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NON-ISOTHERMAL INTERPHASE DIFFUSION WITH CONSECUTIVE REACTIONS

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Abstract—Multi-step heterogeneous consecutive reactions on non-porous catalyst were studied when external mass transfer occuring under non-isothermal condition mainly affects the reaction rates, and useful equations were derived in terms of the dimensionless groups and the measurables by the combination of mass balances with heat balance around the surface of the catalysts.

From the analyses of the equations derived, the enhancements for the surface concentrations of the intermediates were regarded to be identical to those under isothermal condition, and those for the effectiveness factors for the intermediate steps were conculded to be the very multiplications of those through isothermal process by the ratios of the rate constants of the reactions for the corresponding steps at the surface conditions to those at the bulk condition.

INTRODUCTION

As for the effect of diffusion to the reaction rates on heterogeneous catalyst, the references were introduced in the previous study [1] with some examples of the reactions affected by mass transfer.

In the previous study, the kinetics of consecutive reactions were studied and discussed when external mass transfer occuring around nonporous catalyst through isothermal process affects the reaction rates. From that study, the effectiveness factors for the intermediate steps were understood to greatly influenced by the enhancements in simple consecutive reactions, and by the bulk concentrations inclusive of the measurables in additive consecutive reactions.

In non-isothermal process, the explanation of the phenomena around the catalyst particles is not simple because the concentration gradients and a temperature gradient simultaneously affect the reaction rates. In general, the temperature gradient affects the reaction rates more severely than the concentrations do since the temperature gradient directly affects the rate constants. In dealing with the effect of the temperature gradient to the reaction rates, the Arrhenius number and the Prater number are usually introduced besides the Damkholer number introduced in isothermal process. In this study, the equations were derived by introducing these special numbers for the reactions on single particle on the assumption that the concentration gradients are homogeneous within the film around the catalyst regardless of the position of the particle in a reactor and by combining the mass balances with the heat balance for the same reaction schemes as those dealt with in the previous study and also the enhancements were obtained in the same way.

DERIVATION OF EQUATIONS

Simple Consecutive Reactions

Let's consider the following simple multi-step consecutive reactions when external mass transfer occurs through non-isothermal process. The reaction scheme is described as the following

$$A \xrightarrow{k_{1s}} B \xrightarrow{k_{2s}} \cdots \rightarrow i \xrightarrow{k_{js}} \cdots \rightarrow N \xrightarrow{k_{ns}} P \tag{1}$$

If steady state is assumed in the above reactions, the mass balances can be set up in the same way as in isothermal process, but instead the rate constants are given for the surface reactions. A mass balance for the reactant A is written as

$$(k_g a)_A (C_A - C_{AS}) = k_{1S} C_{AS} = R_1 \tag{2}$$

For the intermediates i,

$$(k_g a)_i (C_{is} - C_i) = k_{j-1\,S} C_{(i-1)\,S} - k_{jS} C_{is} = R_{j-1} - R_j \tag{3}$$

For the product P,

$$(k_g a)_P (C_{PS} - C_P) = k_{nS} C_{NS} = R_n \tag{4}$$

If heat produced or removed are due to the reactions, a heat balance can be set up as the following

$$(\mathrm{ha})(T_S - T_b) = \sum_{j=1}^{n} (-\Delta H_j) R_j \tag{5}$$

where the value of (ha) is taken for the mixture of the components in the fluids, and interfacial area for heat transfer is assumed identical to that for mass transfer. The usuage of the subscripts are similar to that in isothermal process while the subcript b means the bulk condition.

In order to derive useful equations, the Damkholer number Da_j, the Arrhenius number ε_j, y and the Prater number β_j were defined as

$$Da_j = \frac{k_{jb}}{(k_g a)_i}, \quad \varepsilon_j = \frac{E_j}{RT_b}, \quad y = \frac{T_S}{T_b}$$

$$\text{and } \bar{\beta}_j = \frac{(j_H/j_D) C_i}{\rho C_P Le^{2/3}} \frac{(-\Delta H_j)}{T_b} \tag{6}$$

