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THE EFFECTS OF CURVATURE
ON ASYMMETRIC STEADY STATES
IN CATALYST PARTICLES

by

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ABSTRACT

The effects of curvature on steady states of chemical catalytic reactions is investigated by studying the cases of the catalytic particle being a spherical or cylindrical shell. Existence and stability of solutions are studied. It is shown that the solutions converge to the solutions for the catalytic slab when the curvature goes to 0 in each case.

The Model

We will use a model for catalyst particles proposed elsewhere (see [1],[3]). We assume that the particle is a homogeneous porous body, and that a catalyst is on the surface of this body. We are interested in the concentration of a limiting substance in a chemical reaction which occurs on the surface of the particle because of the catalyst present there. The limiting substance can diffuse through the particle, and its concentration outside of the particle is kept constant and uniform. We assume the reaction rates do not vary with time.

If we denote the concentration of the limiting substance by C , the diffusion coefficient by D , the coefficient of mass transfer at the surface by κ , and the concentrations of the limiting substance and the model reaction rates at the various surfaces of the particle (the inner and outer surfaces of a cylindrical shell, for example) by C_j and $r_j(C)$, respectively, we find that C satisfies the differential equation

$$\frac{\partial C}{\partial t} = D \Delta C \text{ inside the particle } \Omega^*,$$

$$(1) \quad C(x,y,z,0) = C_0(x,y,z)$$

$$\frac{D}{\kappa} \frac{\partial C}{\partial n} + C - C_j + r_j(C) = 0 \text{ for each boundary surface } \partial \Omega_j^*.$$

Here $\frac{\partial C}{\partial n}$ denotes the derivative of C in the direction normal to $\partial\Omega_j$ and out of Ω .

Using a model reaction rate of Arrhenius type, Pismen and Kharkats [4] were the first to point out the existence of asymmetric or nonconstant steady states for the problem in the slab $\{(x,y,z)|0 < x < L\}$ while assuming $c_1 = c_2$ and $r_1 = r_2$, and $c(x,y,z) = c(x)$, in short, a symmetric problem. Aronson and Peletier [1] and Luss et al. [3] discuss the stability of such solutions. In this paper we will discuss the existence and stability of similar radically symmetric, nonconstant solutions to the problem (1) with Ω^* being a cylindrical or spherical shell.

The model reaction rates $r_j(C)$ are usually assumed to be of Arrhenius type, but for the sake of computational simplicity when we do computations we will assume a model rate attributed to Marek [1],

$$(2) \quad r_j(C) = \frac{K_j^{1*} C}{(1 + K_j^{2*} C)^2}, \quad K_j^{1*}, K_j^{2*} > 0.$$

We will denote by R_1^* and R_2^* the inner and outer radii, respectively, of the cylindrical or spherical shell Ω^* . If we introduce the dimensionless variables $t = (D/(R_2^* - R_1^*)^2)t^*$, $(x,y,z) = (1/(R_2^* - R_1^*)) \cdot (x^*, y^*, z^*)$, $u(x,y,z,t) = C((R_2^* - R_1^*)(x,y,z), ((R_2^* - R_1^*)^2/D) \cdot t)$, then u satisfies the differential equation

$$(3) \quad \begin{aligned} \text{a) } & u_t = \Delta u \text{ in } \Omega, \\ & u(x,y,z,0) = u_0(x,y,z), \\ \text{b) } & \frac{\partial u}{\partial n} + \lambda f_j(u) = 0 \text{ on } \partial\Omega_j, \end{aligned}$$

with $\lambda = \kappa(R_2^* - R_1^*)/D$, and

$$(4) \quad \begin{aligned} f_j(u) &= u - \frac{C_j}{C_1} + \frac{K_j^{1*} u}{(1 + K_j^{2*} C_1 u)^2} \\ &= u - K_j^3 + \frac{K_j^1 u}{(1 + K_j^2 u)^2}. \end{aligned}$$

Here $K_j^1 = K_j^{1*}$, $K_j^2 = C_1 K_j^{2*}$ and $K_j^3 = C_j/C_1$.

