# Theory and Experiments Concerning the Stability of a Reacting System in a CSTR

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The stability and multiplicity of reacting systems have been investigated long ago (Ostwald, 1899; Liljenroth, 1918). Especially after 1950 an increasing number of papers on these subjects were published. Some papers (van Heerden, 1953; Bilous and Amundson, 1955; Aris and Amundson, 1958; Uppal et al., 1974; Schmitz, 1975) deal with a purely theoretical treatment of the multiplicity and stability of chemically reacting systems. Other papers (Hofmann, 1965; Dammers, 1969; Furusawa et al., 1969; Baccaro et al., 1970; Vejtasa and Schmitz, 1970; Razón and Schmitz, 1987) contain both experimental and theoretical work concerning reacting systems under steady-state conditions. However, a quantitative experimental verification of theoretical results can seldom be found in literature. In our department it has been shown that a quantitative agreement between measured and simulated results can only be obtained if the experiments have been carried out very carefully under well-defined conditions, if all of the required physical quantities have been taken into account—including, for example, the heat production due to stirring and the heat capacity of the vessel and its accessories—and if no approximations like the Frank-Kamenetskii approximation for the Arrhenius equation have been applied (White and Johns, 1987). It has been found that experimentally determined multiple steady states, transient states, limit cycles, and runaway reactions occurring in a CSTR can only be described quantitatively under these conditions (Heemskerk, 1980; Heemskerk et al., 1980; Heemskerk and Fortuin, 1983; Vermeulen et al., 1986; Vleeschhouwer et al., 1988). In the present paper a model describing the acid-catalyzed hydration of oxiranemethanol (OM) into glycerol in a CSTR is presented. This model, which consists of two coupled nonlinear differential equations, including a first-order reaction-rate equation, has been investigated by perturbation analysis (Minorsky, 1962). Different types of steady state predicted by perturbation analysis have been verified experimentally with the above-mentioned liquid reacting system and theoretically by computer simulation. Moreover, quantitative agreement between experimental and simulated results has been obtained.

## Mathematical Model

### Assumptions

The following assumptions will be used to derive a model for the description of the behavior of the reacting system in a CSTR:

- The reaction equation can be represented by

$$C_3H_6O_2\,(l) + H_2O\,(l) = C_3H_8O_3\,(l);$$

$$-\Delta H(298\ \mathrm{K}) = 88{,}200\ \mathrm{J/mol}$$

Where $C_3H_6O_2$ is oxiranemethanol and $C_3H_8O_3$ is glycerol

- The reacting system is a gradientless liquid
- The reaction rate equation is of first order in oxiranemethanol and $H^+$
- The temperature and the composition are time-dependent physical quantities
- The other physical quantities may be considered to be constant in the time domain

### Balances for a CSTR

The following equations will be used to describe the thermodynamic behavior of the reacting system:

Component balance

$$m\, dc/dt = \Phi\,(c_f - c) - r \tag{1}$$

Energy balance

$$(mC_p + m_wC_{pw})\frac{dT}{dt} = \Phi\, C_{pf}(T_f - T) + \Delta Q - US(T - T_c) + r\,(-\Delta H) \tag{2}$$

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Reaction rate equation

$$r = mk_c c = mkc_{\mathrm{H}}c \exp\left(-T_A/T\right) \tag{3}$$

Substitution of Eq. 3 into Eqs. 1 and 2 and rearranging the result leads to:

$$\left(\frac{m}{\Phi}\right)\frac{dc}{dt} = (c_f - c) - k\left(\frac{m}{\Phi}\right)c_{\mathrm{H}}c\exp\left(\frac{-T_A}{T}\right) \tag{4}$$

$$\begin{aligned}\left(\frac{m}{\Phi}\right)\left(\frac{C_p}{C_{pf}} + \frac{m_wC_{pw}}{mC_{pf}}\right)\frac{dT}{dt} &= \frac{\Delta Q}{(\Phi C_{pf})} \\ &+ T_f - T_c + \left[1 + \frac{US}{(\Phi C_{pf})}\right](T_c - T) \\ &+ k\left(\frac{m}{\Phi}\right)c_{\mathrm{H}}\left(\frac{c}{c_f}\right)\left(\frac{-\Delta Hc_f}{C_{pf}}\right)\exp\left(\frac{-T_A}{T}\right)\end{aligned} \tag{5}$$