(i = A, B, ---, N, and j = 1, 2, ---, n)

where Le is the Lewis number, j_H and j_D are the j-factors for heat transfer and mass transfer respectively, and the density ρ and the heat capacity C_P are given by the values for the mixture. Also $-\Delta H_j$ denotes heat of the reaction for the j-th step. Defined as the ratios of the rate constants of the surface reactions to those of the bulk ones, α_j are expressed by

$$\alpha_j = k_{jS}/k_{jb} = \exp[\varepsilon_j(1 - \frac{1}{y})] \tag{7}$$

If z_i are defined as C_{iS}/C_i, the effectiveness factors $\bar{\eta}_j$ become

$$\eta_j = \alpha_j z_i \tag{8}$$

Thus z_i are expressed in terms of Da_j and $\bar{\eta}_j Da_j$ (= $R_j/(k_g a)_i C_i$) from the mass balances.

$$z_A = \frac{1}{1 + \alpha_1 Da_1} = 1 - \alpha_1 z_A Da_1 = 1 - \bar{\eta}_1 Da_1 \tag{9}$$

$$\begin{aligned} z_i &= \frac{1 - \alpha_{j-1} \nu_i \gamma_i z_{i-1} Da_{j-1}}{1 + \alpha_j Da_j} \\ &= 1 + \alpha_{j-1} \nu_i \gamma_i z_{i-1} Da_{j-1} - \alpha_j Da_j z_i \qquad (10) \\ &= 1 + \nu_i \gamma_i \bar{\eta}_{j-1} Da_{j-1} - \bar{\eta}_j Da_j \end{aligned}$$

(i = B, C, ⋯, N, and j = 2, 3, ⋯, n)

$$\begin{aligned} z_P &= \frac{1}{1 - \alpha_n \nu_P \gamma_P Da_n} = 1 + \alpha_n \nu_P \gamma_P z_N Da_n \\ &= 1 + \nu_P \gamma_P \bar{\eta}_n Da_n \qquad (11) \end{aligned}$$

The effectiveness factors $\bar{\eta}_j$ are written in terms of Da_j and the measurables $\bar{\eta}_j Da_j$ from the eq. (8). That is,

$$\bar{\eta}_1 = \alpha_1/(1 + \alpha_1 Da_1) = \alpha_1(1 - \bar{\eta}_1 Da_1) \tag{12}$$

$$\begin{aligned} \bar{\eta}_j &= \frac{\alpha_j(1 - \alpha_{j-1} \nu_i \gamma_i z_{i-1} Da_{j-1})}{1 + \alpha_j Da_j} \\ &= \alpha_j(1 + \nu_i \gamma_i \bar{\eta}_{j-1} Da_{j-1} - \bar{\eta}_j Da_j) \qquad (13) \end{aligned}$$

where $\nu_i = (k_g a)_{i-1}/(k_g a)_i$ and $\gamma_i = C_{i-1}/C_i$.

Now y is required to be expressed in terms of the measurables since it is the only unmeasurable. It can be obtained from the eq. (5), and the details of the derivation were explained in the appendix. The result is

$$y = 1 + \sum_{j=1}^{n} \bar{\beta}_j \bar{\eta}_j Da_j \tag{14}$$

From this equation, α_j are expressed explicitly in terms of the measurables: that is,

$$\alpha_j = \exp\{\varepsilon_j[1 - (1 + \sum_{j=1}^{n} \bar{\beta}_j \eta_j Da_j)^{-1}\} \tag{15}$$

Thus $\bar{\eta}_j$ become as the following

$$\bar{\eta}_1 = (1 - \bar{\eta}_1 Da_1) \exp\{\varepsilon_1[1 - (1 + \sum_{j=1}^{n} \bar{\beta}_j \bar{\eta}_j Da_j)^{-1}]\} \tag{16}$$

$$\bar{\eta}_j = (1 + \nu_i \gamma_i \bar{\eta}_{j-1} Da_{j-1} - \bar{\eta}_j Da_j) \exp\{\varepsilon_j[1 - (1 + \sum_{j=1}^{n} \bar{\beta}_j \bar{\eta}_j Da_j)^{-1}]\} \tag{17}$$

