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Chemical Engineering Science, Vol. 43, No. 6, pp. 1410–1413, 1988.
Printed in Great Britain.
0009–2509/88 $3.00 + 0.00
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Effectiveness factors and selectivity for parallel catalytic reactions with Langmuir–Hinshelwood kinetics

(*First received* 7 *January* 1987; *accepted in revised form* 1 *October* 1987)

INTRODUCTION

Today effectiveness factor calculations is one of the most important and cumbersome problem encountered in chemical reactor engineering, particularly in the design of fixed-bed reactors in which complex reactions are taking place with diffusional limitations inside the particle [1]. But, since the differential equations for the pellet constitute a nonlinear boundary value problem, a considerable computational effort is involved in the fixed-bed calculations. However, the computational time can be significantly reduced via algebraic equations expressing the effectiveness factors in terms of system parameters, especially in the case of parallel reactions.

The importance of avoiding numerical calculations of the effectiveness factors in the design of catalytic reactors has been recently emphasized by Froment and Bishoff [2].

In the last few years some efforts have been reported on estimating the effectiveness factor through simple rational expressions based on the asymptotic behavior of η for large and small values of the Thiele modulus [3–7], at least for the case of a single reaction, but only a few for reaction networks [3, 8–10]. Here a new expression [3, 4] is used to analyze coupled parallel reactions with kinetic expressions of the type generally encountered in catalysis, such as those of Langmuir–Hinshelwood.

In this contribution, different hypothetical cases are analyzed as well the specific cohydrogenation of tetraline and paraxylene on a nickel catalyst where the selectivity is also estimated.

ANALYSIS

Consider the case that two species B and C react with a common reactant A to give products, among them P_1 and P_2. The reactions take place in a isothermal catalytic porous slab of half thickness L:

$$A + \nu_B B \rightarrow \nu_{P_1} P_1 \tag{1}$$

$$A + \nu_C C \rightarrow \nu_{P_2} P_2 \tag{2}$$

The reaction rate is given by the Langmuir–Hinshelwood expressions

$$r_1 = \frac{k_1 C_A'^{p} C_B'^{m}}{[1 + K_A C_A' + K_B C_B' + K_C C_C' + K_{P_1} C_{P_1}' + K_{P_2} C_{P_2}]^{\alpha}} \tag{3}$$

$$r_2 = \frac{k_2 C'^q_A C'^n_C}{[1 + K_A C'_A + K_B C'_B + K_C C'_C + K_{P_1} C'_{P_1} + K_{P_2} C'_{P_2}]^\alpha} \quad (4)$$

The dimensionless mass balances for the different species participating in the system can be written as

$$\frac{d^2 C_A}{dX^2} = h_1^2 r_1^* + h_2^2 r_2^* \quad (5)$$

$$\frac{d^2 C_B}{dX^2} = \gamma_B h_1^2 r_1^* \quad (6)$$

$$\frac{d^2 C_C}{dX^2} = \gamma_C h_2^2 r_2^* \quad (7)$$

$$\frac{d^2 C_{P_1}}{dX^2} = -\gamma_{P_1} h_1^2 r_1^* \quad (8)$$

$$\frac{d^2 C_{P_2}}{dX^2} = -\gamma_{P_2} h_2^2 r_2^* \quad (9)$$

where

$$C_i = \frac{C'_i}{C'_{i_S}} \quad h_1^2 = L^2 r_{1S}/D_A C'_{A_S} \quad h_2^2 = L^2 r_{2S}/D_A C'_{A_S}$$

$$\gamma_i = (D_A C'_{A_S}/D_i C'_{i_S})\nu_i \text{ for } i = B, C, P_1 \text{ or } P_2 \quad (10a, b)$$

$$X = x/L$$

and

$$r_i^* = r_i/r_{i_S} \quad i = 1, 2. \quad (11)$$

The subscript S denotes properties evaluated at the external surface of the pellet.

With the boundary conditions:

$$C_i = 1 \quad \text{at} \quad X = 0$$

and (12a, b)

$$\frac{dC_i}{dX} = 0 \text{ at } X = 1 \text{ with } i = A, B, C, P_1 \text{ or } P_2$$

the following relations between dimensionless concentrations are obtained:

$$C_A - C_B/\gamma_B - C_C/\gamma_C = 1 - (1/\gamma_B + 1/\gamma_C) = R \quad (13)$$

$$C_B = 1 + \gamma_B/\gamma_{P_1}(1 - C_{P_1}) \quad (14)$$

$$C_C = 1 + \gamma_C/\gamma_{P_2}(1 - C_{P_2}). \quad (15)$$

It will be assumed that $R > 0$, which means that the concentration of species A can never vanish.

