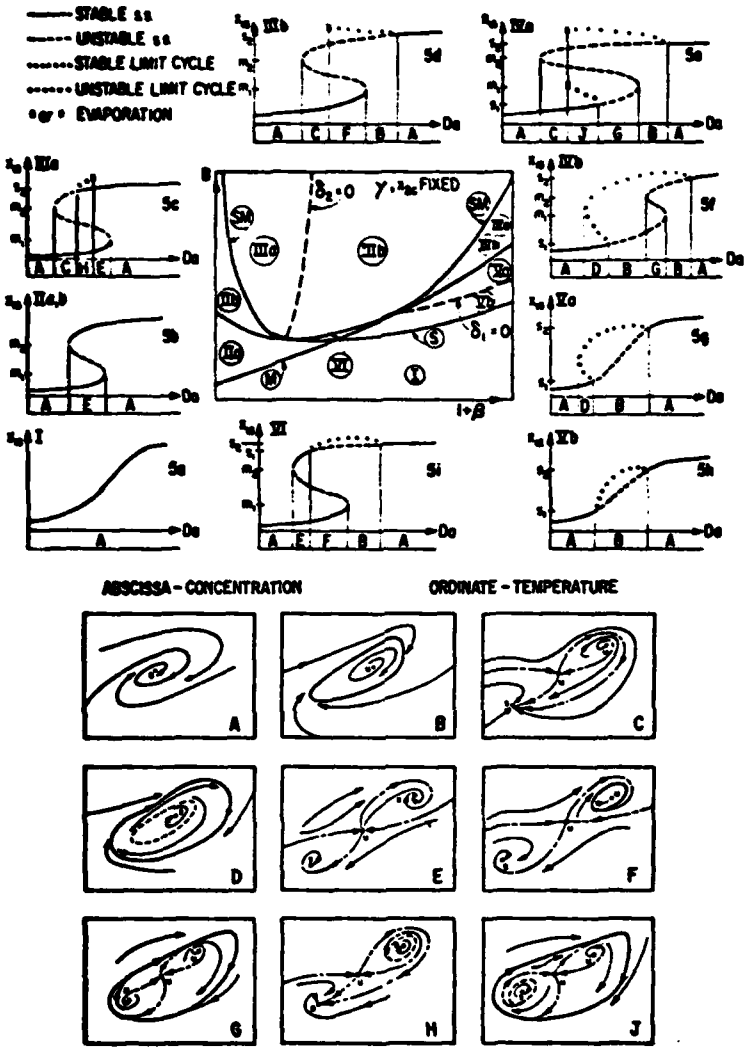


Figure 2. Bifurcation with respect to Da and resulting phase portraits.

The structure of the static and Hopf bifurcation behaviour for Equations (1,2) has been studied (e.g. cf [6, 12, 13]) and may be summarized in Figure 2. The formation of single limit cycles from saddle loops was conjectured in [13] as a means of explaining the

disappearance of limit cycles of large amplitude and the numerical computations in [13] support this conjecture. Recently, some questions have been raised [16,17] about the possible bifurcation of pairs of limit cycles from degenerate saddle loops. Speculations that this could lead to a vast array of new phase portraits have been made [17]; however, no computations have been

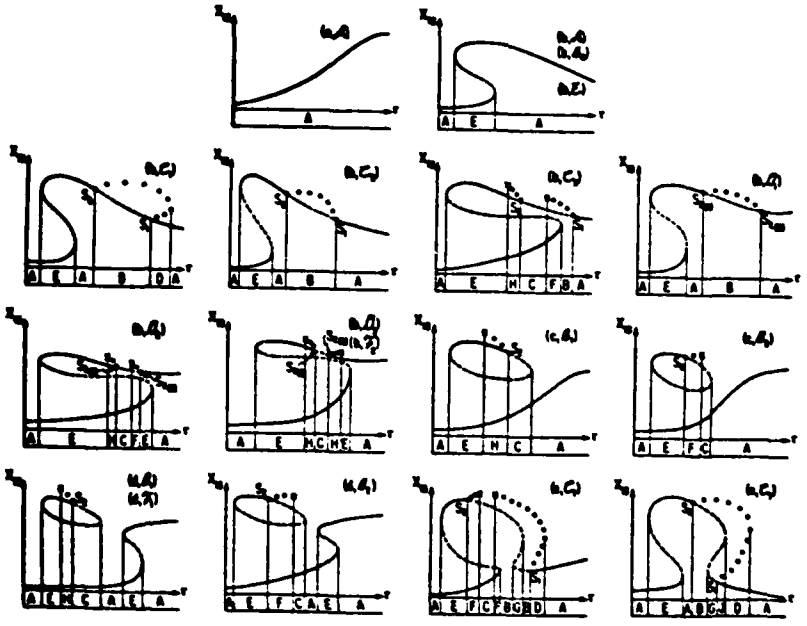

Figure 3. Bifurcation with respect to $\tau$ for $\gamma \to \infty$.