The point yields Y_{iS} by the surface reactions are obtained in terms of z_i, Da_j and $\bar{\eta}_j Da_j$ respectively: that is,

$$Y_{iS} = k_{j-1} \left(\frac{\alpha_{j-1}}{\alpha_1}\right) \left(\frac{z_{i-1}}{z_A}\right) \prod_{i=B}^{i-1} (1/\gamma_i) - k_j \left(\frac{\alpha_j}{\alpha_1}\right) \left(\frac{z_i}{z_A}\right) \prod_{i=B}^{i} (1/\gamma_i) \tag{18}$$

where $k_j = k_{jb}/k_{1b} \cdot O_T$,

$$Y_{iS} = \left(\frac{\alpha_{j-1}}{\alpha_1}\right) \left(\frac{z_{i-1}}{z_A}\right) \left(\frac{Da_{j-1}}{Da_1}\right) \prod_{i=B}^{i-1} (1/\nu_i \gamma_i) - \left(\frac{\alpha_j}{\alpha_1}\right) \left(\frac{z_i}{z_A}\right) \left(\frac{Da_{j-1}}{Da_1}\right) \prod_{i=B}^{i} (1/\nu_i \gamma_i) \tag{19}$$

In terms of $\bar{\eta}_j Da_j$,

$$Y_{iS} = \frac{1}{\bar{\eta}_1 Da_1} [\bar{\eta}_{(j-1)} Da_{j-1} \prod_{i=B}^{i-1} \frac{1}{\nu_i \gamma_i} - \bar{\eta}_j Da_j \prod_{i=B}^{i} \frac{1}{\nu_i \gamma_i}] \tag{20}$$

Since Y_{ib} by the bulk reaction are written as

$$Y_{ib} = k_{j-1} \prod_{i=B}^{i-1} (1/\gamma_i) - k_j \prod_{i=B}^{i} (1/\gamma_i) \tag{21}$$

the comparison can be made between Y_{iS} and Y_{ib}.

Additive Consecutive Reactions

The reaction scheme is described as the following

$$A \xrightarrow[k_{1S}]{+M} B \xrightarrow[k_{2S}]{+M} \cdots \rightarrow i \xrightarrow[k_{jS}]{+M} \cdots \longrightarrow N \xrightarrow[k_{nS}]{+M} P \tag{22}$$

The mass balances and the heat balance can be set up similarly to those in the previous section, and thus z_M, z_i and $\bar{\eta}_j Da_j$, where $\bar{\eta}_j Da_j$ are written as $R_j/(k_g a)_M C_M$, on new definition of ν_i, γ_i, Da_j and $\bar{\beta}_j$: that is,

$$\nu_i = (k_g a)_M/(k_g a)_i,$$

$$\gamma_i = C_M/C_i, \quad Da_j = k_{jb} C_i/(k_g a)_M \quad \text{and}$$

$$\bar{\beta}_j = \frac{(j_H/j_D) C_M}{\rho C_P Le^{2/3}} \frac{(-\Delta H_j)}{T_b} \tag{23}$$

Then z_M and z_i are expressed as the following

$$z_M = (1+\sum_{j=1}^{n} \alpha_j z_i \mathrm{Da}_j)^{-1} = 1 - \sum_{j=1}^{n} \alpha_j z_M z_i \mathrm{Da}_j$$

$$= 1 - \sum_{j=1}^{n} \bar{\eta}_j \mathrm{Da}_j \tag{24}$$

$$z_A = (1+\alpha_1 \nu_A \gamma_A z_M \mathrm{Da}_1)^{-1} = 1 - \alpha_1 \nu_A \gamma_A z_M z_A \mathrm{Da}_1$$

$$= 1 - \alpha_1 \nu_A \gamma_A \bar{\eta}_1 \mathrm{Da}_1 \tag{25}$$

$$z_i = \frac{1+\alpha_{j-1} \nu_i \gamma_i z_M \mathrm{Da}_{j-1}}{1+\alpha_j \nu_i \gamma_i z_M \mathrm{Da}_j}$$

$$= 1 + \nu_i \gamma_i (\alpha_{j-1} z_{i-1} z_M - \alpha_j z_i z_M \mathrm{Da}_j) \tag{26}$$

$$= 1 + \nu_i \gamma_i (\bar{\eta}_{j-1} \mathrm{Da}_{j-1} - \bar{\eta}_j \mathrm{Da}_j)$$

$$z_P = 1 + \alpha_n \nu_P \gamma_P z_M z_N \mathrm{Da}_n = 1 + \nu_P \gamma_P \bar{\eta}_n \mathrm{Da}_n \tag{27}$$