This model of a reacting system in a CSTR consists of two differential equations containing the following nineteen physical quantities:

$$\begin{gathered} c, c_f, c_{\mathrm{H}}, C_p, C_{pf} \\ C_{pw}, \Delta H, \Delta Q, k, m \\ m_w, S, t, T, T_A \\ T_c, T_f, U, \Phi \end{gathered}$$

where it is assumed that only $c$, $t$, and $T$ are time dependent.

### Introduction of derived physical quantities

Equations 4 and 5 can be reduced by introducing the following derived physical quantities:

$$\begin{aligned} &\tau = \frac{m}{\Phi};\quad \left\{\gamma = \frac{C_p}{C_{pf}} + \frac{m_wC_{pw}}{mC_{pf}}\right\};\quad \tau_h = \frac{mC_{pf}}{US} \\ &T_r = T_c + \left[\frac{\Delta Q}{(\Phi C_{pf})} + T_f - T_c\right]\left(1 + \frac{\tau}{\tau_h}\right)^{-1} \\ &\Delta_aT = \frac{(-\Delta Hc_f)}{C_{pf}} \\ &\Delta_mT = \frac{\Delta_aT}{(1 + \tau/\tau_h)} \end{aligned} \tag{6}$$

Substitution of Eqs. 6 into Eqs. 4 and 5 leads to:

$$\tau\frac{dc}{dt} = c_f - c - k\tau c_{\mathrm{H}}c\exp\left(\frac{-T_A}{T}\right) \tag{7}$$

$$\begin{aligned}\gamma\tau\frac{dT}{dt} &= (1 + \tau/\tau_h)(T_r - T) \\ &+ k\tau c_{\mathrm{H}}(c/c_f)\Delta_aT\exp\left(\frac{-T_A}{T}\right)\end{aligned} \tag{8}$$

In the following section the derived physical quantities will be used in describing the behavior of the reacting system under steady-state conditions.

### Steady-state conditions

In the steady state the following relations hold:

$$\begin{aligned} &\frac{dc}{dt} = 0;\quad \frac{dT}{dt} = 0;\quad c = c_s \\ &T = T_s;\quad r = r_s \end{aligned} \tag{9}$$

As a consequence we obtain from Eqs. 6–9 after elimination of $c_s$:

$$T_s - T_r = \Delta_mT\left[1 + (k\tau c_{\mathrm{H}})^{-1}\exp\left(\frac{T_A}{T_s}\right)\right]^{-1} \tag{10}$$

If no chemical reaction occurs $\Delta_mT = 0$ and $T_s = T_r$; that is, the reference temperature $T_r$ is equal to the temperature $T_s$ of the nonreacting system in the vessel during steady-state operation. From Eq. 6 it is further obtained that during steady-state operation of a reacting system, $T_r = T_c$ if the sum of the temperature $T_f$ of the feed and the temperature rise due to stirring and mixing is equal to the temperature $T_c$ of the coolant. From Eqs. 6 and 10 it can be obtained that under steady-state conditions the maximum temperature rise due to chemical reaction is always smaller than $\Delta_mT$.

