# Stability of the Ignited State in Packed-Bed Reactor

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*The ignited state is associated with a thin reaction zone and a steep temperature gradient ahead of the front. The stability of this front is analyzed for small radial and azimuthal perturbations. The near-equidiffusional assumption is made, and the deviation of the Lewis number from unity is considered as the bifurcation parameter. The analysis shows that the planar front becomes unstable at Lewis numbers above unity after a Hopf bifurcation. The oscillatory front collapses into hot spots or rings which oscillate around the axial axis. For Lewis numbers less than one, a steady-state cellular front structure is found.*

## Introduction

Although packed-bed reactor models have been formulated, updated, and analyzed for more than 30 years, it still remains an area of interest. The existence of multiple fixed points and oscillatory solutions for certain parameter ranges has also been the focus of much research. Although most of the research was done on one dimensional (1-D) models, several studies of 2-D models were published. Most of those studies were focused on the estimation of parameters and the correction of lumped parameters which are used for 1-D models. Considerable work was also done to obtain some decision support for the choice of an appropriate model. Previous studies of instabilities of fixed-bed reactors were mostly done for 1-D models, e.g., Gatica et al. (1987) studied traveling wave behavior of nonadiabatic reaction systems.

In this study we will consider a 3-D pseudo-homogeneous model for an adiabatic packed-bed reactor. In particular we are interested in the stability of the solution when the reactor is operated in the ignited state. Although this is not usually the operating state, there are several reactions which are carried out in the ignited state, e.g., methane oxidation. The motivation for this research stems from discussions H. J. Viljoen held with engineers at SASOL (RSA). When reactors are scaled up, heat removal is usually a critical issue and it is a practice to merely duplicate a tested configuration (shell-and-tube reactors). However, industrial adiabatic reactors often have large diameters. When the reactor is designed for a combustion-like reaction, some interesting instabilities can occur. It is well-known in the combustion literature that planar flame fronts become unstable if the spatial domain in which the front exists is sufficiently large.

The practical implications of this research go beyond immediate effects. It is suspected that fluctuations in the position of the reaction front lead to large-scale mechanical failure of catalysts supports, sintering of catalysts, and excessive wear of reactor linings.

## Model

A pseudo-homogeneous model of the packed-bed reactor will be used in this analysis. It is assumed that the thermophysical properties are constant and the reactor is operated adiabatically. Admittedly this model is not the best choice for reactor design, but it is most amenable for the analysis of the reaction front by matched asymptotics. Furthermore, we expect that more sophisticated models will contribute at best quantitatively to the findings of this analysis.

The assumption of near-equidiffusivity will be used: the Lewis number is of the order of unity. The scales for length and time are chosen as $\eta\kappa/U$ and $\kappa/U^2$. Temperature and concentration will be scaled with the adiabatic temperature rise, $T_a - T_o$ and inlet concentration $c_o$. A dimensionless activation energy $\gamma$ is defined as $E/R_gT_a$ and the Damköhler

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number is defined as $k_o e^{-\gamma}\kappa/U^2$. The energy and species balances can then be written in nondimensional form as

$$\frac{\partial\theta}{\partial\tau}+\frac{\partial\theta}{\partial x}=\nabla\theta+DaCe^{\frac{\gamma(\theta-1)}{1+\sigma}} \tag{1}$$

$$\frac{\partial C}{\partial\tau}+\frac{\partial C}{\partial x}=\frac{1}{Le}\nabla C-DaCe^{\frac{\gamma(\theta-1)}{1+\sigma}}, \tag{2}$$

subject to the boundary conditions

$$\frac{d\theta}{dx}-\theta=\frac{1}{Le}\frac{dC}{dx}+1-C=0,\ x=0 \tag{3}$$

$$\frac{d\theta}{dx}=\frac{dC}{dx}=0,\ x=1 \tag{4}$$

$$\frac{d\theta}{dr}=\frac{dC}{dr}=0,\ r=R \tag{5}$$

and the system is closed by the obvious condition of periodicity in the azimuthal direction.

Since we use a thermal length scale which is relatively short, the dimensionless length of the reactors can become quite large; similarly, the radius can be large as well. However, in the analysis to come, we will show that the outlet conditions become disconnected from the eigenvalue problem and the crucial length variable is really the diameter.