Also, since $\bar{\eta}_j = \alpha_j z_M z_i$, $\bar{\eta}_j$ are expressed as the following

$$\bar{\eta}_1 = \alpha_1 (1+\sum_{j=1}^{n} \alpha_j z_i \mathrm{Da}_j)^{-1}$$

$$[1+\alpha_1 \nu_A \gamma_A \mathrm{Da}_1 (1+\sum_{j=1}^{n} \alpha_j z_i \mathrm{Da}_j)^{-1}]^{-1}$$

$$= \alpha_1 (1 - \sum_{j=1}^{n} \bar{\eta}_j \mathrm{Da}_j)\ (1 - \nu_A \gamma_A \bar{\eta}_1 \mathrm{Da}_1) \tag{28}$$

$$\bar{\eta}_j = \alpha_j (1+\sum_{j=1}^{n} \alpha_j z_i \mathrm{Da}_j)^{-1} [1+\nu_i \gamma_i (1+\sum_{j=1}^{n} \alpha_j z_i \mathrm{Da}_j)^{-1}$$

$$(\alpha_{j-1} z_{i-1} \mathrm{Da}_{j-1} - \alpha_j z_i \mathrm{Da}_j)]^{-1} \tag{29}$$

$$= \alpha_j (1 - \sum_{j=1}^{n} \bar{\eta}_j \mathrm{Da}_j) [1 + \nu_i \gamma_i (\bar{\eta}_{j-1} \mathrm{Da}_{j-1} - \bar{\eta}_j \mathrm{Da}_j)] \tag{29}$$

(i=B,C, --, N, and j=2, 3, ---, n)

The point yields Y_{iS} are expressed in terms of z_i, Da_j and $\bar{\eta}_j \mathrm{Da}_j$ as the following

$$Y_{iS} = \gamma_A (\alpha_1 z_A)^{-1} (\alpha_{j-1} k_{j-1} z_{i-1}/\gamma_{i-1} - \alpha_j k_j z_i/\gamma_i) \tag{30}$$

$$= (\alpha_1 z_A \mathrm{Da}_1)^{-1} (\alpha_{j-1} z_{i-1} \mathrm{Da}_{j-1} - \alpha_j z_i \mathrm{Da}_j) \tag{31}$$

In terms of $\bar{\eta}_j \mathrm{Da}_j$

$$Y_{iS} = (\bar{\eta}_{j-1} \mathrm{Da}_{j-1} - \bar{\eta}_j \mathrm{Da}_j) / (\bar{\eta}_1 \mathrm{Da}_1) \tag{32}$$

The point yields Y_{ib} by the bulk reactions are

$$Y_{ib} = (k_{j-1} \gamma_{i-1} - k_j \gamma_i) / \gamma_A \tag{33}$$

Thus the comparison can be made between Y_{iS} and Y_{ib}.

RESULTS AND DISCUSSION

Results

The effects of $\bar{\eta}_j \mathrm{Da}_j$, $\bar{\beta}_j$ and γ_i at fixed ε_j to the effectiveness factors in two-step additive consecutive reactions were shown in the figures (1)—(8) by assuming that $\nu_i = 1$ (see the previous study for this assumption). As shown in the graphs, the effects of these are regarded to be considerable. But the graphs for simple consecutive reactions were not drawn because the drawings can be done similarly to additive consecutive reactions.

Surface Concentrations

From the results derived, it is understood that the enhancements for the surface concentrations of the intermediates are regarded to be same as those in isothermal process from the comparisons of the eq. (10) and (26) with the results in isothermal process. That is, the enhancements ϕ_i become

$$\phi_i = \nu_i \gamma_i \bar{\eta}_{j-1} \mathrm{Da}_{j-1} \tag{34}$$

in simple consecutive reactions as well as in additive consecutive reactions. The discussion about the enhancements in isothermal process was given in the previous study.