reported which show these new phase portraits actually exist. Some attempts at such calculations have been unsuccessful [16].

When the reparameterization given by Equation (3,4) is made there is very complex static and Hopf bifurcation behaviour. For the case where $\gamma \to \infty$, the structure has been calculated (cf. [14]) and may be seen in Figure 3. Recent results by Golubitsky and Keyfitz [18] as well as by Huang and Varma [19] seem to indicate that for relatively small values of $\gamma$ and extreme values of $x_{2c}$, even more variety in static bifurcation may occur (cf. Figure 4).

The effect of the Lewis number, $Le$ on the CSTR behaviour has been analyzed by Schmitz et al [20] and by Ray and Hastings [21]. In reference [20], theoretical predictions of bifurcation behaviour were found to agree very well with observed experimental results for the case of a second order exothermic reaction. Ray and Hastings [21] show that for sufficiently small values of $Le$, Hopf bifurcation occurs and the resulting limit cycle approaches a relaxation oscillation as $Le \to 0$.

If one considers more complex kinetics or multiple coupled stirred tank reactors, the behaviour becomes even more interesting. For example, Halbe and Poore [22] have carried out the bifurcation analysis for the case of two consecutive reactions $A \to B \to C$ in a CSTR and have shown secondary static branching and a rich variety of Hopf bifurcation behaviour. Recently, Aris and his

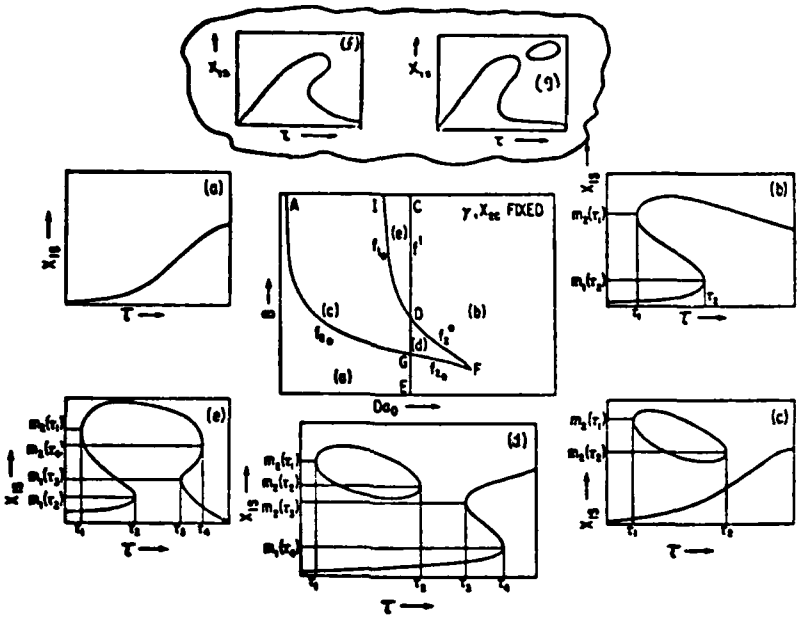


*Figure 4.* Static bifurcation with respect to $\tau$ for both finite and infinite $\gamma$ .

students [26] have also studied the bifurcation behaviour of this problem. Marek [23-25] and Aris [26] have considered the behaviour of systems of coupled stirred tanks, and their results illustrate quite clearly that for multiple CSTRs the story of CSTR bifurcation behaviour is far from complete.

3. Catalytic Reactors. There are many types of catalytic chemical reactors which exhibit interesting bifurcation behaviour. These range from the catalytic converter used in automobile exhaust purefication to large 10 story high fluidized bed catalytic crackers used in oil refineries. The oscillations, multiple steady states, and wave phenomena which arise in catalytic systems may be instigated by the catalyst, the reactor configuration or the interaction of both the catalyst and the reactor. (cf [4-7] and references therein for more details) However, those effects due solely to the reactor configuration are not specific to catalytic systems and would arise for non-catalytic reactions in the same reactor. Thus our discussion here will center on the bifurcation behaviour of catalytic surfaces.