## Stability Analysis

The analysis is based on the assumption that the activation energy is large. This assumption can become debatable for some catalyzed reactions, but for the oxidation of methane, e.g., the activation energy drops from 130 kJ/mol for the homogeneous reaction (Sathe et al., 1990) to 92 kJ/mol for the catalytic reaction (Chaouki et al., 1994). Even for the catalytic reaction, the activation energy is large enough to limit the reaction to a thin section of the reactor. As Norbury and Stuart (1987) described it, the large activation energy is acting like a switch and the reaction is turned off, except in the thin zone where the temperature is large enough. This concept forms the basis to analyze the reactor in three parts, a pre-heating zone, the reaction zone, and a post-combustion zone.

This type of analysis has been mostly done for problems defined on an infinite domain, where the steady state is associated with a freely propagating flame. An equivalent formulation to our problem is obtained by a coordinate transformation to the flame front, except that the unknown quantity is the speed of uniform propagation. In our problem, the flame is stabilized at some position in the reactor and we need to determine the position $F_o$ of the flame. It is well-known that the flame is anchored near the inlet. Solutions exist for flames near the outlet, but these solutions are unstable. For convenience sake, we will transform our coordinate system to one that is attached to the front. If the front's position is generally described by $x=F(r,\phi,\tau)$, we define $z=x-F(r,\phi,\tau)$. The inlet is at $z=-F_o$ for the basic solution.

Following Sivashinsky (1979, 1980), the basic solution will be sought in the form of a perturbation series, using $1/\gamma$ as the expansion parameter. The Lewis number is expressed as

$$Le=1-\frac{\lambda}{\gamma}. \tag{6}$$

If the basic solution is generically presented as

$$y_o=y_{oo}+\frac{1}{\gamma}y_{o1}+\ldots$$

the basic solutions to Eqs. 1–5 in the pre-heating and post-combustion zones are given by

$$\theta_{oo}=\begin{cases}e^z, & z<0\\ 1, & z>0\end{cases} \tag{7}$$

and

$$C_{oo}=\begin{cases}1-e^z, & z<0\\ 0, & z>0.\end{cases} \tag{8}$$

Before solving the next order of approximation, note that the following identity exists

$$C_{oo}+\theta_{oo}=1.$$

Let $P$ be defined as $\theta_1+C_1$, then $P_o$ $(=\theta_{o1}+C_{o1})$ is the solution of

$$\frac{d^2P_o}{dx^2}-\frac{dP_o}{dx}=\lambda e^x \tag{9}$$

and it is given by

$$P_o=\begin{cases}\lambda z e^z, & z<0\\ 0, & z>0.\end{cases} \tag{10}$$

Before we proceed with the linear stability analysis, we will analyze the reaction zone to determine the position of the flame inside the reactor and to find the necessary jump and continuity conditions across the reaction front. In order to analyze the reaction zone, the spatial variable is expanded as follows

$$v=\gamma z. \tag{11}$$

The temperature and concentration fields in the reaction zone are sought as perturbation series in $1/\gamma$. The Damköhler number is also expanded

$$\theta_f=\theta_{f0}+\frac{1}{\gamma}\theta_{f1}+\ldots \tag{12}$$

$$C_f=C_{f0}+\frac{1}{\gamma}C_{f1}+\ldots \tag{13}$$

$$Da=\gamma^2 Da_o+\ldots \tag{14}$$

Collecting terms of the same order, it follows that

$$\frac{d^2\theta_{f0}}{dv^2} = 0 \tag{15}$$

and matching the solution of this equation with $\theta_{oo}$, it follows that $\theta_{f0} = 1$. The following limits exist for the next order of approximation

$$\theta_{f1}(+\infty) = \theta_1(0,r,\phi,\tau) \tag{16}$$

$$C_{f1}(+\infty) = 0 \tag{17}$$

$$\frac{d\theta_{f1}(+\infty)}{dv} = \frac{d\theta_{oo}^{+}}{dz} \tag{18}$$

$$\frac{dC_{f1}(+\infty)}{dv} = \frac{dC_{oo}^{+}}{dz}. \tag{19}$$

In terms of the transformed coordinate system, the equations of order $(1/\gamma)$ are

$$\left(1+\left(\frac{\partial F}{\partial r}\right)^2+\frac{1}{r^2}\left(\frac{\partial F}{\partial \phi}\right)^2\right)\frac{\partial^2\theta_{f1}}{\partial v^2}+Da_oC_{f1}e^{\frac{\theta_{f1}}{1+\sigma}}=0. \tag{20}$$

$$\left(1+\left(\frac{\partial F^2}{\partial r}\right)+\frac{1}{r^2}\left(\frac{\partial F}{\partial \phi}\right)\right)^2\frac{\partial^2 C_{f1}}{\partial v^2}-Da_oC_{f1}e^{\frac{\theta_{f1}}{1+\sigma}}=0. \tag{21}$$