The main difficulty in solving this system is the unknown relationship between C_B and C_C. At low h_1 and h_2 values this difficulty can be easily overcome but the system cannot be uncoupled when h_1 and h_2 are $\gg 1$. Cuckierman and Lemcoff [11] suggested the use of the analytical relation between C_B and C_C, which is strictly valid for first-order uncoupled reactions for large Thiele moduli. Thus when h_1 and $h_2 \gg 1$ it will always be assumed that

$$C_C = C_B^w \quad (16)$$

with

$$w = h_2/h_1 (\gamma_C/\gamma_B)^{1/2} \quad (17)$$

The dimensionless form of the kinetic expressions were found using the generalized approach of Roberts and Satterfield [12] which, together with eq. (16), yields

$$r_1^* = \frac{C_A^p C_B^m (1 + K_1 + K_2)^\alpha}{[(1 + K_1 C_B + K_2 C_B^w)]^\alpha} \quad (18)$$

$$r_2^* = \frac{C_A^q C_C^n (1 + K_1 + K_2)^\alpha}{[(1 + K_1 C_C^{1/w} + K_2 C_C)]^\alpha} \quad (19)$$

where

$$K_1 = \left[\frac{K_A C'_{A_S}}{\gamma_B} + K_B C'_{B_S} - K_{P_1} C'_{P_1} \frac{\gamma_{P_1}}{\gamma_B}\right] \Big/ \left\{1 + K_A C'_{A_S}\left[1 - \left(\frac{1}{\gamma_B} + \frac{1}{\gamma_C}\right)\right] + K_{P_1} C'_{P_{1S}}\left(1 + \frac{\gamma_{P_1}}{\gamma_B}\right) + K_{P_2} C'_{P_{2S}}\left(1 + \frac{\gamma_{P_2}}{\gamma_C}\right)\right\} \quad (20)$$

and

$$K_2 = \left[\frac{K_A C'_{A_S}}{\gamma_C} + K_C C'_{C_S} - K_{P_2} C'_{P_{2S}} \frac{\gamma_{P_2}}{\gamma_C}\right] \Big/ \left\{1 + K_A C'_{A_S}\left[1 - \frac{1}{\gamma_B} + \frac{1}{\gamma_C}\right] + K_{P_1} C'_{P_{1S}}\left(1 + \frac{\gamma_{P_1}}{\gamma_B}\right) + K_{P_2} C'_{P_{2S}}\left(1 + \frac{\gamma_{P_2}}{\gamma_C}\right)\right\}. \quad (21)$$

In addition and since the order of the reaction set is arbitrary, it can be chosen in such way that

$$\lambda = \frac{h_2}{h_1} \leq 1. \quad (22)$$

A rigorous solution of the system of differential eqs (5)–(9) is extremely arduous, even with novel numerical techniques. Nevertheless, a general solution can be obtained using the perturbation and matching technique recently developed [13] which is based on the knowledge of the asymptotic behavior of the solutions for small and large values of the Thiele moduli.

Following the procedure sketched for the technique just indicated, when $h_1 \to 0$ it is proposed that

$$C_B = 1 + h_1^2 B_1 + 0(h_1^4) \quad (23)$$

$$C_C = 1 + h_2^2 C_1 + 0(h_2^4) \quad (24)$$

Which after introduction into eqs (18) and (19) and taking into account terms up to the order of h^2 leads to:

$$r_1^* = 1 + h_1^2\left[p\frac{B_1}{\gamma_B} + p\lambda^2\frac{C_1}{\gamma_C} + mB_1 - \alpha(1 + K_1 + K_2)^{-1}(K_1 B_1 + K_2 \lambda^2 C_1)\right] + 0(h_1^4) \quad (25)$$

$$r_2^* = 1 + h_2^2\left[q\frac{C_1}{\gamma_C} + q\frac{B_1}{\gamma_B}\frac{1}{\lambda^2} + nC_1 - \alpha(1 + K_1 + K_2)^{-1}\left(K_1\frac{B_1}{\lambda^2} + K_2 C_1\right)\right] + 0(h_2^4) \quad (26)$$