Although there have been many chemical explanations put forth for the wide variety of dynamic behaviour exhibited by catalytic surfaces (e.g.; [4-7]), these phenomena are so pervasive that a physical explanation would seem plausible. Recently, the authors developed such a physical model for unsupported [27,28] and supported [29] catalytic surfaces.

For unsupported catalysts such as catalytic wires, the model arose from the following experimental observations:

(i) Completely smooth wires do not readily ignite, but must be "activated" by heat treatment which roughens the surface.

(ii) Smooth wires do not oscillate and analysis of simple models confirms that such oscillations should not be possible.

(iii) Roughened wires oscillate for a wide range of catalysts and reactant gases, and these oscillations tend to be very complex.

(iv) Optical observations of the roughened oscillating surface show very high oscillating temperatures locally on the roughened surface.

When one considers all of this evidence together, a new view of the dynamics of a catalytic metal surface emerges. In particular, it appears that the roughened surface of a catalytic wire or gauze plays a key role in the dynamic behaviour of the reaction. Electron micrographs of platinum and platinum alloy catalytic wires such as shown in Figure 5 [31] clearly show the presence of rough, porous protrusions on the surface of the catalytic wire. To mathematically model this "fuzzy" wire, we shall choose the simple picture shown in Figure 6. The protrusions on the wire surface are modelled as cylinders of radius $R_p$ and length $L_p$. These protrusions have a size distribution, $F(R_p)\ dR_p$ which defines the number of protrusions per unit area of bulk wire surface in each size range $R_p$ to $R_p + dR_p$. Each size protrusion can oscillate with a

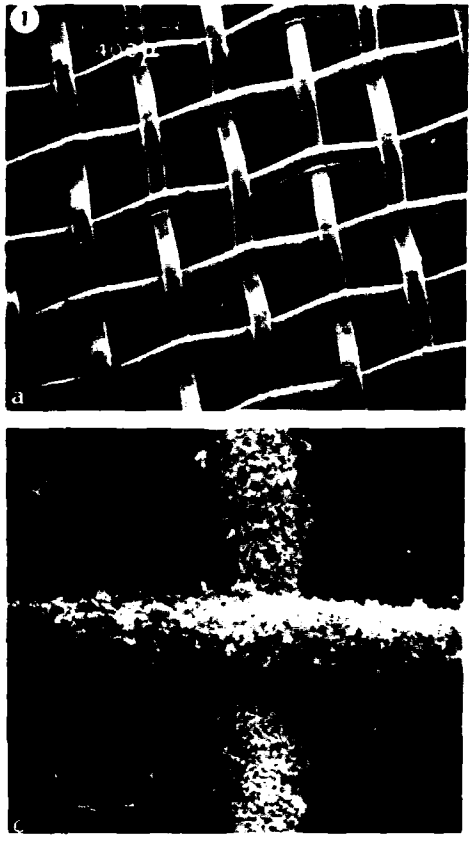

Figure 5. Scanning electron micrographs of platinum rhodium gauzes used for ammonia oxidation, above: smooth wires before use, below: rough wires after use. (Photograph kindly provided by Professor L. D. Schmidt.)

frequency dependent on $R_p$ and the distribution of protrusions can give rise to complex oscillations on the wire. Thermal communication between each protrusion and the bulk wire occurs at the end of the protrusion over a circular cross section of area

$\pi R_p^2$ . The fraction of the bulk wire surface covered by these protrusions is thus given by

$$\varepsilon = \int_0^\infty F(R_p)\pi R_p^2 \, dR_p \qquad 0 < \varepsilon < 1 \tag{5}$$

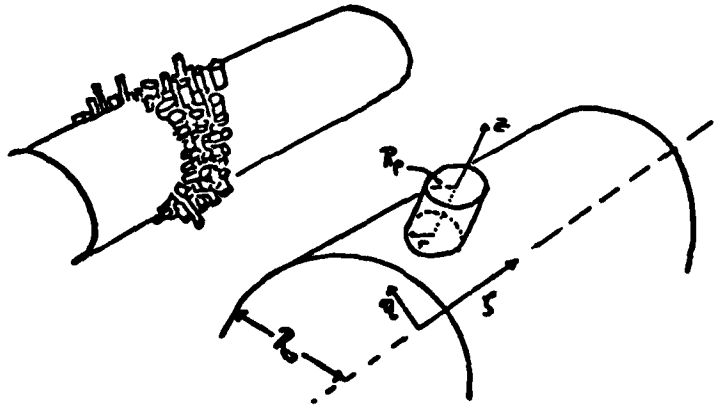

Figure 6. The fuzzy wire model.

