

PREDICTION OF THE TEMPERATURE PROFILES OF ADIABATIC MOVING-BED REACTORS

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ABSTRACT

The axial profiles of fluid-and solid-temperatures as well as concentration of reactant are examined based on a simple model for an adiabatic moving-bed catalytic reactor with exothermic reaction. An analytical method is proposed to predict the hot spot temperature and the desirable operating condition. The simultaneous differential equations of this model are calculated numerically for several sets of parameter, and the results are compared with the predicted values by our method. Thus the analytical treatments shown in this paper should prove to be extremely useful for design and operation of the reactor.

The problems in reference to the design and operation of a moving-bed catalytic reactor received some attentions in recent years. Munro and Amundson⁴⁾ discussed heat exchange between solid and fluid, taking into consideration the resistance to heat transfer by conduction in solid spheres, but they did not consider the effect of reactant concentration on the reaction rate. Muchi³⁾ studied a mathematical description of a simple model of the reactor with the first order irreversible reaction, assuming its rate constant is uniform.

The solid catalyst temperature profile is especially important to estimate the hot spot temperature in the reactor. In this paper, analytical and numerical treatments are carried out on a simple model similar to Muchi's one³⁾, in which the first order reaction rate with Arrhenius type rate constant is considered. The 'locus of maxima' method by Barkelew¹⁾ and the graphical method by Liu and Amundson²⁾ for stability analysis of a fixed-bed reactor are applicable to the analytical consideration of the model. A comparison is made between the analytical result and the numerical one in various operating conditions. It is shown that the former can predict not only the effects of operating conditions on the hot spot temperature but also the desirable conditions of the reactor operation.

1. BASIC EQUATIONS

A simple model for an adiabatic counter current operation of a moving-bed catalytic reactor is illustrated in Fig. 1. Assumptions of this model are as follows.

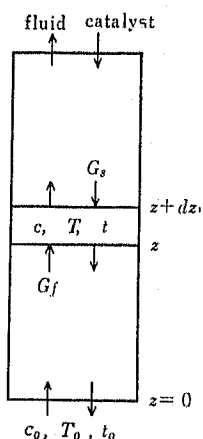


Fig. 1 Model for an adiabatic moving-bed reactor

i) Because it is adiabatic, radial transport of heat and mass by diffusion is not considered.

The axial transport of heat and mass by diffusion is negligible compared with transport by flow.

ii) The velocity profiles of fluid and catalyst particles' flows are both uniform in the cross and the longitudinal sections. Temperature effect on the velocity of fluid flow is neglected.

iii) Resistance to heat transfer is lumped at the particle-fluid interface.

iv) Taking the first order and irreversible catalytic reaction into consideration, the reaction rate constant is expressed by the Arrhenius expression. Differential equations are obtained from these restrictions for steady state operation. The mass balance for the interstitial fluid at the increment dz can be written :

$$-\frac{G_f dc}{\rho_f dz} = (1 - \varepsilon) \rho_s kc \quad (1)$$

The heat balance of fluid phase is :

$$G_f C_f \frac{dT}{dz} = \frac{6(1 - \varepsilon) h_p}{\phi D_p} (t - T) \quad (2)$$

As the reaction occurs in the catalyst particle, the heat balance for the particles at the increment dz is :

$$-G_s C_s \frac{dt}{dz} = (1 - \varepsilon) \rho_s (-\Delta H) kc - \frac{6(1 - \varepsilon) h_p}{\phi D_p} (t - T) \quad (3)$$

Note that;

$$k = k_0 \exp(-E/Rt) \quad (4)$$

where kc is the rate of disappearance of reactant in moles per unit time per unit mass of catalyst. Initial conditions are

$$z = 0; \quad c = c_0, \quad T = T_0, \quad t = t_0 \quad (5)$$

Simultaneous differential equations, Eqs. (1) to (4), are nonlinear, and cannot be solved analytically.