Construction of Solutions

We will look for nonconstant steady state solutions of (3) for the cylindrical shell $\Omega = \{(x,y,z) | R_1^2 < x^2 + y^2 < R_2^2\}$ that have the property $u(x,y,z) = v(\rho)$, where $\rho = \sqrt{x^2 + y^2}$. The only functions harmonic in Ω having this property are of the form $u(x,y,z,t) = a \cdot \log(\rho) + b$. If we specify u to be p on $\partial\Omega_1 = \{\rho = R_1\}$ and to be q on $\partial\Omega_2 = \{\rho = R_2\}$, we have

$$(5) \quad u = \frac{q-p}{\log(R_2/R_1)} \log(\rho) + \frac{\log(R_2)p - \log(R_1)q}{\log(R_2/R_1)}.$$

Substituting this value for u into the boundary conditions 3(b), we find

$$\begin{aligned} -\frac{(q-p)}{\log(R_2/R_1)} \cdot \frac{1}{R_1} + \lambda f_1(p) &= 0 \quad \text{on } \partial\Omega_1 \\ \frac{(q-p)}{\log(R_2/R_1)} \cdot \frac{1}{R_2} + \lambda f_2(q) &= 0 \quad \text{on } \partial\Omega_2 \end{aligned}$$

or

$$\begin{aligned} q &= p + R_1 \log(R_2/R_1) \lambda f_1(p) \\ p &= q + R_2 \log(R_2/R_1) \lambda f_2(q). \end{aligned}$$

For a steady state solution to exist we must therefore have

$$\begin{aligned} 0 &= \lambda \log(R_2/R_1) (R_1 f_1(p) + R_2 f_2(q)) \\ \text{or} \\ (6) \quad (R_1/R_2) f_1(p) + f_2(q) &= 0 \end{aligned}$$

and also

$$\begin{aligned} 2(q-p) &= \lambda \log(R_2/R_1) (R_1 f_1(p) - R_2 f_2(q)) \\ \text{or} \\ (7) \quad \lambda &= \frac{2(q-p)}{R_2 \log(R_2/R_1) ((R_1/R_2) f_1(p) - f_2(q))}. \end{aligned}$$

In view of (6), (7) may be rewritten as

$$(8) \quad R_2 \log(R_2/R_1) \lambda = - \frac{(q-p)}{f_2(q)} .$$

Fixing λ and finding the possible values of p and q can be difficult, and we will do as others have done and graph λ against q .

This is done by picking q and finding the roots p of (6). There are at most three positive roots, because our reaction function f_j , like the Arrhenius reaction, is differentiable and increases monotonically from $f_j(0) < 0$ to a local maximum at a point we will denote by α_j , then decreases monotonically to a local minimum at β_j and then increases monotonically to ∞ . Such model equation will be called "suitable," and unless stated otherwise all of the following analyses holds for any "suitable" model equation. For each root p we may calculate a λ from (7), which must be positive to have physical significance.

We will have found a constant steady state solution if $p = q$, i.e. $(R_1/R_2)f_1(p) + f_2(p) = 0$. This will be a steady state only for $\lambda = 0$ if $f_1(p) \neq 0$, or for any $\lambda \geq 0$ if $f_1(p) = 0$. In particular this second case is true if $f_1 = f_2$.

We have derived the equation for a cylindrical shell. For the spherical shell $\Omega = \{R_1^2 \leq x^2 + y^2 + z^2 \leq R_2^2\}$ we may derive a similar set of equations, to wit

$$(6') \quad (R_1/R_2)^2 f_1(p) + f_2(q) = 0$$

$$(8') \quad (R_2/R_1) \lambda = - \frac{(q-p)}{f_2(q)} .$$

Note that both sets of equations are of the form

$$(9) \quad (R_1/R_2)^{n-1} f_1(p) + f_2(q) = 0$$

$$(10) \quad \mu = - \frac{(q-p)}{f_2(q)}$$

where $n=3$ for the spherical shell and $n=2$ for the cylindrical shell (and $n=1$ for the slab, for that matter), and so for each value of $(R_c/R_1)^{n-1}$ we

will graph μ against q . The correspondence between λ and μ is easy to see for each domain.