Adding Eqs. 20 and 21 and integrating once between $v$ and $+\infty$, one obtains

$$-\frac{d\theta_{f1}}{dv}+\frac{d\theta_{oo}^{+}}{dz}=-\frac{dC_{oo}^{+}}{dz}+\frac{dC_{f1}}{dv}. \tag{22}$$

Note that $d\theta_{oo}/dz + dC_{oo}/dz$ becomes zero identically. Equation 22 can be integrated between $v$ and $v \to +\infty$ and the following relationship between $C_{f1}$ and $\theta_{f1}$ is found

$$C_{f1} = P(0) - \theta_{f1}. \tag{23}$$

Note that $P(0) \equiv \theta_1$.

We consider $\gamma$ as a large but finite number; therefore, a lower bound exists for $v$, namely $v_{\min}$. Also recognize the fact that the flame is stabilized close to the inlet, i.e., $F \ll 1$, hence

$$F = \frac{1}{\gamma}\mathfrak{F}$$

and

$$v_{\min} = \mathfrak{F}.$$

Substituting for $C_{f1}$ from Eq. 23 in Eq. 20, it can be integrated between $v_{\min}$ and $v \to +\infty$ after multiplying by $2d\theta_{f1}/dv$. The integrated equation can be written as

$$0-\left(1+\left(\frac{\partial F}{\partial r}\right)^2+\frac{1}{r^2}\left(\frac{\partial F}{\partial \phi}\right)^2\right)\left(\frac{d\theta_{oo}}{dz}\right)^2 = 2Da_o(1+\sigma)\left[P(0)e^{-\frac{\mathfrak{F}}{1+\sigma}}+\mathfrak{F}e^{-\frac{\mathfrak{F}}{1+\sigma}} - (1+\sigma)\left(e^{\frac{P(0)}{1+\sigma}}-e^{\frac{\mathfrak{F}}{1+\sigma}}\right)\right]. \tag{24}$$

Since this result also holds for the basic case, i.e., $P(0) = 0$, we can solve for $\mathfrak{F} = \mathfrak{F}_o$

$$-1 = 2Da_o(1+\sigma)\left[\mathfrak{F}_oe^{-\frac{\mathfrak{F}_o}{1+\sigma}}-(1+\sigma)\left(1-e^{-\frac{\mathfrak{F}_o}{1+\sigma}}\right)\right]. \tag{25}$$

In the subsequent analysis, we can substitute $2Da_o(1+\sigma)$ in terms of the flame position for the basic solution $\mathfrak{F}_o$. The temperature gradient disappears in the post-combustion zone and Eq. 24 becomes the following jump condition

$$\sqrt{\left(1+\left(\frac{\partial F}{\partial r}\right)^2+\frac{1}{r^2}\left(\frac{\partial F^2}{\partial \phi}\right)\right)}\left[\frac{d\theta}{dz}\right] + \sqrt{\frac{\theta_1e^{-\frac{\mathfrak{F}}{1+\sigma}}+\mathfrak{F}e^{-\frac{\mathfrak{F}}{1+\sigma}}-(1+\sigma)\left(e^{\frac{\theta_1}{1+\sigma}}-e^{-\frac{\mathfrak{F}}{1+\sigma}}\right)}{\mathfrak{F}_oe^{-\frac{\mathfrak{F}_o}{1+\sigma}}-(1+\sigma)\left(1-e^{-\frac{\mathfrak{F}_o}{1+\sigma}}\right)}} = 0, \tag{26}$$

where $[\cdot] \equiv (\cdot)^+ - (\cdot)^-$.