Moreover these equations must be solved essentially as a boundary value problem, but such a calculation is difficult and not practical because a large amount of computation time is required. Therefore, we solved them numerically as an initial value problem under the bottom conditions, Eq. (5). The details of computing method will be shown in the later section.

2. DEVELOPMENT OF EQUATIONS

Eqs. (1) to (5) can be expressed to dimensionless forms as follows.

$$\frac{dX}{d\xi} = -M(1-X) \exp(\alpha/\theta) \quad (6)$$

$$\frac{d\Theta}{d\xi} = \tau\theta - \Theta \quad (7)$$

$$\frac{d\theta}{d\xi} = \frac{Mq}{\beta\tau}(1-X)\exp(\alpha/\theta) + \frac{\tau\theta - \Theta}{\beta\tau} \quad (8)$$

$$\xi = 0; \quad X = 0, \quad \Theta = \theta = 1 \quad (9)$$

ξ , X , Θ and θ are dimensionless variables for reactor length, conversion, fluid temperature and catalyst temperature, respectively, while the others are dimensionless parameters. $d\theta/dX$ is derived from Eqs. (6) and (8) as follows.

$$\frac{d\theta}{dX} = -\frac{q}{\beta\tau} \frac{\tau\theta - \Theta}{\beta\tau M} \frac{\exp(-\alpha/\theta)}{1-X} \quad (10)$$

Total heat balance of the system over any section including the bottom of reactor gives

$$G_f C_f (T_0 - T) + G_s C_s (t - t_0) + (-\Delta H)(G_f / \rho_f)(c_0 - c) = 0$$

The dimensionless form is :

$$\Theta = 1 + \beta\tau(\theta - 1) + qX \quad (11)$$

This may be substituted into Eq. (10) to give

$$\frac{d\theta}{dX} = \frac{q}{\beta\tau} \frac{Q_I - Q_{II}}{Q_{II}} \quad (12)$$

where

$$\left. \begin{aligned} Q_I &= -\frac{\tau}{qM} \frac{1-\beta}{1-X} \theta + \frac{1-\beta\tau+qX}{qM(1-X)} \\ Q_{II} &= \exp(\alpha/\theta) \end{aligned} \right\} \quad (13)$$

Some interest informations on the profiles of catalyst temperature are given by Eq. (12). When the catalyst temperature takes maximum, $d\theta/dX$ is equal to zero. Therefore,

$$Q_{I_m} = -\frac{\tau}{qM} \frac{1-\beta}{1-X_m} \theta_m + \frac{1-\beta\tau+qX_m}{qM(1-X_m)} = \exp(\alpha/\theta_m) = Q_{II_m} \quad (14)$$

The subscript m is used for the indication that the relation is valid only at the point of maximum temperature. The same treatment for stability analysis of a packed-bed reactor has been done by Barkelew¹. Hereafter, as he used, the term 'profile' will refer to a numerical solution of Eq. (12) or Eqs. (6) to (8), and the term 'locus of maxima' or simply 'locus', respectively, to the relation between X_m and θ_m expressed by Eq. (14).