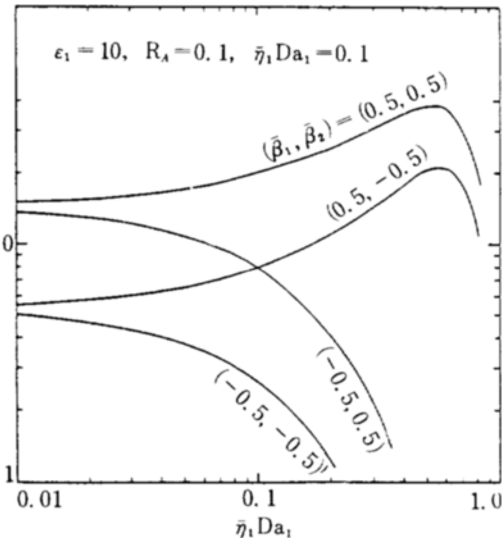

Fig. 1. $\bar{\eta}_1$ vs. $\bar{\eta}_1 \mathrm{Da}_1$ in two-step additive consecutive reactions.

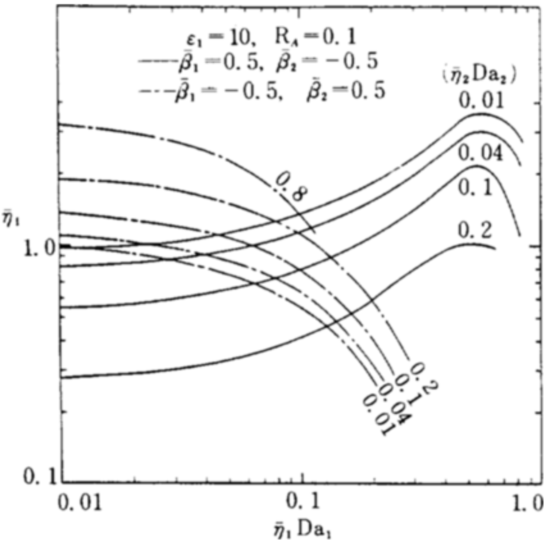

Fig. 2. $\bar{\eta}_1$ vs. $\bar{\eta}_1 \mathrm{Da}_1$ in two-step additive consecutive reactions.

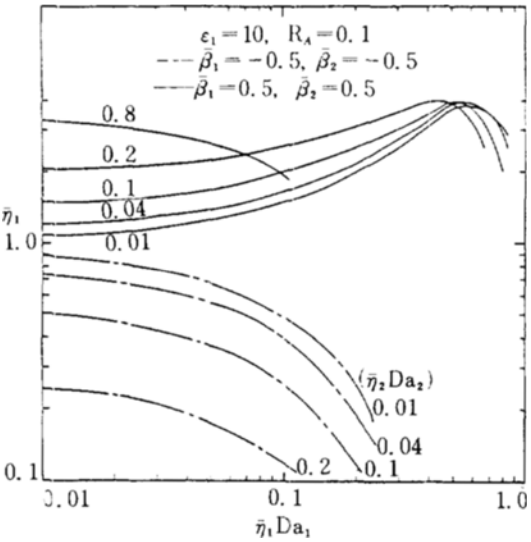

Fig. 3. $\bar\eta_1$ vs. $\bar\eta_1 Da_1$ in two-step additive consecutive reactions.

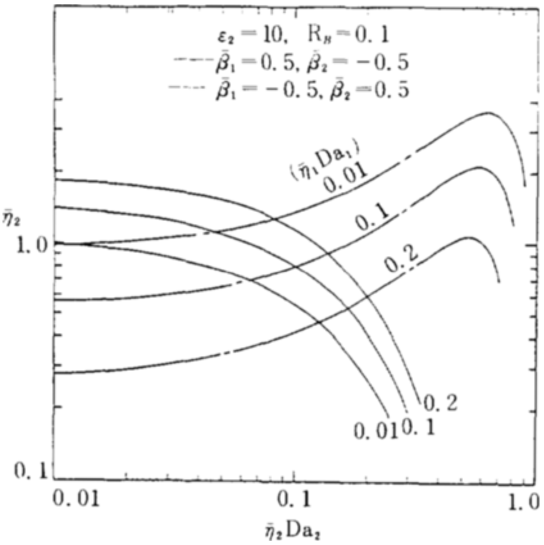

Fig. 5. $\bar\eta_2$ vs. $\bar\eta_2 Da_2$ in two-step additive consecutive reactions.