The basic solution is perturbed by an infinitesimally small amount

$$\theta = \theta_{oo} + \epsilon\tilde{\theta} \tag{27}$$

$$P = P_o + \epsilon\tilde{P} \tag{28}$$

$$\mathfrak{F} = \mathfrak{F}_o + \epsilon\tilde{\mathfrak{F}} \tag{29}$$

The stability of the basic solution is described by the following eigenvalue problem

$$\frac{\partial\tilde{\theta}}{\partial\tau}+\frac{\partial\tilde{\theta}}{\partial z}=\frac{\partial^2\tilde{\theta}}{\partial z^2}+\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial\tilde{\theta}}{\partial r}\right)+\frac{1}{r^2}\frac{\partial^2\tilde{\theta}}{\partial\phi^2} - \frac{d\theta_{oo}}{dz}\left[\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial\tilde{\mathfrak{F}}}{\partial r}\right)+\frac{1}{r^2}\frac{\partial^2\tilde{\mathfrak{F}}}{\partial\phi^2}-\frac{\partial\tilde{\mathfrak{F}}}{\partial\tau}\right] \tag{30}$$

$$\frac{\partial \tilde{P}}{\partial \tau}+\frac{\partial \tilde{P}}{\partial z}=\frac{\partial^2 \tilde{P}}{\partial z^2}+\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial \tilde{\theta}}{\partial r}\right)+\frac{1}{r^2}\frac{\partial^2 \tilde{P}}{\partial \phi^2}$$
$$-\frac{dP_o}{dz}\left[\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial \tilde{\mathcal{F}}}{\partial r}\right)+\frac{1}{r^2}\frac{\partial^2 \tilde{\mathcal{F}}}{\partial \phi^2}-\frac{\partial \tilde{\mathcal{F}}}{\partial \tau}\right]$$
$$-\lambda\left\{\frac{\partial^2 \tilde{\theta}}{\partial z^2}+\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial \tilde{\theta}}{\partial r}\right)+\frac{1}{r^2}\frac{\partial^2 \tilde{\theta}}{\partial \phi^2}\right.$$
$$\left.-\frac{d\theta_{oo}}{dz}\left[\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial \tilde{\mathcal{F}}}{\partial r}\right)+\frac{1}{r^2}\frac{\partial^2 \tilde{\mathcal{F}}}{\partial \phi^2}\right]\right\}. \quad (31)$$

The following continuity and jump conditions hold at $z=0$

$$[\tilde{P}]=0 \quad (32)$$
$$[\tilde{\theta}]=0 \quad (33)$$
$$\left[\frac{\partial \tilde{P}}{\partial z}\right]=\lambda\left[\frac{\partial \tilde{\theta}}{\partial z}\right] \quad (34)$$
$$\left[\frac{\partial \tilde{\theta}}{\partial z}\right]=-\tilde{P}\mathcal{K}_1+\tilde{\mathcal{F}}\mathcal{K}_2 \quad (35)$$
$$\tilde{\theta}(z,r,\phi,\tau)=0,\ z>0, \quad (36)$$

The boundary conditions at $z=-F_o$ are

$$\frac{\partial \tilde{P}}{\partial z}=\tilde{P}+\lambda\frac{\partial \tilde{\theta}}{\partial z} \quad (37)$$
$$\frac{\partial \tilde{\theta}}{\partial z}=\tilde{\theta}. \quad (38)$$

The two functions $\mathcal{K}_1$ and $\mathcal{K}_2$ are obtained by linearizing Eq. 26; they are

$$\mathcal{K}_1=\frac{1-e^{-\frac{\mathcal{F}_o}{1+\sigma}}}{2(1+\sigma)\left[1-e^{-\frac{\mathcal{F}_o}{1+\sigma}}\left(1+\frac{\mathcal{F}_o}{1+\sigma}\right)\right]}$$
$$\mathcal{K}_2=\frac{\mathcal{F}_o e^{-\frac{\mathcal{F}_o}{1+\sigma}}}{2(1+\sigma)^2\left[1-e^{-\frac{\mathcal{F}_o}{1+\sigma}}\left(1+\frac{\mathcal{F}_o}{1+\sigma}\right)\right]}$$

The solution of the eigenvalue problem is

$$\tilde{\theta}=\begin{cases}(e^z+Ae^{\lambda_1 z}+Be^{\lambda_2 z})\tilde{\mathcal{F}}, & z<0\\ 0, & z>0.\end{cases} \quad (39)$$

$$\tilde{P}=\begin{cases}(\lambda e^z(1+z)+Cze^{\lambda_1 z}+De^{\lambda_1 z}+Gze^{\lambda_2 z}+He^{\lambda_2 z})\tilde{\mathcal{F}}, & z<0\\ Me^{\lambda_2 z}\tilde{\mathcal{F}}, & z>0\end{cases} \quad (40)$$