where B_1 and C_1 are the solution of the following set of differential equations, obtained by replacing eqs (23)–(26) into (5)–(7) and equating terms of like power of h_1 or h_2:

$$\frac{d^2 B_1}{dX^2} = \gamma_B \quad (27)$$

$$\frac{d^2 C_1}{dX^2} = \gamma_C \quad (28)$$

subject to

$$X = 0 \qquad B_1 = C_1 = 0 \quad (29)$$

$$X = 1 \qquad \frac{dB_1}{dX} = \frac{dC_1}{dX} = 0. \quad (30)$$

Since the effectiveness factor η of each reaction is defined as the ratio of the pellet volume averaged reaction rate to the rate under outside surface conditions, then

$$\eta_1 = \int_0^1 r_1^* \, dX = 1 - h_1^2 \sigma_1 + 0(h_1^4) \quad (31)$$

$$\eta_2 = \int_0^1 r_2^* \, dX = 1 - h_2^2 \sigma_2 + 0(h_2^4) \quad (32)$$

with

$$\sigma_1 = \frac{1}{3}[p + p\lambda^2 + m\gamma_B$$
$$- \alpha(1 + K_1 + K_2)^{-1}(K_1\gamma_B + K_2\lambda^2\gamma_C)] \quad (33)$$

$$\sigma_2 = \frac{1}{3}\left[q + \frac{q}{\lambda^2} + n\gamma_C\right.$$
$$\left. - \alpha(1 + K_1 + K_2)^{-1}(K_1\gamma_B/\lambda^2 + K_2\gamma_C)\right]. \quad (34)$$

The asymptotic solutions for h_1 and $h_2 \gg 1$ can be found using the Clareaut substitution in eqs (6) and (7). As the reactions are irreversible and the common reactant A is not limiting; the solution becomes, after taking into account relation (13);

$$\eta_1 = \frac{1}{h_1}\left(\frac{2}{\gamma_B}\right)^{1/2}\left[\int_0^1 \left[1 + \frac{C_B - 1}{\gamma_B} + \frac{C_B^w - 1}{\gamma_C}\right]^p \frac{C_B^m(1 + K_1 + K_2)^\alpha \mathrm{d}C_B}{[1 + K_1C_B + K_2C_B^w]^\alpha}\right]^{1/2} = \frac{\rho_1}{h_1} \quad (35)$$

$$\eta_2 = \frac{1}{h_2}\left(\frac{2}{\gamma_C}\right)^{1/2}\left[\int_0^1 \left[1 + \frac{C_C - 1}{\gamma_B} + \frac{C_C - 1}{\gamma_B}\right]^q \frac{C_C^n(1 + K_1K_2)^\alpha}{[1 + K_2C_C + K_1C_C^{1/w}]^\alpha}\mathrm{d}C_C\right]^{1/2} = \frac{\rho_2}{h_2}. \quad (36)$$

So far, if we look at eq. (22), $h_1 \to \infty$ does not necessarily imply $h_2 \to \infty$. Nevertheless, the opposite situation, $h_2 \to 0$, is not of practical interest due to the fact that both reactions under these unusual conditions are nearly uncoupled.

With the aim of obtaining expressions which permit the estimation of η_1 and η_2 for all range of h_1 of h_2 values, the formula proposed by Gonzo *et al.* [4] was used:

$$\eta_1 = [\hat{h}_1^2 + \exp(-\theta_1\hat{h}_1^2)]^{-1/2} \quad (37)$$

$$\eta_2 = [\hat{h}_2^2 + \exp(-\theta_2\hat{h}_2^2)]^{-1/2} \quad (38)$$

in which

$$\hat{h}_1 = h_1/\rho_1 \quad \text{and} \quad \hat{h}_2 = h_2/\rho_2. \quad (39a, b)$$

The unknowns can be determined by comparing the expansion of eqs (37) and (38), when $\hat{h}_1$ and $\hat{h}_2$ are very small, with eqs (31) and (32). This yields

$$\theta_1 = 1 - 2\sigma_1\rho_1^2 \quad (40)$$

$$\theta_2 = 1 - 2\sigma_2\rho_2^2 \quad (41)$$

On the other hand, when h_1 and $h_2 \to \infty$, eqs (37) and (38) reproduce exactly eqs (35) and (36).