Effectiveness Factors

Unlike the surface concentrations, the effectiveness factors in nonisothermal process are considerably influenced by the temperature gradients around the catalyst particles. In case of single exothermic reaction, in general $\bar\beta$ enhances $\bar\eta$ because α has the larger value than the unity due to the positive value of $\bar\beta$. But, in case of endothermic reaction, the reverse effect is expected on account of the negative value of $\bar\beta$.

Then, in consecutive reactions, α_j are greatly affected by $\sum_{j=1}^{n} \bar\beta_j \bar\eta_j Da_j$ and therefore this summation is regarded to be important for enhancements of η_j in the consecutive reactions. From the consideration that the above summation is the total of the β multiplied by the measurable $\bar\eta$ Da of every step, the prediction of the enhancements is not simple. From the comparisons of the eq. (12) with the eq. (13) and of the eq. (28) with the eq. (29), the enhancements Φ_j in simple consecutive reactions are

$$\Phi_j = \alpha_j \nu_i \gamma_i \bar\eta_{j-1} Da_{j-1} \tag{35}$$

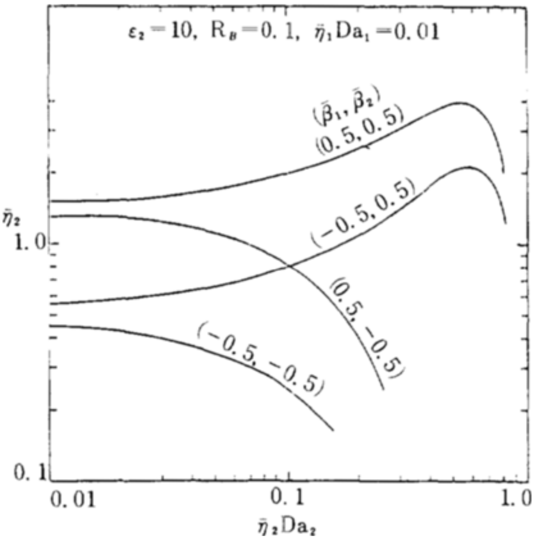

Fig. 4. $\bar\eta_2$ vs. $\bar\eta_2 Da_2$ in two-step additive consecutive reactions.

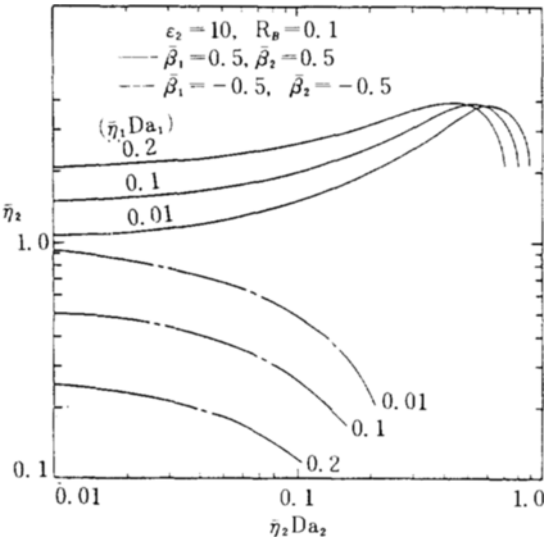

Fig. 6. $\bar\eta_2$ vs. $\bar\eta_2 Da_2$ in two-step additive consecutive reactions.

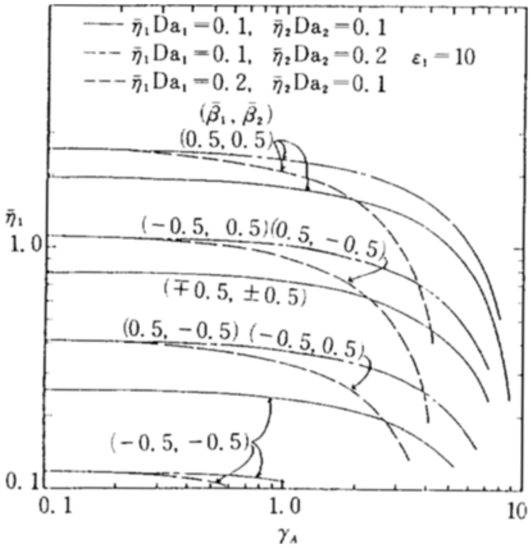

Fig. 7. $\bar{\eta}_1$ **vs.** γ_A **in two-step additive consecutive reactions.**