The perturbed flame front is

$$\tilde{\mathcal{F}}=aJ_n(k_r r)e^{i\omega\tau+in\phi}. \quad (41)$$

The complex values $\lambda_{1,2}$ are given by

$$\lambda_{1,2}=\frac{1+\sqrt{1+4(\omega+k_r^2+n^2)}}{2},\quad \lambda_1>\lambda_2.$$

Note that $\tilde{P}$ in the post-combustion zone does not contain any term of the form $e^{\lambda_1 z}$. The post-combustion length is $UL_r/\kappa\times(1-F_o)\approx UL_r/\kappa\gg 1$, hence $\tilde{P}^+$ will satisfy Eq. 4, approximately. In other words, the length of the reactor becomes disconnected from the eigenvalue problem. This approximation will become invalid in the case of extremely short beds. The coefficients A, B, C, D, G, H and M are defined in the Notation section. Define an overall wave number $K$ as

$$K^2=k_r^2+n^2.$$

The dispersion relation can be written as

$$\lambda=\frac{(\lambda_2-\lambda_1)\alpha_1\left(\frac{1+\lambda_1 A+\lambda_2 B-\mathcal{K}_2}{\mathcal{K}_1}\right)}{(\lambda_2-\lambda_1)(\alpha_1+\alpha_3 A+\alpha_4 B)+(\alpha_1-\alpha_2)\alpha_6}. \quad (42)$$

The definitions of $\alpha_1-\alpha_6$ are also given in the Notation section.

## Results

In the limit $\mathcal{F}_o\to\infty$, the dispersion relation takes the form

$$\frac{\lambda}{2(1+\sigma)}=\frac{(\lambda_1-\lambda_2)^2(\lambda_1-\lambda_2-1)}{\lambda_1-\lambda_2-1-2\omega} \quad (43)$$

which is exactly the result obtained by Sivashinsky (1979) for a freely propagating flame. However, it should be pointed out that $K$ can be considered continuous in an infinite domain, but in our case, where the reactor has a finite radius, only those $K$ values which also satisfy Eq. 5a are permissible. But we conclude that the behavior is correct in this limit.

Let us now consider some reactor conditions. The parameters have been chosen in accordance with the values published by Chaouki et al. (1994), for the oxidation of a 3% methane stream and a feed velocity of 0.1 m/s. For this example our length scale is $\eta\kappa/U=1/162.5$. First Eq. 25 is solved and a value of $\mathcal{F}_o=3.4$ is found. This is the dimensionless distance from the inlet of the reactor to the reaction front. The real distance can be determined by multiplying with the length scale. Flames are not stabilized near the outlet and the position close to the inlet is the realistic solution. This stationary, planar reaction front can become unstable. When $\omega=0$, a principal exchange of stability occurs and the result is shown in Figure 1. An important difference from the limiting case ($\mathcal{F}_o\to\infty$) is that a singularity develops as $K\to 0$. The minimum value of $\lambda$ is found at $K=0.9$. A Hopf type bifurcation occurs at negative values of $\lambda$. In Figure 2 the

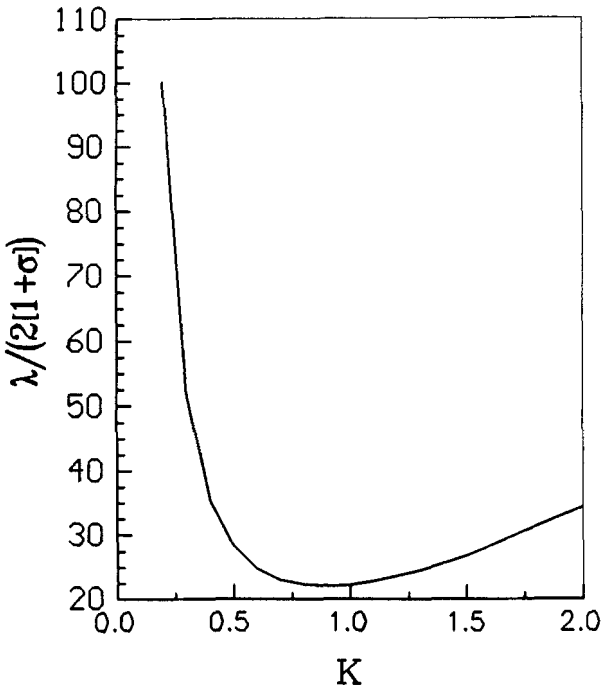


**Figure 1. Neutral stability for $\omega = 0$.**

dispersion relation is shown when $\omega \neq 0$. The function $\lambda(K)$ does not have a local maximum at $K > 0$. In previous results (for infinite domain) a local, albeit very slight maximum exists for $\lambda$ at $K > 0$. The maximum $\lambda$ for our case is at $K = 0$.