Introducing the steady-state temperature rise due to chemical reaction,

$$\Delta_sT = (T_s - T_r) \tag{11}$$

Eqs. 10 and 11 result in:

$$T_r = T_A\left\{\ln\left[k\tau c_{\mathrm{H}}\left(\frac{\Delta_mT}{\Delta_sT} - 1\right)\right]\right\}^{-1} - \Delta_sT \tag{12}$$

### Perturbation analysis

If in the reacting system infinitesimally small perturbations $\delta c$, $\delta T$, and $\delta r$ from steady-state conditions occur, the following relations hold:

$$c = c_s + \delta c,\quad r = r_s + \delta r,\quad \text{and}\quad T = T_s + \delta T \tag{13}$$

From Eqs. 7, 8, 9, 13, and the first-order approximation of the Maclaurin series:

$$\delta r = \left(\frac{\partial r}{\partial c}\right)_T\delta c + \left(\frac{\partial r}{\partial T}\right)_c\delta T \tag{14}$$

it can be obtained that

$$\frac{d^2(\delta T)}{dt^2} - 2A\frac{d(\delta T)}{dt} + B^2(\delta T) = 0 \tag{15}$$

The characteristic equation of differential Eq. 15 is:

$$\lambda^2 - 2A\lambda + B^2 = 0 \tag{16}$$

Equation 15 has been derived in a previous paper (Heemskerk and Fortuin, 1983). It can be obtained from Eqs. 3, 7, 8, 13, 14, and 15 that:

$$2A = \left(\frac{\Delta_s T T_A}{T_s^2} - 1\right)\left(1 + \frac{\tau}{\tau_h}\right)(\tau\gamma)^{-1} - \left[\tau\left(1 - \frac{\Delta_s T}{\Delta_m T}\right)\right]^{-1} \tag{17}$$

$$B^2 = \left[\left(1 - \frac{\Delta_s T}{\Delta_m T}\right)^{-1} - \Delta_s T \frac{T_A}{T_s^2}\right] \frac{(1 + \tau/\tau_h)}{(\gamma\tau^2)} \tag{18}$$

Further, the discriminant of Eq. 16 follows from:

$$D = 4(A^2 - B^2) \tag{19}$$

The type of steady state depends on the combination of the signs of the values of $2A$, $B^2$, and $D$; each set of three signs indicates a separate steady-state type, as discussed next.

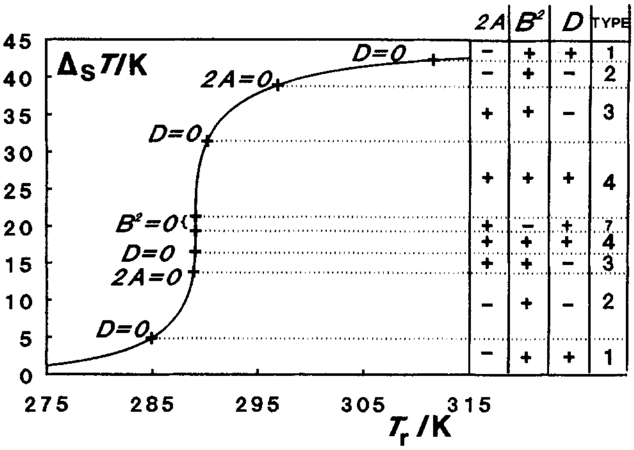

**Figure 1. Types of steady state with various $T_r$ values.**

Curve and indicated points calculated with Eqs. 12, 17, 18, 19, using data of Table 1

## Analysis of Steady States

### *Steady states with various $T_r$ values*

In order to demonstrate the behavior of a chemically reacting system in a CSTR under different steady-state operating conditions, we have chosen the suitable and realistic values of the physical quantities represented in Table 1. After substitution of these suitable data into Eq. 12 the S-shaped curve of Figure 1 is obtained. Each point of this S-shaped curve represents a steady state of the chemically reacting system in the CSTR. It gives the temperature rise $\Delta_s T$ of the system due to chemical reaction as a function of the reference temperature $T_r$, that is, the temperature the system would have if no chemical reaction occurred.