and is assumed uniform over the entire wire surface. One may formulate heat and material balances for this composite system in a very general way [27,28]. However, a particularly simple model may be developed by assuming a first order irreversible reaction on the wire (in excess oxygen) with rate of mass transfer controlled by adsorption to the surface. If one assumes both the wire and protrusions to be uniform in temperature and concentration and the heat transfer between protrusions and wire to be modelled by Newton's law of cooling, then the modelling equations take the dimensionless form

$$\frac{dx_{1p}}{dt} = -x_{1p} + Da(1 - x_{1p}) \exp \frac{x_{2p}}{1 + x_{2p}/\gamma} \tag{6}$$

$$Le \frac{dx_{2p}}{dt} = -(1 + \beta)x_{2p} + BDa(1 - x_{1p}) \exp \frac{x_{2p}}{1 + x_{2p}/\gamma} + \beta x_{2w} \tag{7}$$

$$\frac{dx_{1w}}{dt} = -x_{1w} + Da(1 - x_{1w}) \exp \frac{x_{2w}}{1 + x_{2w}/\gamma} \tag{8}$$

$$Le_w \frac{dx_{2w}}{dt} = -(1 - \varepsilon + \beta_w \varepsilon)x_{2w} + \beta_w \varepsilon \langle x_2 \rangle + (1 - \varepsilon)B_w Da(1 - x_{1w}) \exp \frac{x_{2w}}{1 + x_{2w}/\gamma} \tag{9}$$

where $x_{1p}$, $x_{2p}$ are the composition and temperature of a protrusion of size $R_p$ and $x_{1w}$, $x_{2w}$ are the corresponding values on the bulk wire. Here

$$(10) \qquad \langle x_2 \rangle = \frac{\int_0^\infty a_c F(R_p) x_{2p} dR_p}{\int_0^\infty a_c F(R_p) dR_p}$$

is the dimensionless area average protrusion temperature. For purposes of simulation it is useful to assume a discrete set of protrusion sizes, $R_{p_i}$, so that

$$(11) \qquad F(R_p) = \sum_{i=1}^{N} n_{p_i} \delta(R_p - R_{p_i})$$

where $n_{p_i}$ is the number of protrusions/unit wire area of size $R_{p_i}$. Thus Equations (6 - 10) represent a set of $2N + 2$ model equations.

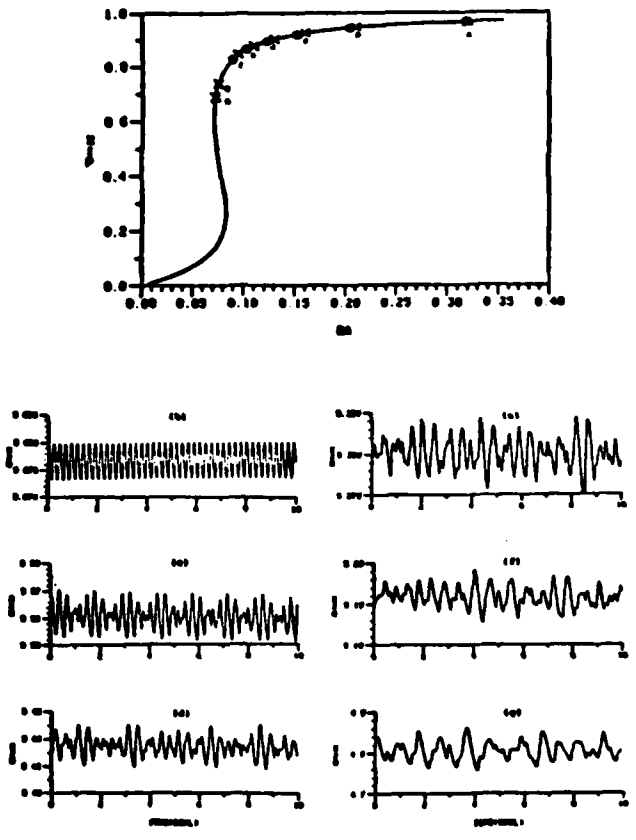

Figure 7. Hopf bifurcation points for the fuzzy wire model simulations, with varying catalyst activitiy, (.) bifurcation point, (x) simulation. The wire totally covered by six protrusion sizes evenly distributed, B = 6.0, Le = 0.20, 0.17, 0.14, 0.11, 0.08, 0.05, $\beta$ = 1.0, $\gamma$ =20.0, $\tau$ =2.0.