To bring these results in perspective with previous results on traveling waves, it is important to note that a stationary solution only exists for velocities in a certain range, i.e., $U \in \mathcal{I} = [U_{\min}, U_{\max}]$. If either bound is exceeded, the stationary solution ceases to exist and the front moves out of the bed. The analysis presented in this work deals with instabilities which occur for velocities which lie within the range $\mathcal{I}$.

Both figures represent a continuous relationship between $\lambda$ and $K$. However, for a specific reactor only certain points on these curves are permissible. Continuing with our example, let us further suppose that the reactor is 0.2 m in diameter. The dimensionless radius is $R = 0.1 \times 162.5 = 16.25$. First, neglect any angular variations, in other words $n = 0$, hence $K = k_r$. The reaction front is then described by

$$\mathcal{F} = \epsilon J_o(k_r r)$$

and $k_r$ is a root of

$$\frac{dJ_o(k_r r)}{dr} = J_1(k_r r) = 0, \quad r = R.$$

It follows from Figure 1 that the root closest to $K = k_r = 0.9$ defines the eigenfunction that exists near the minimum value of $\lambda$. Since $J_o(k_r r)$ is an oscillating function, there are an infinite number of roots, but we are interested in the one for which $| J_1(16.25k_r) - J_1(0.9 \times 16.25) |$ is a minimum. In this case it is the fifth root

$$16.25k_r = 13.324$$

Close to the bifurcation point, the shape of the front is described by superimposing the eigenfunction (multiplied by a small constant) on the planar solution. It takes on a wrinkled form, as shown in Figure 3, although the symmetry is not broken in the angular direction. As the diameter is increased, the minimum value of the bifurcation parameter ($\lambda$) is found at higher roots of Eq. 5a; hence more oscillations of the Bessel function can fit and the front will become even more wrinkled. Also note that $J_o$ is an oscillatory function with a decreasing amplitude, and the hot rings which form will be progressively weaker as one moves towards the perimeter. When