The perturbation analysis carried out earlier and applied to steady states of the reacting system resulted in Eq. 15. In this equation the two different signs of the values of the three parameters $2A$, $B^2$, and $D$ result in $2^3$ combinations of signs and consequently in eight different types of steady state, Table 2. In order to be able to characterize the different types of steady state represented by the locus of steady states in Figure 1, the S-shaped curve has been divided into nine parts by eight characteristic points:

Two points where $2A = 0$
Two points where $B^2 = 0$
Four points where $D = 0$

It must be noted that substitution of less suitable values into Eq. 12 will generally result in an S-shaped curve with a smaller number of characteristic points. Each of the nine parts of the curve in Figure 1 refers to a steady-state type of which the values of $2A$, $B^2$, and $D$ have a fixed combination of signs, Table 2. In Figure 1 five different types of steady state occur. The combinations of types 1 to 4 of Table 2 fulfill the slope condition $B^2 > 0$; they refer to stable steady states. For combinations 1 and 2 the dynamic condition $2A < 0$ holds. This means that a perturbation of the temperature will decrease in time. This decrease will be an asymptotic damping if $D > 0$ (type 1) and an approach to a spiral point if $D < 0$ (type 2). In Figure 1, type 3 covers stable oscillations and type 4 asymptotic oscillations provided that no limiting value (e.g., the bubble point of the reacting system) is exceeded. The combinations where $B^2 < 0$ belong to the unstable steady states (types 5 to 8).

**Table 1. Data Used in Figure 1**

| Physical Quantity | Value |
|---|---|
| $c_H$ | 0.15 mol · kg$^{-1}$ |
| $k$ | $8.5 \times 10^{10}$ kg · mol$^{-1}$ · s$^{-1}$ |
| $T_A$ | 8,827 K |
| $\gamma$ | 1 |
| $\Delta_a T$ | 319 K |
| $\tau$ | 167 s |
| $\tau_h$ | 26.5 s |

### *Steady states with various $T_r$ and $\tau_h$ values*

In Figure 2 twenty S-shaped curves have been drawn. These curves have been calculated with Eq. 12 using the values of Table 3 except the $\tau_h$ value. The $\tau_h$ values substituted into Eq. 12 to calculate the curves of Figure 2 have been obtained from:

$$\tau_h = \tau_{h1} U_1 S/(US)$$

where

$$\tau_{h1} U_1 S = 750 \text{ J/K} \tag{20}$$

The values of $US$ substituted in Eqs. 12 and 20 are 3, 6, 9, . . . , 60 W/K. On each of the S-shaped curves of Figure 2 characteristic points have been indicated where $2A = 0$, $B^2 = 0$, $D = 0$. The coordinates of these points have been calculated using Eqs. 12, 17, 18, and 19. In Figure 2 similar characteristic points have been connected by characteristic curves indicated by $2A = 0$,

**Table 2. Types of Steady State**

| Type | Sign 2A | Sign $B^2$ | Sign D | Behavior After a Perturbation |
|---|---|---|---|---|
| 1 | − | + | + | Asymptotic damping |
| 2 | − | + | − | Spiral point |
| 3 | + | + | − | Stable oscillations |
| 4 | + | + | + | Asymptotic oscillations |
| 5 | − | − | + | Unstable steady state |
| 6 | − | − | − | Unstable steady state |
| 7 | + | − | + | Unstable steady state |
| 8 | + | − | − | Unstable steady state |

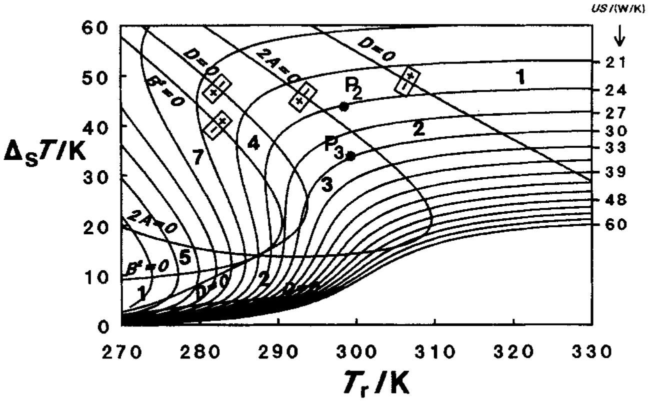

**Figure 2. Steady states with various $T_r$ and $\tau_h$ values.**

Curves and other solid lines calculated with Eqs. 12, 17, 18, 19, using data of Table 3