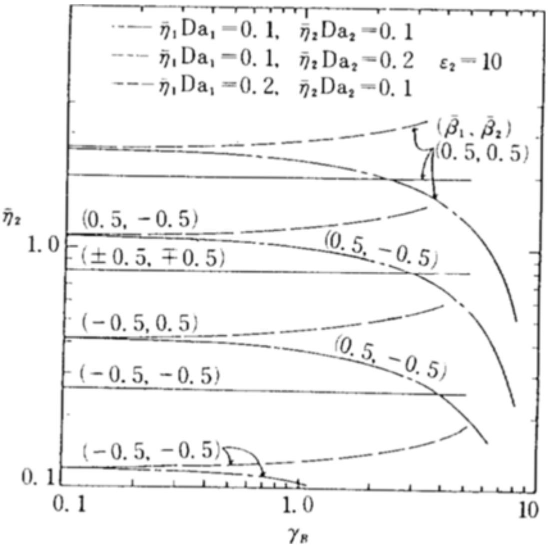

Fig. 8. $\bar{\eta}_2$ **vs.** γ_B **in two-step additive consecutive reactions.**

and those in additive consecutive reactions

$$\Phi_j = \alpha_j \nu_i \gamma_i \bar{\eta}_{j-1} Da_{j-1} \left(1 - \sum_{j=1}^{n} \bar{\eta}_j Da_j\right) \tag{36}$$

what is obvious is that $\bar{\eta}_j$ increase if all the steps are exothermic, but decrease if all the steps are endothermic. In other cases, the enhancements are dependent upon the magnitudes of the $\bar{\eta}_j Da_j$. These effects in two-step additive consecutive reactions were shown in the graphs. Also, from the comparisons with those in isothermal process, it is concluded that the enhancements in non-isothermal process are the multiplications of those in isothermal process by α_j Here α indicates the effect of the temperature gradient. Further, the effectiveness factors are the very multiplications of those under isothermal process by the ratios α_j regardless of the types of the reactions.

From the eq. (35) and (36), the bulk concentrations γ_i affect the enhancements proportionally, but don't the effectiveness factors themselves. While α_j affect proportionally to $\bar{\eta}_j$, γ_i don't do that. In general, the effects of γ_i are regarded to be not so much sensitive as those α_j inclusive of $\bar{\beta}_j$ and ε_j because α_j vary exponentially with ε_j and $1-1/y$ including $\bar{\beta}_j$.

In the previous study, the effects of the diffusivity and the Reynolds number to the mass-transfer resistance around a sphere were discussed. In non-isothermal process, the effect of heat-transfer must be simultaneously considered.

General correlation for mass transfer around a sphere can be written by

$$Sh\left(\propto \frac{k}{D}\right) = 2.0 + f(Re,\ Sc) \tag{37}$$

and that for heat transfer by

$$Nu\left(\propto \frac{h}{k_f}\right) = 2.0 + g(Re,\ Pr) \tag{38}$$

where k_f is thermal conductivity of the fluid. The dividing the eq. (37) by the eq. (38) produces

$$\frac{k_g}{h}\left(\frac{k_f}{D}\right) \propto \frac{2.0 + f(Re,\ Sc)}{2.0 + g(Re,\ Pr)} \tag{39}$$

In case of low Re or motionless fluid, the ratio k_g/h is directly proportional to D/k_f. Therefore, at low Re, the Prater number $\bar{\beta}$ is regarded to be affected mainly by these properties D and k_f.

In case of high Re, the eq. (39) becomes

$$\frac{k_g}{h} \propto \left(\frac{D}{k_f}\right) \cdot F(Le) \tag{40}$$

Therefore, at high Re, $\bar{\beta}$ is affected by Le as well as (D/k_f).

CONCLUSION

Multi-step consecutive reactions with non-isother interphase diffusion were studied. From the analyses of the equations derived, the enhancements for the surface concentrations of the intermediates are identical to those under isothermal condition, and the enhancements for the effectiveness factors for the intermediate step the multiplication of those under isothermal process by the ratio α. Further, the effectiveness factors turned out to be the very multiplications of those under isothermal process by the ratio α like single reaction.