This model gives an amazing variety of ignition, extinction and oscillatory behaviour [28], but space only allows the presentation of a few results here. For example, in the case where there are 6 different protrusion sizes on the wire, there is secondary, tertiary, and higher order Hopf bifurcation as each size protrusion begins to oscillate with decreasing values of Da. The bifurcation points for the individual protrusions and the resulting complex dynamic behaviour of the entire wire is shown in Figure 7.

An even more interesting case involving only two protrusion sizes may be seen in Figure 8. Here as Da decreases the smaller protrusion bifurcates first and begins to oscillate as shown in Figure 8(d). Then as Da is decreased only slightly, the larger protrusion is "excited" and participates in a large complex oscillation in tandem with the smaller protrusion. As Da is decreased still further, the larger protrusion bifurcates and oscillations become less complex and more nearly regular. The fact that this rather complex oscillation arises for only two protrusion sizes on an inert bulk wire suggests that even simple interacting reaction systems can show a wide variety of dynamic behaviour.

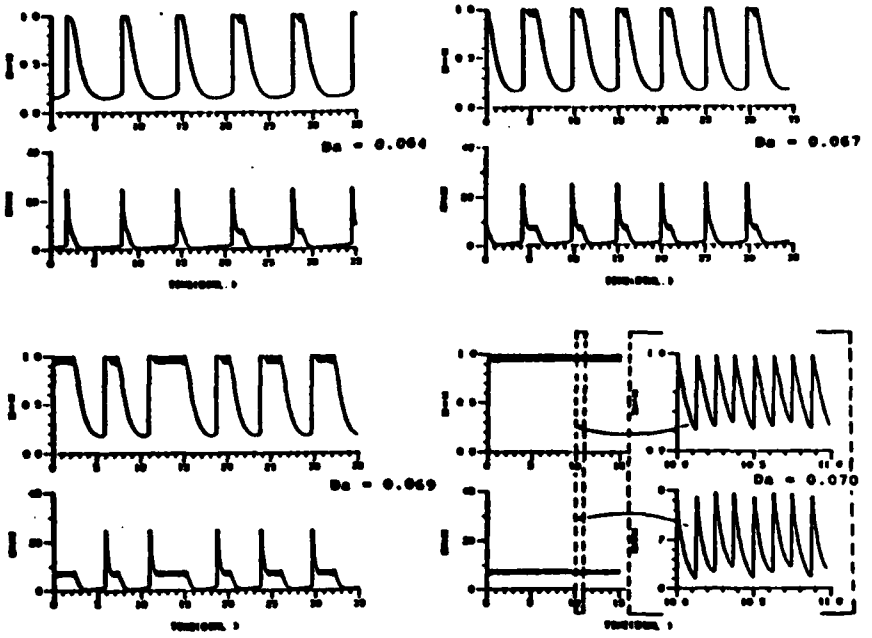

Figure 8. Bulk wire oscillations for varying catalytic activity. The wire is totally covered by equal areas of two protrusion sizes, B = 7.2, Le = 0.09, 0.005, $\beta$ = 2.0, $\gamma_0$ = 30.0, $\tau$ = 0.053
(a) Da = 0.064, (b) Da = 0.067, (c) Da = 0.069, (d) Da = 0.070.

Two forms of oscillating behaviour are possible when the bulk wire is only partially covered by protrusions. In the first case the bulk wire serves to attenuate the protrusion generated oscillations [27], while in the second and more interesting case the wire also sustains oscillations. A particularly interesting case arises when both the protrusion and wire are about the same size, in this case the Hopf-bifurcation points for wire and protrusion equations can coincide. Figure 9 illustrates the time evolution of the state variables, $x_{1p}$ and $x_{2w}$ for a wire half covered by protrusions of a uniform size as Da is decreased away from such a multiple Hopf-bifurcation point. Initially the oscillations are complex, but as Da decreases further they become single peaked due to synchronization of wire and protrusion dynamics.