*US* value range is 3–60 W/K, in increments of 3 W/K

1–5, 7 refer to steady-state types, Table 2

$B^2 = 0$, and $D = 0$. These characteristic curves are the boundaries of the areas covering steady states of the same type. In Figure 2 six different regions of steady states have been indicated. Each region has its own set of signs of values of $2A$, $B^2$, and $D$, Table 2. In each of the areas, points can be chosen representing steady states of the same type. The steady states represented by points $P_2$ and $P_3$ in Figure 2 are discussed in more detail in the next section.

## Behavior of a Reacting System under Various Steady-state Conditions

According to Table 2, eight different types of steady state can be distinguished for the acid-catalyzed hydration of oxiranemethanol (OM) in a CSTR. In order to verify this behavior we give results obtained by experiments and by numerical simulation.

### *Experimental behavior*

The hydration of OM was carried out in a 250 mL vessel provided with internals as described in a previous publication (Vermeulen, 1986). Aqueous solutions of oxiranemethanol and sulfuric acid were used as feed streams of constant flow rate into the stirred and cooled reactor vessel. The temperature was digitally recorded at intervals of 5 s, as represented in Figure 3. During the first steady-state period stable temperature oscillations were obtained after about 2 h of operation at a stirrer speed of $n = 75\ s^{-1}$. After 7,775 s the stirrer speed was reduced from 75 to 50 $s^{-1}$. This reduction of stirrer speed at the start of the second steady-state period resulted in a lower $U$ value and consequently in a higher $\tau_h$ value than in the first period. From Figure 3 it can be concluded that under the experimental conditions considered, two types of steady state have been obtained. During the first steady-state period, represented by $P_3$ in Figure 2, a stirrer speed of $n = 75\ s^{-1}$ ($\tau_h = 24.74$ s) exists and stable temperature oscillations occur. During the second steady-state period, represented by $P_2$ in Figure 2, where $n = 50\ s^{-1}$ ($\tau_h = 30.30$ s), the temperature oscillations are damped (type 2 in Table 2). After 9,640 s the stirrer speed was increased to its original value, resulting in an approach to the third steady-state period, $P_3$ in Figure 3, where temperature oscillations (type 3 in Table 2) similar to those in the first period occur.

**Table 3. Data Used in Figure 2**

| Physical Quantity | Stirrer<br>$n = 75\ s^{-1}$<br>$P_3$; Type 3 | <br>$n = 50\ s^{-1}$<br>$P_2$; Type 2 | Dimension |
|---|---|---|---|
| $2A$ | $+4.1 \times 10^{-3}$ | $-3.0 \times 10^{-2}$ | $s^{-1}$ |
| $B^2$ | $+1.1 \times 10^{-3}$ | $+1.9 \times 10^{-3}$ | $s^{-2}$ |
| $D$ | $-4.4 \times 10^{-3}$ | $-6.7 \times 10^{-3}$ | $s^{-2}$ |
| $c_f$ | 8.59 | 8.59 | $mol \cdot kg^{-1}$ |
| $c_H$ | 0.16 | 0.16 | $mol \cdot kg^{-1}$ |
| $k$ | $8.5 \times 10^{10}$ | $8.5 \times 10^{10}$ | $kg \cdot mol^{-1} \cdot s^{-1}$ |
| $T_A$ | 8,827 | 8,827 | K |
| $T_r$ | 299.3 | 298.5 | K |
| $\Delta_a T$ | 303.1 | 303.1 | K |
| $\Delta_s T$ | 34.7 | 43.5 | K |
| $\gamma$ | 1.5* | 1.5* | 1 |
| $\tau$ | 166.67 | 166.67 | s |
| $\tau_h$ | 24.75 | 30.3 | s |