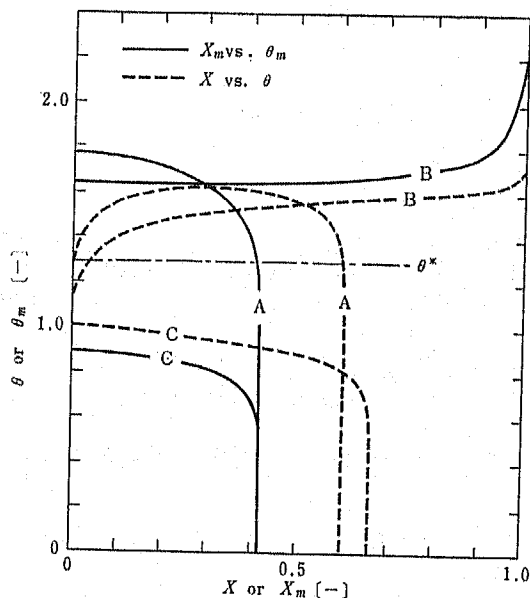


Fig. 2 X_m vs. θ_m and exact solutions

The locus for $\beta=1$ is obtainable directly from Eq. (14), but for $\beta \neq 1$, it can be got as follows. Q_{II_m} gives an exponential curve when plotted as a function of θ_m , while for fixed X_m , Q_{I_m} is a linear function of θ_m and gives a straight line. A series of intersections of Q_{I_m} and Q_{II_m} gives the locus of maxima. Typical loci are shown by solid line in Fig. 2. Examples of numerical solutions of Eq. (12) are shown by dotted line in the same figure for some appropriate parameters. $d\theta/dX$ is always positive to the left of the locus and negative to the right. In the case A, the catalyst temperature θ increases until θ^* without reaction, indicating that this region ($1.0 \sim \theta^*$) is a heat exchange zone, and continues to increase slowly until the maximum, at which the path crosses the locus. After this point, θ decreases and then enters again the heat exchange zone.

In the case B, θ cannot take a maximum because the locus, θ_m , continues to increase until $X=1.0$. In the case C, θ_m at $X_m=0$ is smaller than 1.0. Hence, $d\theta/dX$ must be negative throughout the reaction. θ will decrease continuously without taking a maximum.

Therefore, it appears that if the locus of maxima decreases monotonously and if θ_m at $X_m=0$ is larger than 1.0, θ takes a maximum in the reactor.

Thus one can know without numerical solution whether the catalyst temperature takes a maximum or not.

From the standpoint of commercial operation, it can be said that a convex profile of θ similar to the case A is desirable for operation, because the case A has two heat exchange zones at the both sides of reaction zone. While, such an operation as has an increasing or decreasing profile may not be a desirable one.

Furthermore conversion of effluent fluid should not be below the maximum X_m in the locus.

Estimation of the maximum catalyst temperature

The maximum or, in other words, hot spot temperature of catalyst in the reactor can be obtained from the conception of the locus. When the catalyst temperature takes a maximum, the maximum of θ , namely θ_{max} never exceeds the maximum of the locus. Accordingly one can take this value as the estimation of the maximum catalyst temperature, θ'_{max} . The estimated values were compared with the exact ones for 65 sets of example data. The differences were within 1 to 30% of exact values, and 54 sets of them were presented within 1 to 10%.

Now, it can be shown briefly where the catalyst temperature takes a maximum in the reactor, as follows. The slope of θ at $\xi=0$ is got by putting $\theta = \theta = 1$ and $X = 0$ in Eq. (8). The straight line with this slope through the point $\theta=1$ and $\xi=0$ is illustrated in Fig. 3. Parameters are identical with the

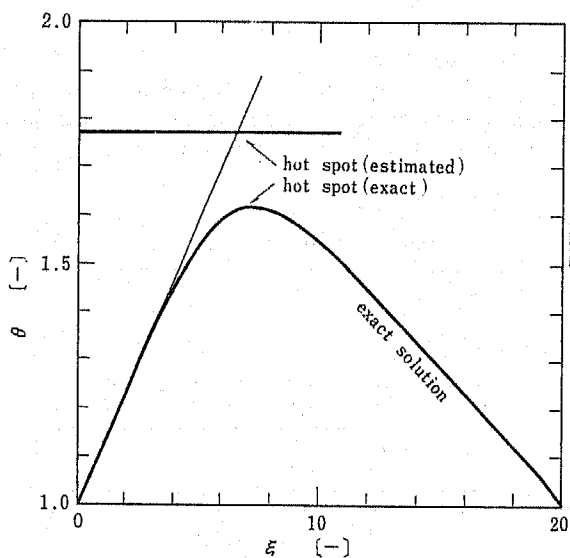


Fig. 3 Estimation of hot spot

case A in Fig. 2. While, the estimated maximum temperature θ'_{max} is the horizontal line. The intersection of the lines will give an expectable position of the maximum temperature. The exact profile by numerical calculation is shown by the solid line in Fig. 3. Though this method is very concise the exact position of θ_{max} does not part so far from the predicted point in most cases. Thus, the value and the position of the hot spot temperature are estimated without numerical calculation, and further the estimated value is always found in so-called the safety side.