To illustrate the prediction of apparent chaotic behaviour from our model and a comparison with experiment, we provide in Figure 10 a simulation with four protrusion sizes and the parameters for the catalytic oxidation of butane over Pt. Note that the chaotic temperature dynamics predicted by the model are in rather good agreement with the experimental observations of Edwards et al [30].

4. Concluding Remarks. From the very few examples presented here it is clear that commonly encountered chemical reactors provide an abundance of mathematically interesting bifurcation phenomena. In the case where the number of chemical reactor equations rises above two, then complex oscillations, chaos, and other exotic

behaviour are predicted even from simple models. The fact that these dynamic systems are also very important from the practical viewpoint, means that they make very good candidates for further study both by engineers and mathematicians.

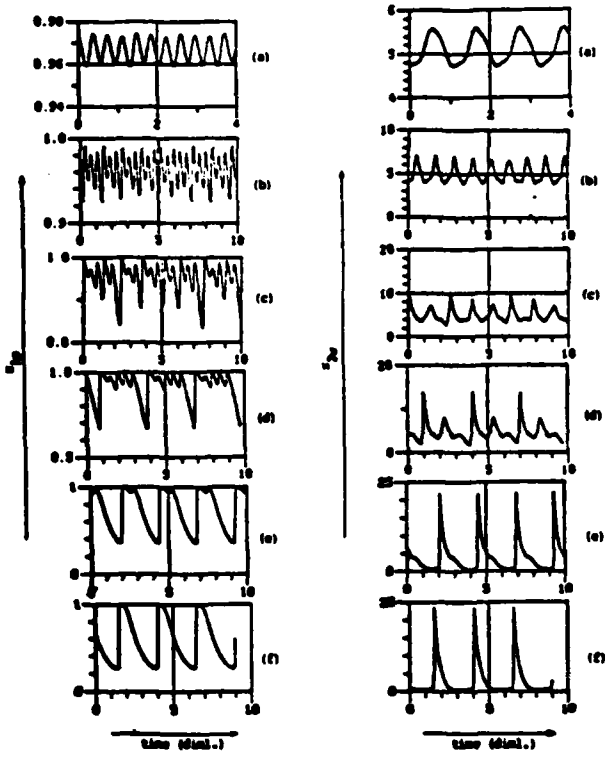

Figure 9. Oscillations in $x_{1p}$ and $x_{2w}$ for Da decreasing away from a multiple Hopf-bifurcation point Da = 0.1089, B = 8.32, B = 4.99, Le = 0.1, $Le_w$ = 0.1, $\beta$ = 0.1, $\beta_w$ = 0.3, $\gamma$ = 20, $\epsilon$ = 0.5 (a) Da = 0.1083, (b) Da = 0.1029, (c) Da = 0.09808, (d) Da = 0.09696, (e) Da = 0.09179, (f) Da = 0.07637.

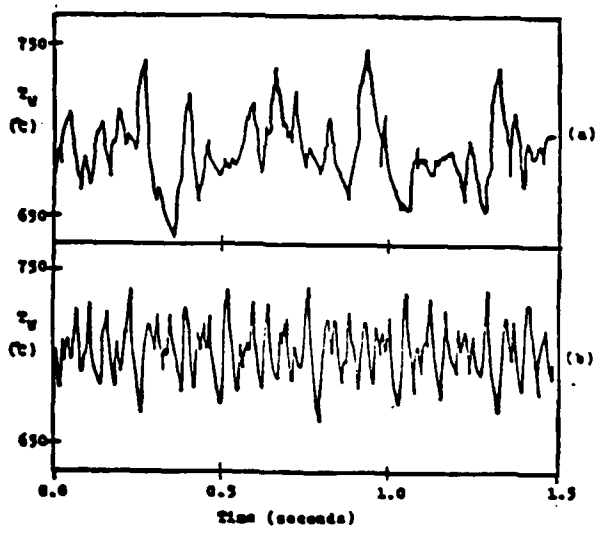


Figure 10. Comparison of experimentally observed and theoretically predicted oscillations in butane oxidation. (a) Experiments (Ref. [30]), (b) Fuzzy wire model prediction.

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

A review of the present state of understanding of continuous stirred tank reactors is presented along with certain new results on bifurcation phenomena for catalytic surfaces. A few examples are discussed which show repeated Hopf bifurcation, complex oscillations, and chaotic behaviour arising from a simple catalyst surface model.

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