Numerical Results

A series of diagrams of μ versus q for the value of the parameter $(R_1/R_2)^{n-1} = 2/3$ using our specific model equations is given in Figures 1-7.

The branches of μ where the steady state solutions of the problem for the spherical shell are stable are indicated by solid lines, the rest by dashed lines. We make the conjecture that the solutions for the cylindrical shell have the same region of stability. The stability analyses are given in the next section.

The first set of diagrams were drawn with $f_1 = f_2$ (consequently $K_1^3 = K_2^3 = 1$). The value of $K^1 = K_1^1 = K_2^1$ was kept fixed, and the value of $K^2 = K_1^2 = K_2^2$ was changed to indicate the qualitative behavior of the μ vs. q diagrams. The values of K^1 and K^2 were chosen so that f_1 has three real positive roots. These roots are all necessarily between 0 and 1 because $f_1(0) = -1$ and the curve is otherwise above the line $y = u-1$.

Each of these roots provides us with a steady state solution for all μ , and these are indicated by the vertical lines in the diagram. As indicated, the two outside lines are stable, and the inner one is not.

In each diagram there is at least one interval, and sometimes two, where there are no steady state solutions u of (3) with the value q on the outside surface of the cylindrical or spherical shell for any positive μ . This can happen quite generally, even if $f_1 \neq f_2$. We will assume that f_1 is not necessarily equal to f_2 , but that both are "suitable" and that the position of the local extremes of f_2 , α_2 and β_2 , are contained in the smallest interval containing the roots of $f_1(x) = 0$. (This assumption allows us to ignore the behavior of f_1 and f_2 outside of the smallest intervals containing their respective zeros.) Then gaps occur if

$$(15) \quad \begin{aligned} (a) \quad & (R_1/R_2)^{n-1} f_1(\alpha_1) + f_2(\beta_2) < 0, \text{ or} \\ (b) \quad & (R_1/R_2)^{n-1} f_1(\beta_1) + f_2(\alpha_2) > 0. \end{aligned}$$

If (a) is true then there is an interval around β_2 for which there corresponds no positive μ for any q in the interval, because the solution of (9) yielding positive μ is impossible for such q ; similarly if (b) is true. If there is equality in (a) then there is a critical point at β_2 , and similarly for (b).

If we consider our specific model equation for the reaction rate and assume that $f_1 = f_2$, then there is a particularly elegant description of when equality in (15)a or (15)b will occur. As was pointed out by Kaper, et al. [2], for each value of K^1 there is a range of K^2 , depending on K^1 , such that if $K_L^2 < K^2 < K_U^2$ then f has exactly three positive real roots. This set $\{(K^1, K^2) | f \text{ has three positive real roots}\}$ forms a wedge, the vertex of which is (27,8). (Recall $K_1^3 = K_2^3 = 1$.) The upper and lower boundaries are asymptotically

$$(16) \quad K_U^2 = \frac{1}{4} K^1 + 1 + O(1/K^1) \quad \text{as } K^1 \rightarrow \infty \text{ and}$$

$$(17) \quad K_L^2 = 2(K^1)^{1/2} - 2 + O((K^1)^{-1/2}) \quad \text{as } K^1 \rightarrow \infty.$$

For $K^2 = K_L^2$, f_1 has a double root at β_1 , and for $K^2 = K_U^2$, f_1 has a double root at α_1 . Kaper et al. found that, for the slab, their single condition for a critical point was

$$f_1(\alpha_1) + f_1(\beta_1) = 0$$

and that this occurred when K^2 was exactly equal to the average of K_L^2 and K_U^2 . In our case the situation is completely analogous. We state, without proof, the following result.