Characteristics of the X_m vs. θ_m diagram

Following relations are obtained by an analysis of the sign of dQ_{1m}/dX . For $\beta=1$ and $q+1-\tau>0$, θ_m decreases monotonously and for $\beta=1$ and $q+1-\tau<0$, θ_m increases. For $\beta>1$ and $q+1-\beta\tau>0$, θ_m is always a monotonous decreasing function, and for $\beta<1$ and $q+1-\beta\tau<0$, always increasing function. But, in other cases, namely, when $\beta>1$ and $q+1-\beta\tau<0$, or $\beta<1$ and $q+1-\beta\tau>0$, we can not forecast how θ_m varies with X_m .

3. NUMERICAL CALCULATIONS AND DISCUSSIONS

Simultaneous differential equations, Eqs. (6), (7) and (8), were solved numerically under the condition of Eq. (9), as to various combinations of parameters by a digital computer. The procedure of calculation was Hamming Method, a set of predictor-corrector formulas. For the discussions of effects of parameter on the profile, several sets of illustrative example were chosen. These data are listed in Table 1. The best increment, $d\xi$, was 0.5 for data 1, and 5.0 for data 2 and 3. These values seemed to give satisfactory accuracy without excessive machine time. Smaller increments showed little improvement in profiles.

Axial temperature and concentration profiles

Figs. 4-(a), (b) and (c) show the axial temperature profiles of fluid and catalyst as well as concentration profiles. In Fig. 4-(d), the catalyst temperature is plotted against the conversion X to compare it with the locus of maxima.

Table 1 Illustrative examples

	G_s	G_f	C_s	C_f	ρ_s	ρ_f $\times 10^3$	E $\times 10^{-4}$	h_0 $\times 10^{-6}$	$(-ΔH)$ $\times 10^{-4}$	h_p $\times 10^{-3}$
data 1	0.15	0.168	0.28	0.25	2.6	1.3	1.79	1.45	1.96	6.1
data 2	0.078	0.1	0.28	0.22	1.7	0.87	1.58	1.0	2.2	8.7
data 3	0.078	0.1	0.28	0.22	1.7	0.87	1.58	1.0	2.2	8.7
	ϕD_p	ϵ	c_0 $\times 10^6$	T_0 $\times 10^{-2}$	t_0 $\times 10^{-2}$	α	β	M $\times 10^{-4}$	q	τ
data 1	1.0	0.5	2.0	4.0	4.5	-20.0	1.0	-3.33	0.301	1.125
data 2	1.0	0.55	0.115	3.4	3.5	-22.7	1.0	-0.62	0.039	1.029
data 3	1.0	0.55	0.449	6.5	7.0	-11.4	1.0	-0.62	0.0794	1.077