One of the most important features of eqs (37) and (38) is that the unknowns θ_1 and θ_2 are determined by a set of linear algebraic equations in a very simple and explicit form.

RESULTS AND DISCUSSION

Results obtained with expressions (37) and (38) with auxiliary conditions (40) and (41) were compared with numerical findings. The values of η_{1N} and η_{2N} were calculated by the orthogonal collocation procedure of Villadsen and Michelsen [14], although this requires a considerable amount of computing time, especially for values of h_1 greater than one. Figure 1 shows a comparison for three cases where we can observe the influence of α, γ_B, γ_C, K_1 and K_2. When the latter parameters are increased the general order of reaction goes to zero and the most unfavorable situation is encountered. It becomes quite clear that eqs (37) and (38) are able to predict effectiveness factors values in very close agreement with the corresponding values obtained by numerical integration, in the entire range of h_1 values. Nevertheless, as can be seen in Fig. 1 the asymptotic behavior of η_1 and η_2 for high values of h_1 or h_2, as predicted by our analysis, presents a systematic deviation which is due to the

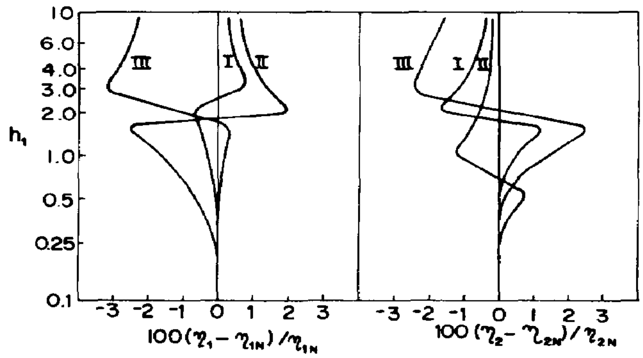

Fig. 1. Deviation between estimated (η) and exact (η_N) values of the effectiveness factors: (I) $p = 1, q = 1, m = 0.8, n = 0.5, \gamma_B = \gamma_C = 2, \lambda = 0.5, K_1 = 2, K_2 = 1, \alpha = 1$; (II) $p = q = m = n = 1, \gamma_B = 5, \gamma_C = 3, \lambda = 0.5, K_1 = 10, K_2 = 5, \alpha = 1$; (III) $p = q = m = n = 1, \gamma_B = 1.5, \gamma_C = 3, \lambda = 0.5, K_1 = 1.5, K_2 = 0.3, \alpha = 2$.

approximation [eq. (16)] used to obtain in the relation between C_B and C_C.

We have also studied the catalytic cohydrogenation of tetraline and paraxylene in a liquid phase with nickel catalyst (Wauquier and Jungers [15]). The reactions are carried out at 443 K with a hydrogen pressure of 60 atmospheres. The concentration of tetraline (B) and paraxylene (C) are 0.2 and 0.1 mole/l. The solubility of hydrogen (A) estimated from Cysewski and Prausnitz [16] is 0.2 mole/l. The system is described by the following rate expressions:

$$r_1 = \frac{k_1k_BC'_AC'_B}{K_BC'_B + k_CC_C} \quad (42)$$

$$r_2 = \frac{k_2k_CC'_AC'_C}{k_BC'_B + k_CC'_C} \quad (43)$$

with $k_1 = 6.7 \times 10^{-3}$ mole/g catalyst min, $k_2 = 12.9 \times 10^{-3}$ mole/g catalyst min and $\frac{k_1k_B}{k_2k_C} = 2.80$.

Under these conditions the dimensionless forms of eqs (42) and (43) become

$$r_1^* = \frac{0.2185C_AC_B}{0.2C_B + 0.01855C_C}. \quad (44)$$

and

$$r_2^* = \frac{0.2185C_AC_C}{0.2C_B + 0.01855C_C} \quad (45)$$

with $\gamma_B = 2.71$, $\gamma_C = 4.85$, $w = 0.564$ and $\lambda = 0.4217$.

To calculate γ_B and γ_C a relation between effective diffusion coefficients inversely proportional to the square root of the correspondent molecular weight has been assumed.