Proposition: a) There is a critical point at $q = \alpha_1$ if

$$K^2 = \frac{K_L^2 + (R_1/R_2)^{n-1} K_U^2}{1 + (R_1/R_2)^{n-1}}.$$

b) There is a critical point at $q = \beta_1$ if

$$K^2 = \frac{(R_1/R_2)^{n-1} K_L^2 + K_U^2}{(R_1/R_2)^{n-1} + 1}.$$

This problem (3) for the cylindrical or spherical shell is singular in the following sense. If f_2 is any perturbation of f_1 such that f_1 and f_2 have different roots, then there is a solution of

$$(R_1/R_2)^{n-1}f_1(q_0) + f_2(q_0) = 0$$

for which $f_2(q_0) \neq 0$, and so $\mu = 0$ for this (constant) steady state. Because of at least one sided continuity at the point $(q_0, 0)$, there are arbitrarily small positive μ , and hence λ , for which there exist non-constant steady state solutions of (3). This is false for the case $f_1 = f_2$, for which there is a minimum $\mu_0 > 0$ such that, if $\mu < \mu_0$, there is no nonconstant steady state solutions corresponding to μ . This phenomenon is illustrated in [2].

Because the μ versus p curves are qualitatively different from the μ versus q curves, we include also several μ vs. p diagrams. Their qualitative behavior can be explained in a manner similar to the μ vs. q diagrams.

Stability

In this section we assume only that the f_j 's are differentiable. A linear stability analysis about a steady state solution u of (3) requires the study of the stability of the linearized equation

$$\begin{aligned} (18) \quad & v_t = \Delta v \quad \text{in } \Omega, \\ & v(x, y, z, 0) = v_0(x, y, z), \\ & \frac{\partial v}{\partial n} + \lambda f'_j(u)v = 0 \quad \text{on } \partial\Omega_j. \end{aligned}$$

From variational principles we know that all solutions v of (18) will converge to 0 if $f'_j(u) > 0$ everywhere, and some will blow up if $f'_j(u) < 0$ everywhere. Thus, the solution u of (3) is stable if $f'_j(u) > 0$ everywhere, and is unstable if $f'_j(u) < 0$ everywhere. We would like to find the other regions of stability for our special domains Ω and steady states u .

For the steady state solutions $u(x, y, z, t) = p + (q - p)x$ for problem (3) in the slab $\Omega = \{(x, y, z) | 0 < x < 1\}$ with $\partial\Omega_1 = \{x=0\}$, $\partial\Omega_2 = \{x=1\}$, we find stability if and only if

$$e^{2\sigma} = \frac{(\lambda f'_2(q) - \sigma)(\lambda f'_1(p) - \sigma)}{(\lambda f'_2(q) + \sigma)(\lambda f'_1(p) + \sigma)}$$

has no nonzero real solution σ . This condition is fulfilled if and only if

$$(19) \quad a) \quad f'_2(q) > 0 \quad \text{and} \quad f'_1(p) > 0, \quad \text{or}$$

$$b) \quad \lambda + \frac{1}{f'_1(p)} + \frac{1}{f'_2(q)} < 0 \quad \text{if} \quad f'_1(p)f'_2(q) < 0.$$

(See Pismen and Kharkats [4].)

We may derive a similar expression for the problem (3) in the spherical shell.

We assume a solution of the form $v(x, y, z, t) = e^{\sigma^2 t} w(x, y, z)$, where σ^2 is real, positive or negative, because (18) is a selfadjoint problem. We may derive an equation for w

$$(20) \quad a) \quad -\Delta w + \sigma^2 w = 0 \quad \text{in } \Omega$$

$$b) \quad \frac{\partial w}{\partial r} + \lambda f'_j(u) w = 0 \quad \text{on } \partial\Omega_j.$$

Now w must be radially symmetric, i.e. $w = w(\rho)$, since Ω , the differential operator, and the boundary conditions are radially symmetric and the problem is linear.