Units are written in Nomenclature

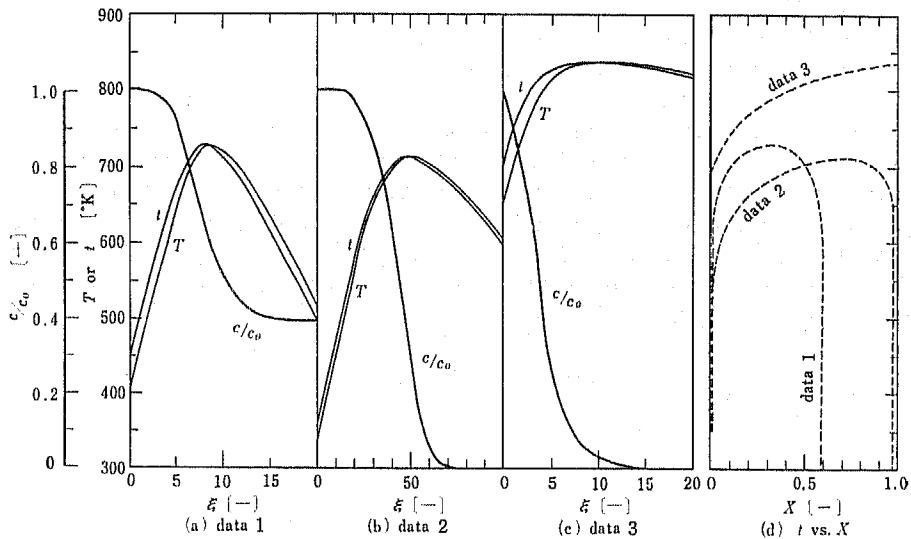


Fig. 4 Typical temperature and concentration profiles

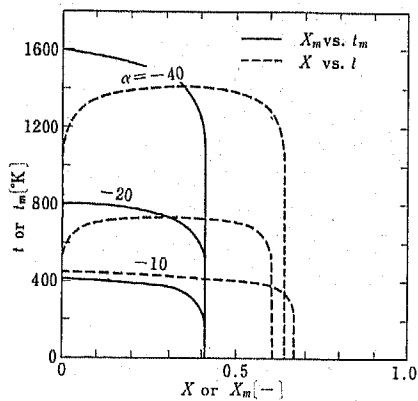


Fig. 5 Effects of changes in α

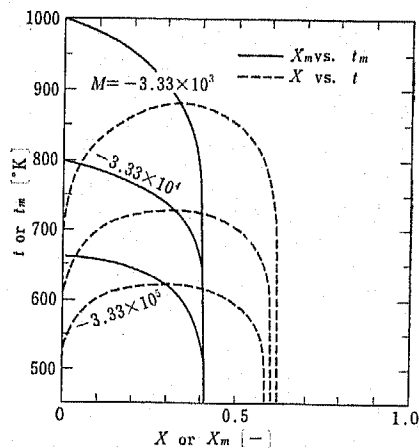


Fig. 6 Effects of changes in M

In the example data 1 and 2 there are two heat exchange zones but not in the data 3, which is the case that the inlet temperature is so high as the reaction has already started at the bottom. The data 1 is chosen to show the effects of parameter on the profile.

Hereafter, θ (or t) vs. X coordinates are adopted to compare with the locus of maxima.

Effects of reaction rate constant

Fig. 5 shows t vs. X family in which α varies and all other parameters are identical with data 1. Fig. 6 shows the change in M . Variations in α and M may be interpreted as variation in the activation energy and the frequency factor term, respectively. It is apparent from these figures that the variation in α or M affects the maximum temperature, but scarcely affects the conversion of effluent fluid. These effects can be presumed by the locus of maxima without numerical results. That is, the estimated maximum temperature changes on account of the variation in α or M , but the maximum of X_m does not change. In the case of $\alpha = -40.0$ in Fig. 5, the value t_m at $X_m = 0$ on the locus is smaller than t_0 , so that, dt/dX is always negative. Therefore, t does not take a maximum in spite of a decreasing locus.