NOMENCLATURE

A : Reactant for initiation
a : Effective interfacial area for heat and mass transfer
C : Concentration
C_P : Heat capacity of the fluids
D : Effective diffusivity
Da : Damkholer number
E : Activation Energy
$-\Delta H$: Heat of reaction
h : Heat-transfer coefficient
j_D : j-factor for mass transfer
j_H : j-factor for heat transfer
kg : Mass-transfer coefficient
k : Rate constant
k_f : Thermal conductivity
k_j : Ratios defined as k_{jb}/k_{1b}
Le : Lewis number
M : Reactant added to each step
N : Intermediate from the (n−1)- th step
n : Number of the total steps
Nu : Nusselt number
P : Final product
Pr : Prandtl number
R : Gas constant or reaction rate
Re : Reynolds number
Sc : Schmidt number
Sh : Sherwood number
T : Temperature
Y : Yield
y : Ratio defined as T_s/T_b
z_i : Ratios defined as C_{is}/C_i

Greek Letters

α_j : Ratios defined as k_{js}/k_{jb}
$\bar{\beta}$: Interphase adiabatic temperature rise: Prater number
γ_i : Ratios defined as C_{i-1}/C_i in simple consecutive reactions and as C_M/C_i in additive consecutive reactions
ε : Arrhenius number difined as E/RT_b
ν_i : Ratios defined as $(k_ga)_{i-1}/(k_ga)_i$ simple consecutive reactions and as $(k_ga)_M/(k_ga)_i$ in additive consecutive reactions
: Interphase effectiveness factor
ρ : Density of the fluids
ϕ : Enhancement of the effectiveness factor
Φ : Enhancement of the surface concentration

Subscripts

b : for the bulk condition
i : for species i(i = A,B,C,..., N,P)
j : for the j-th step
s : for the surface condition

APPENDIX

The eq. (14), the expression as for the ratio T_s/T_b, derived as the following. From the j-factor correlations about heat and mass transfer, the ratio of the heat-transfer coefficient of the fluids to the masstransfer coefficient of a component is written in simple consecutive reactions as

$$\frac{(ha)}{(k_ga)_i} = \rho C_P Le^{2/3} \left(\frac{j_D}{j_H}\right) \tag{a}$$

The division of the both sides of the eq. (5) by $(k_ga)_i C_i T_b$ gives

$$\begin{aligned}\frac{(ha)}{(k_ga)_i C_i}(y-1) &= \frac{R_1(-\Delta H_1)}{(k_ga)_i C_i T_b} + \cdots + \frac{R_j(-\Delta H_j)}{(k_ga)_i C_i T_b} + \cdots \\ &= \frac{-\Delta H_1}{(T_b)} \frac{C_A}{(C_i)} \frac{(k_ga)_A}{(k_ga)_i} \bar{\eta}_1 Da_1 + \cdots \\ &\quad + \frac{-\Delta H_j}{(T_b)} \frac{C_i}{(C_i)} \frac{(k_ga)}{(k_ga)} \bar{\eta}_j Da_j + \cdots\end{aligned} \tag{b}$$

$$\begin{aligned}y-1 &= \frac{(k_ga)_A C_A}{(ha)} \left(\frac{-\Delta H_1}{T_b}\right) \bar{\eta}_1 Da_1 + \cdots \\ &\quad + \frac{(k_ga)_i C_i}{(ha)} \left(\frac{-\Delta H_j}{T_b}\right) \bar{\eta}_j Da_j\end{aligned} \tag{c}$$

The introduction of the definition of $\bar{\beta}_j$ in the eq. (6) and the eq. (a) into the eq. (c) gives

$$\begin{aligned}y &= 1 + \bar{\beta}_1 \bar{\eta}_1 Da_1 + \cdots + \bar{\beta}_j \bar{\eta}_j Da_j + \cdots \\ &= 1 + \sum_{j=1}^{n} \bar{\beta}_j \bar{\eta}_j Da_j\end{aligned} \tag{14}$$

In additive consecutive reactions, the replacement of $(k_ga)_i$ in the relation (a) with $(k_ga)_M$ and the introduction of Da_j and $\bar{\beta}_j$ defined in the eq. (23) produce the same expression as the eq. (14).

REFERENCE

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