The solution w therefore is of the form

$$(21) \quad w(\rho) = a(\sigma\rho)^{-1} e^{\sigma\rho} + b(\sigma\rho)^{-1} e^{-\sigma\rho}.$$

Substituting this into the boundary conditions 20(b) and denoting $\lambda f'_1(p)$ by g_1 , and $\lambda f'_2(q)$ by g_2 we require

$$(22) \quad \begin{aligned} \frac{a}{R_2} e^{\sigma R_2} \left(1 + \frac{R_2 g_2 - 1}{\sigma R_2} \right) + \frac{b}{R_2} e^{-\sigma R_2} \left(-1 + \frac{R_2 g_2 - 1}{\sigma R_2} \right) &= 0 \\ \frac{a}{R_1} e^{\sigma R_1} \left(-1 + \frac{R_1 g_1 + 1}{\sigma R_1} \right) + \frac{b}{R_1} e^{-\sigma R_1} \left(1 + \frac{R_1 g_1 + 1}{\sigma R_1} \right) &= 0. \end{aligned}$$

If this set of linear equations has a nonzero solution (a,b) for some nonzero real σ , then σ^2 will be positive and v will be unbounded in time. Thus we wish to see, for fixed values of R_1 and R_2 , for which values of g_1 and q_2 is there a real nonzero σ such that the determinant of (21) is zero.

This condition can be written as

$$(23) \quad e^{2\sigma} = \frac{\left(\frac{R_2 q_2 - 1}{R_2} - \sigma\right) \left(\frac{R_1 q_1 + 1}{R_1} - \sigma\right)}{\left(\frac{R_2 q_2 - 1}{R_2} + \sigma\right) \left(\frac{R_1 q_1 + 1}{R_1} + \sigma\right)}.$$

(Recall $R_2 - R_1 = 1$.)

For stability we require that there be no real roots σ of this equation. This occurs if and only if

$$(24) \quad \begin{aligned} & \text{a) } R_2 q_2 - 1 > 0 \quad \text{and} \quad R_1 q_1 + 1 > 0, \quad \text{or} \\ & \text{b) } (R_2 q_2 - 1)(R_1 q_1 + 1) < 0 \quad \text{and} \end{aligned}$$

$$1 \leq - \left(\frac{R_2}{R_2 q_2 - 1} + \frac{R_1}{R_1 q_1 + 1} \right).$$

(Compare with (19).)

These conditions can be reformulated as

$$(25) \quad \begin{aligned} & \text{a) } q_2 = \lambda f'_2(q) > 0 \quad \text{and} \quad g_1 = \lambda f'_1(p) > 0, \quad \text{or} \\ & \text{b) } f'_2(q) f'_1(p) < 0 \quad \text{and} \end{aligned}$$

$$\frac{R_2}{R_1} \lambda + \frac{1}{(R_1/R_2)^2 f'_1(p)} + \frac{1}{f'_2(q)} \leq 0.$$

It was pointed out by Aronson and Peletier [1] for $f_1 = f_2$ for the slab that condition (19)b is equivalent to the statement that $f'_1(p) f'_2(q) < 0$ and $\frac{d\lambda}{dq} > 0$. It is an easy derivation from (6') and (8') to conclude the following result.

Proposition. The radially symmetric steady state solution u of (3), where Ω is a spherical shell, taking the values p and q on the inner and outer surfaces of radii R_1 and R_2 respectively is stable to small perturbations if and only if

$$(26) \quad \begin{aligned} & \text{a) } f'_1(p) > 0 \text{ and } f'_2(q) > 0 \quad \text{or} \\ & \text{b) } f'_1(p)f'_2(q) < 0 \quad \text{and} \quad \frac{d\lambda}{dq} > 0 \quad /// \end{aligned}$$

A similar analysis may be made when Ω is a cylindrical shell. There the analysis is complicated because the form of w is

$$(27) \quad w = aI_0(\sigma\rho) + bK_0(\sigma\rho)$$

where $I_\nu(z)$ and $K_\nu(z)$ are modified Bessel functions of order ν . The analysis gives

Proposition. The radially symmetric steady state solution u of (3), with Ω a cylindrical shell, taking the values p and q on the inner and outer surfaces of radii R_1 and R_2 respectively is stable to small perturbations if and only if:

there is no real solution σ of the equation

$$(28) \quad \begin{aligned} & [\lambda f'_1(p)K_0(R_1\sigma) + \sigma K_1(R_1\sigma)] \times [\lambda f'_2(q)I_0(R_2\sigma) + \sigma I_1(R_2\sigma)] \\ & - [\lambda f'_1(p)I_0(R_1\sigma) - \sigma I_1(R_1\sigma)] \times [\lambda f'_2(q)K_0(R_2\sigma) + \sigma K_1(R_2\sigma)] = 0 \quad /// \end{aligned}$$

We conjecture that condition (28) is equivalent to condition (26).