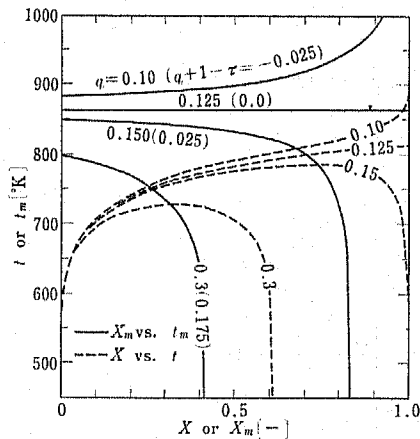


Fig. 7 Effects of changes in q

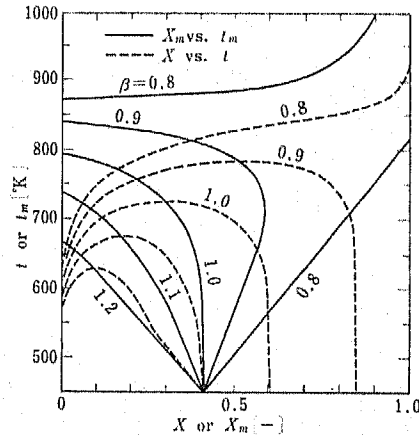


Fig. 8 Effects of changes in β

Effects of concentration

Fig. 7 shows t vs. X family, when changed the value the inlet fluid concentration c_0 , namely the dimensionless term q .†

These examples show, as mentioned above, that if $q+1-\tau > 0$, t_m decreases and t takes a maximum, while if $q+1-\tau < 0$, it does not take a maximum.

Furthermore, if the reactor is sufficiently high, it is seen that any conversion of effluent fluid is not smaller than the maximum of X_m .

† At a glance, these profiles seem to differ from our experience, that is, if c_0 increases, the maximum temperature must rise, too. This irrationality is resulted from the fact that the equations were solved as an initial value problem.

Effects of catalyst flow rate

Fig. 8 shows the effects of changes in the catalyst mass velocity G_s , viz, the dimensionless parameter β . Similarly as above, the informations on the maximum temperature and the conversion can be obtained from the corresponding locus.