The expression for estimating η_1 and η_2 in accordance with eqs (37) and (38) are

$$\eta_1 = [\hat{h}_1^2 + \exp(-0.5315\hat{h}_1^2]^{-1/2} \quad (47)$$

$$\eta_2 = [\hat{h}_2^2 + \exp(-1.9993\hat{h}_2^2]^{-1/2}. \quad (48)$$

The selectivity toward reaction r_1 can be estimated as

$$S_{1-2} = (\eta_1/\eta_2)/\lambda^2. \quad (49)$$

The results are presented in Table 1. Again the subscript N is used to denote values of η obtained by the collocation method. Once again, it can be seen that the estimation of the effectiveness factors by the approximate expressions (47) and (48) produce fairly accurate results, since the maximum deviation is below 1%. The effect of the internal diffusional

Table 1. Hydrogenation of tetraline and paraxylene

h_1	η_1	η_{1N}	η_2	η_{2N}	S_{1-2}
0.25	0.9724	0.9729	1.0104	1.0106	5.412
0.5	0.8938	0.8991	1.0379	1.0413	4.843
1.0	0.6646	0.6703	1.0868	1.0976	3.439
1.5	0.4780	0.4818	1.0079	1.0179	2.667
2.0	0.3624	0.3591	0.8352	0.8269	2.440
3.0	0.2419	0.2407	0.5696	0.5667	2.388
4.0	0.1814	0.1807	0.4274	0.4257	2.387
6.0	0.1209	0.1205	0.2849	0.2840	2.387

resistance on the selectivity can also be observed in the last columm of Table 1.

The behavior of η_2 in Table 1 deserves some comment. Values of η_2 greater than unity result over a range of values of the Thiele modulus. This is a consequence of the fact that the rate equation · (45) possesses a maximum under certain conditions [see eqs (16), (46) and (45)]. In terms of the Langmuir–Hinshelwood model, this maximum results from a competition between reactants B and C for sites on the catalyst surface. A similar situation was observed for η_2 in case II (Fig. 1). It should be noticed that eq. (48) was able to predict effectiveness factors greater than one.

CONCLUSIONS

By the applications of the perturbation and matching technique and using the new expression for the effectiveness factors estimation it is shown that, for two coupled parallel reactions with Langmuir–Hinshelwood kinetics, this procedure can be used safely in packed-bed catalytic reactor design when the heterogeneous model with intraparticle gradients is considered [1].

The problem of selectivity, which is extremely important in these cases, is immediately determined since the fundamental advantage arise from the fact to have analytical expressions which allows a rapid and accurate calculations of the effectiveness factors.

The effect of competition between reactants species can produce effectiveness factors greater than one, but this situation can also be handled by the matching expression.

Although the analysis was restricted to a porous slab, it can easily be extended for other geometrical shapes of the catalyst.

Acknowledgements—The authors are grateful to Consejo Nacional de Investigaciones Cientificas y técnicas (CONICET) for supporting this work and to Prof. G. Froment who suggested the use of Langmuir–Hinshelwood kinetics expressions to test our procedure.

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NOTATION

B_1	auxiliary expansion function defined by eq. (23)
C	dimensionless concentration
C'	dimensional concentration
C_1	auxiliary expansion function defined by eq. (24)
D	effective diffusivity coefficient
h	Thiele modulus
k_1 and k_2	reaction constants for reactions (1) and (2), respectively
K_1 and K_2	parameters defined by eqs (20) and (21)
K_i for $i = A, B, C, P_1$ and P_2	adsorption equilibrium constants
L	half thickness of the slab particle
m	order of reaction for species B
n	order of reaction for species C
p and q	Order of reaction for species A in reactions (1) and (2), respectively
R	constant defined by eq. (13)
w	parameter defined by eq. (17)
X	dimensionless coordinate
x	dimensional coordinate

Greek letters

α	coefficient defined by eqs (18) and (19)
γ_B and γ_C	parameters defined by eqs (10a)
η_1 and η_2	effectiveness factors for reactions (1) and (2)
λ	Thiele moduli ratio
ν	stoichiometric coefficients
ρ_1 and ρ_2	parameters defined by eqs (35) and (36)
σ_1 and σ_2	parameters defined by eqs (33) and (34)

Subscripts

A, B, C, P_1, P_2	for species A, B, C, P_1 or P_2
S	external surface value

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