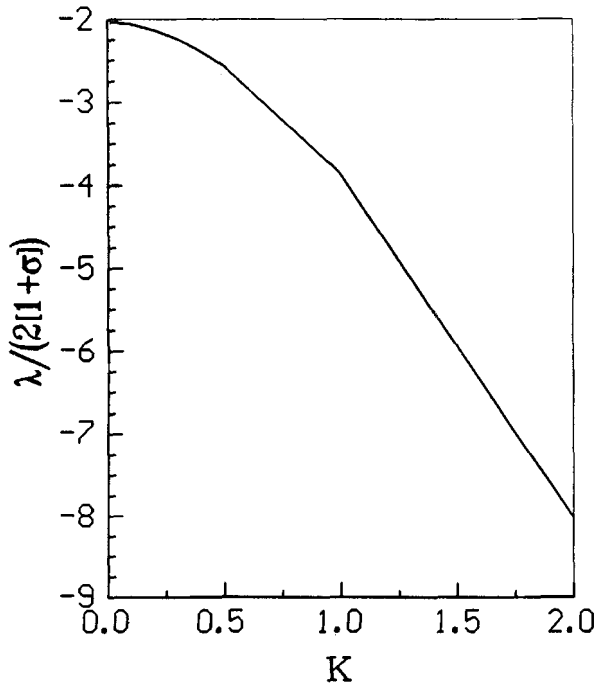


**Figure 2. Neutral stability for $\omega \neq 0$.**

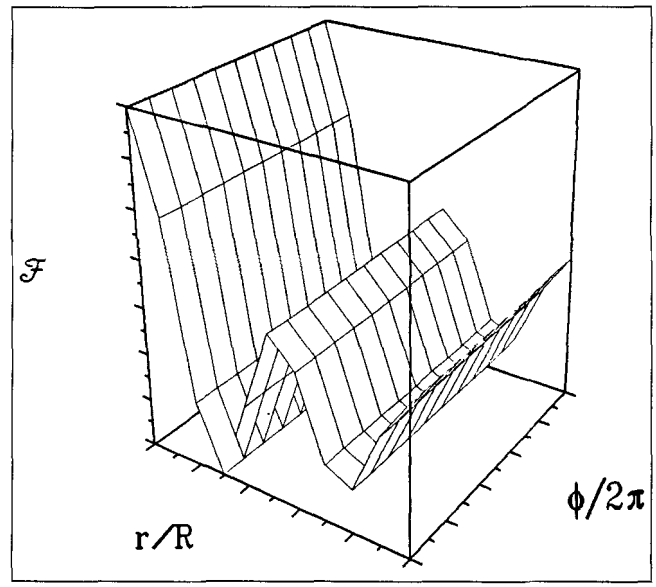


**Figure 3. Reaction front for $n = 0$ and $\omega = 0$.**

$K$ is larger than 1, eigenfunctions with nonzero azimuthal wave numbers come into consideration and $k_r^2 = K^2 - n^2$. However, different eigenmodes can start to compete with each other and a nonlinear analysis is required to describe the behavior properly.

Turning our attention to Figure 2, let us first comment on the feasibility of negative values for $\lambda$. In packed beds, the axial dispersion coefficient is considerably larger than diffusion coefficients. Formulas which have been advanced for the calculation of the dispersion coefficient (Wicke, 1975) include the diameter of the pellet, the superficial velocity and tortuosity factors. The Lewis number is determined by the choice of reactants and by reactor conditions and negative values for $\lambda$ are possible.

When $K = 0$, the stationary front becomes unstable without any changes in its transversal dimensions, i.e., no symmetry is broken in either the radial or the angular directions and the planar shape is preserved, but the front is no longer stationary. One possible scenario is that the front oscillates back and forth around $z = 0$, like a vibrating diaphragm. This instability may have interesting acoustic implications. Unfortunately, the linear analysis is not able to determine the amplitude of the oscillations. If this amplitude is twice as large as $\mathcal{F}_o$, the front moves upstream during one part of the period and can actually move out of the bed. At $K$ values between 1 and 2, an azimuthal wave number of 1 can be chosen while the radial wave number $k_r$ depends on the reactor diameter. To illustrate this case, consider again the diameter of 0.20 m (i.e., $R = 16.25$) and let $n = 1$. The front is now described by (cf. Eq. 41)

$$\mathcal{F} = \epsilon J_1(k_r r) e^{i\omega\tau + i\phi}$$

and $k_r$ is a root of the equation

$$\frac{dJ_1(k_r r)}{dr} = 0.$$

The first root is

$$16.25 k_r = 1.871.$$

The eigenfunction associated with these wave numbers has the form shown in Figure 4, and it is representative of the temperature on the front as well. A hot spot develops at the perimeter and a cooler spot diagonally across from it. As time progresses, the hot spot rotates around the center and Figure 4 represents only a snapshot in time. At larger diameters, higher roots of Eq. 5a can be chosen and hot spots can exist at the perimeter and on the interior, as described by the shape of the Bessel function. Higher azimuthal wave numbers lead to multiple hot spots at the perimeter or at the interior.

When the velocity increases, so does $\mathcal{F}_o$ and the neutral stability diagrams approach those for the infinite case, i.e., $\mathcal{F}_o \to \infty$. In practice, the maximum value for $\mathcal{F}_o$ is found just before blow-off occurs. It is evident that the behavior can be quite complex and the complexity increases for larger reactor diameters. A proper picture of the reaction front can only be constructed if a nonlinear analysis is done. The Kuramoto-Sivashinsky equation is an example of such an analysis. An equivalent of this equation should be derived for the reactor problem to get a better understanding of the behavior of reaction fronts in a packed bed.