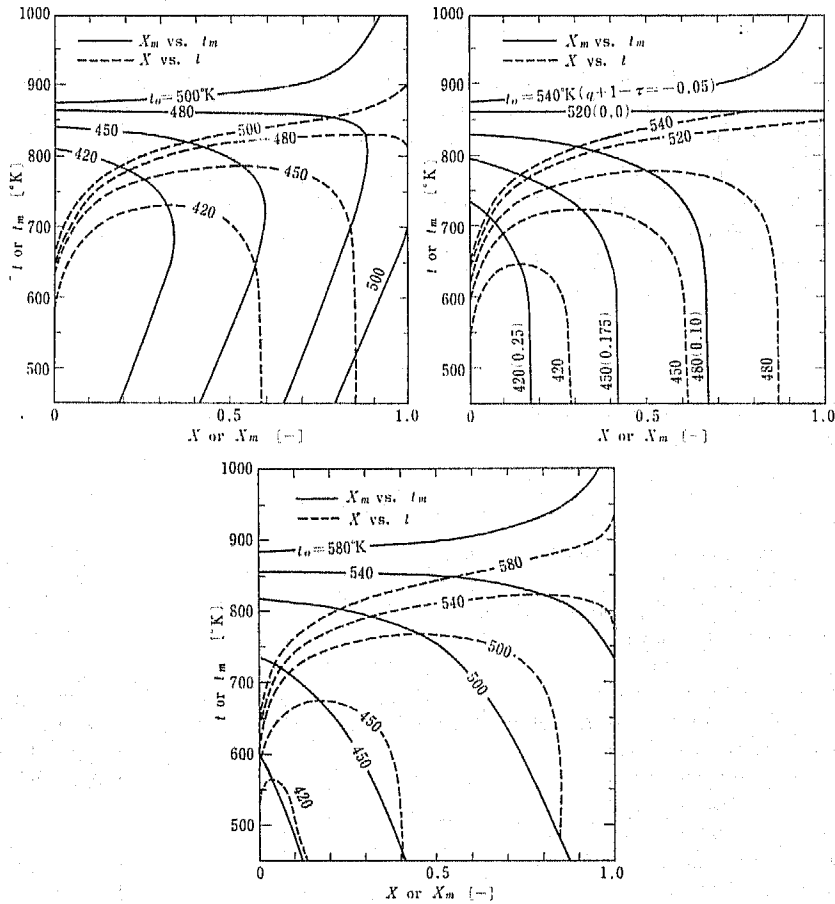


Fig. 9-(a) Effects of changes in t_0 ($\beta=0.9$) Fig. 9-(b) Effects of changes in t_0 ($\beta=1.0$)
 Fig. 9-(c) Effects of changes in t_0 ($\beta=1.1$)

Effects of temperature of catalyst

Figs. 9-(a), (b) and (c) show the effects of changes in the catalyst temperature at the bottom of the reactor, t_0 , for $\beta=0.9$, 1.0 and 1.1, respectively.

A very interesting information for the inlet catalyst temperature can be obtained from these figures. In Fig. 9-(b), for example, if t_0 takes below 520°K, t takes a maximum and never exceeds 864°K that is t_m at $t_0=520$ °K.

It can be said from another standpoint that the inlet catalyst temperature must be controlled below 864°K for the desired operation. But, when it exceeds the limit, the profile does not take a maximum. Similar discussions can also be done for Figs. 9-(a) and (c). For $\beta=0.9$, the limit is 860°K and for $\beta=1.1$, it is 855°K. Thus the upper limit to desirable operating condition is also obtainable from the locus of maxima.

4. CONCLUSIONS

A simple model of a moving-bed catalytic reactor expressed by Eqs. (6) to (8) was treated numerically to determine the axial profiles of concentration and fluid- and catalyst-temperature. The 'locus of maxima' based on Eq.(14) proved a useful method for predicting the behaviors of the reactor, especially the solid catalyst temperature profile, without numerical treatment. Thus, the analytical considerations showed how to find the desirable operating condition and how to estimate the hot spot temperature as well as the effects of the individual parameter on the profile.

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NOMENCLATURE

c	= concentration of reactant in fluid	[gmol/cm ³]
C	= specific heat	[cal/g·°K]
D_p	= diameter of catalyst particle	[cm]
E	= activation energy	[cal/gmol]
G	= mass velocity	[g/cm ² ·sec]
$(-\Delta H)$	= heat of reaction	[cal/gmol]
h_p	= heat transfer coefficient between fluid and solid	[cal/cm ² ·sec·°K]
k	= reaction rate constant	[cm ³ /g-cat·sec]
k_0	= frequency factor	[cm ³ /g-cat·sec]
M	= $-\phi D_p C_f k_0 \rho_f \rho_s / 6 h_p$	[—]
q	= $c_0 (-\Delta H) / \rho_f C_f T_0$	[—]
Q_I, Q_{II}	= abbreviations for Eq. (13)	[—]
R	= gas constant	[cal/gmol·°K]
T	= fluid temperature	[°K]
t	= catalyst particle temperature	[°K]
X	= $1-c/c_0$, conversion	[—]
z	= axial distance from the bottom of bed	[cm]
α	= $-E/Rt_0$	[—]
β	= $G_s C_s / G_f C_f$	[—]
ϵ	= void fraction	[—]
θ	= t/t_0 , dimensionless catalyst temperature	[—]
θ^*	= reaction starting temperature, same form as θ	[—]
θ_{max}	= dimensionless maximum catalyst temperature	[—]
θ'_{max}	= estimated value of θ_{max}	[—]

θ	= T/T_0 , dimensionless fluid temperature	[—]
ξ	= $6(1-\varepsilon)h_p z / \phi D_p G_f C_f$, dimensionless axial distance	[—]
ρ	= density	[g/cm ³]
τ	= t_0/T_0	[—]
ϕ	= shape factor	[—]

Subscripts

0	= value at the bottom
f	= fluid
m	= value at maximum
s	= solid catalyst

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