Limiting Cases

As $(R_1/R_2) \rightarrow 1$, each of our shells looks more and more like a slab. We would therefore expect that, given q , our steady state solutions u , our λ 's and the regions of stability, for each of these shells, converge to their counterpart in the slab. For each q the corresponding p , u and λ for the slab will be denoted by p_s , u_s and λ_s .

We will have occasion to use the functions

$$(29) \quad \begin{array}{ll} h_1 : [0, \alpha_1] \rightarrow R & h_1(x) = f_1(x) \\ h_2 : [\alpha_1, \beta_1] \rightarrow R & h_2(x) = f_1(x) \\ h_3 : [\beta_1, \infty] \rightarrow R & h_3(x) = f_1(x) \end{array}$$

These are restrictions of f_1 that are 1-1 and hence have well defined converses.

We will deal first with the spherical shell. We may rewrite (6') as

$$p(q) = h_j^{-1}(f_2(q) \times (R_2/R_1)^2)$$

for each branch h_j of f . As $R_1/R_2 \rightarrow 1$, since h_j^{-1} is continuous $p(q) \rightarrow p_s$. We also have that

$$\lambda = -(R_1/R_2) \frac{(q-p)}{f_2(q)} \rightarrow -\frac{(q-p_s)}{f_2(q)} = \lambda_s$$

as $R_1/R_2 \rightarrow 1$.

What we'd like to show is that, with $u = u(\rho)$, $\|u''\|_{L^\infty[R_1, R_2]} \rightarrow 0$, because then u would converge to the straight line joining q and p_s , i.e. u_s . The solution is

$$u(\rho) = \frac{-R_1 R_2 (q-p)}{\rho} + R_2 q - R_1 p.$$

If we differentiate twice with respect to ρ we get

$$u''(\rho) = \frac{2R_1 R_2 (p-q)}{\rho^3} \rightarrow 0 \quad \text{for } \rho \in [R_1, R_2] \text{ as } R_1 \rightarrow \infty.$$

Hence $u \rightarrow u_s$ in the suitable sense.

The sign of $f_1(p)$ is obviously the same as the sign of $f_1(p_s)$ (the opposite of the sign of $f_2(q)$), and so the question of stability is settled if $\frac{d\lambda}{dq} \rightarrow \frac{d\lambda_s}{dq}$ as $R_1/R_2 \rightarrow 1$. This can be shown by direct calculation.

For the case of the cylindrical shell we may show that $u \rightarrow u_s$, $p \rightarrow p_s$, and $\lambda \rightarrow \lambda_s$ in a completely analogous fashion, using the form of the solution (5). Because of the form of the stability condition (28) we were not

able to show the convergence of the curves of stable solutions (although if our conjecture there were true, it would follow immediately, as before).

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Figure 1
Solutions of (9),(10) with $K_2=50$, $K_1=12.594684$

Figure 2
Solutions of (9),(10) with $K_2=50$, $K_1=12.598694$

Figure 3
Solutions of (9),(10) with $K_2=50$, $K_1=12.602673$

Figure 4
Solutions of (9),(10) with $K_2=50$, $K_1=12.765120$

Figure 5
Solutions of (9),(10) with $K_2=50$, $K_1=12.928872$

Figure 6
Solutions of (9),(10) with $K_2=50$, $K_1=12.931540$

Figure 7
Solutions of (9),(10) with $K_2=50$, $K_1=12.934197$

Figure 1'

Solutions of (9),(10) with $K_2=50$, $K_1=12.594684$

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