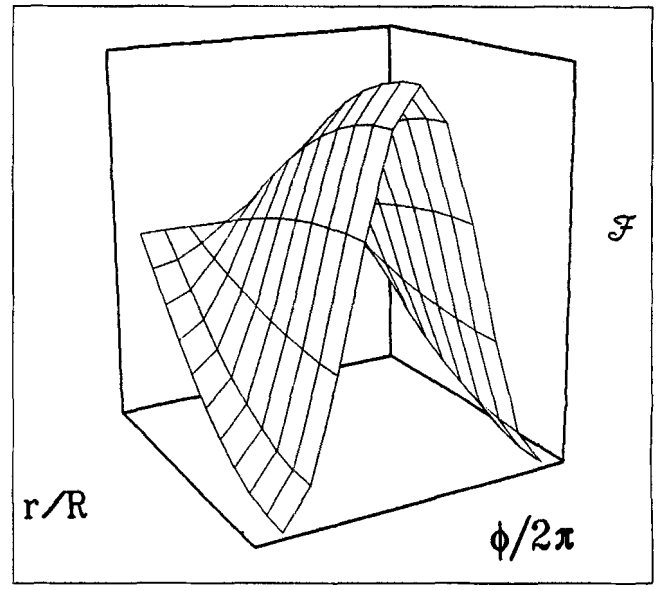


**Figure 4. Reaction front for $n = 1$ and $\omega \neq 0$.**


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## Notation

$c$ = concentration, mol/m$^3$
$C$ = dimensionless concentration, $c/c_o$
$Cp$ = specific heat capacity J/kg·K
$\mathfrak{D}$ = diffusion coefficient, m$^2$/s
$Da$ = Damköhler number, $k_o e^{-\gamma_\kappa}/U^2$
$E$ = activation energy, J/mol
$\mathcal{F}$ = flame position in expanded scale
$k$ = thermal conductivity, W/(m·K)
$k_o$ = frequency factor, s$^{-1}$
$k_r$ = radial wave number
$L_r$ = length of reactor, m
$Le$ = Lewis number, $\kappa/\mathfrak{D}$
$n$ = azimuthal wave number
$R$ = dimensionless radius
$R_g$ = universal gas constant
$t$ = time, s
$T$ = temperature, K
$U$ = velocity, m/s
$v$ = expanded axial coordinate
$x$ = axial coordinate
$z$ = translated axial coordinate

### *Greek letters*

$\gamma$ = dimensionless activation energy, $E/(R_g T_a)$
$\eta = [(\rho Cp)_{gas}]/[\rho Cp)_{eff}]$
$\theta$ = dimensionless temperature, $(T - T_o)/(T_a - T_o)$
$\kappa = k_{eff}/(\rho Cp)_{gas}$
$\lambda$ = deviation of Lewis number from unity (Eq. 6)
$\lambda_{1,2} = [1 \pm \sqrt{1 + 4(\omega + K^2)}]/2$, $\lambda_1 > \lambda_2$
$\rho$ = density, kg/m$^3$
$\sigma = T_o/T_a$

$\tau$ = dimensionless time, $\eta\kappa/U^2$
$\phi$ = azimuthal angle
$\omega$ = complex eigenvalue

### Subscripts

$a$ = adiabatic
eff = effective value
$f$ = reaction zone
gas = gas phase
$o$ = inlet

### Coefficients

$$A = \frac{-1}{1 + \frac{\lambda_1 - 1}{1 - \lambda_2} e^{(\lambda_2 - \lambda_1)F_o}}$$

$$B = \frac{\lambda_1 - 1}{1 - \lambda_2} e^{(\lambda_2 - \lambda_1)F_o} A$$

$$C = -\frac{\lambda A(\lambda_1^2 - K^2)}{1 - 2\lambda_1}$$

$$G = -\frac{\lambda B(\lambda_2^2 - K^2)}{1 - 2\lambda_2}$$

$$\alpha_1 = \lambda_2 - 1$$

$$\alpha_2 = (\lambda_1 - 1)e^{(\lambda_2 - \lambda_1)F_o}$$

$$\alpha_3 = \lambda e^{(\lambda_2 - \lambda_1)F_o}\left[\lambda_1 + \frac{\lambda_1^2 - K^2}{1 - 2\lambda_1}(1 + F_o - \lambda_1 F_o)\right]$$

$$\alpha_4 = \lambda\left[\lambda_2 + \frac{\lambda_2^2 - K^2}{1 - 2\lambda_2}(1 + F_o - \lambda_2 F_o)\right]$$

$$\alpha_5 = \frac{1 + \lambda_1 A + \lambda_2 B - \mathcal{K}_2}{\mathcal{K}_1}$$

$$\alpha_6 = \lambda\left(-\lambda_2 + 1 - \frac{\lambda_1^2 - K^2}{1 - 2\lambda_1} A - \frac{\lambda_2^2 - K^2}{1 - 2\lambda_2} B\right)$$

$$D = \lambda \frac{\alpha_6}{\lambda_2 - \lambda_1}$$

$$H = \lambda\left(\frac{\alpha_3 A}{\alpha_1} + \frac{\alpha_4 B}{\alpha_1}\right) - \frac{\alpha_2 D}{\alpha_1}$$

$$M = \lambda + D + H$$

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