*Estimated mean value

### *Simulated behavior*

In Table 3 the values of $2A$, $B^2$, and $D$ calculated with Eqs. 12, 17, 18, and 19 have been represented for steady-state conditions indicated by points $P_2$ and $P_3$ of Figure 2. From the combination of signs of the three characteristic values represented in Table 3 it appears that during the first and the third period ($n = 75\ s^{-1}$) steady states of type 3 (stable oscillations) can be expected, and during the second period ($n = 50\ s^{-1}$) a steady state of type 2 (spiral point) will occur. Under the operating conditions represented by points $P_2$ and $P_3$ in Figure 2, the temperature–time relationship has been simulated numerically with a computer using Eqs. 7 and 8 and the values of the physical quantities presented in Table 3. The results of this numerical simulation are represented in Figure 4. For this simulation $U$ is calculated for each stirrer speed, as published previously, and $C_p$ is obtained by taking 80% of the sum of the partial heat capacities (Vermeulen, 1986; Vleeschhouwer et al., 1988).

### *Discussion*

It is interesting to note that in both the experimental results, Figure 3, and the simulated results, Figure 4, there is a small

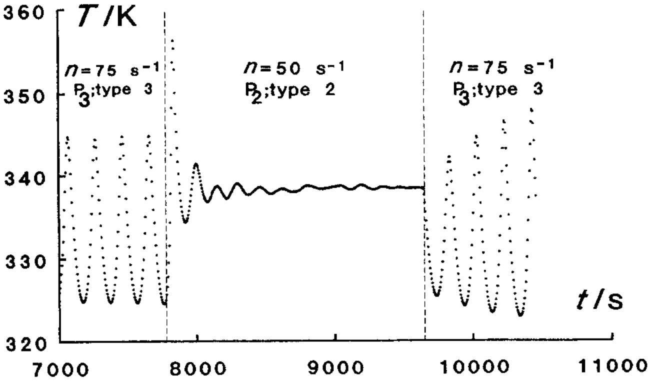

**Figure 3. Experimentally obtained temperature oscillations under steady-state conditions in Table 3.**

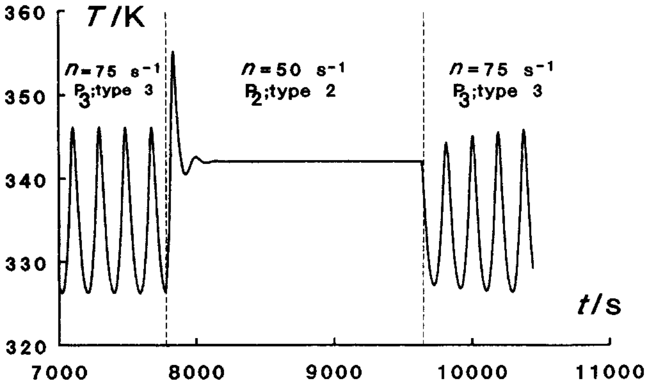

**Figure 4. Simulated temperature oscillations for steady-state conditions in Table 3.**

difference between the oscillations during the first and the third periods under similar operating conditions. The amplitude of the oscillations referring to the first period is constant in time, while the amplitude of the third period shows a small increase. This difference is due to the fact that only the end of the first period and the start of the third period have been represented in these figures. In the first period the limit cycle is sufficiently approached after 2 h operation, resulting in oscillations of constant amplitude. At the third period, even after 15 min operation the transient state is still approaching steady-state oscillations, resulting in a small increase of the amplitude. Consequently we may state that the experimental results obtained agree quantitatively with the behavior of the reacting system predicted by the model.

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## Notation

$2A$ = dynamic condition parameter, Eq. 17, $s^{-1}$
$B^2$ = slope condition parameter, Eq. 18, $s^{-2}$
$c$ = concentration of key component, oxiranemethanol, mol · $kg^{-1}$
$c_f$ = concentration of oxiranemethanol in feed, mol · $kg^{-1}$
$c_H$ = $H^+$ concentration in reaction vessel, mol · $kg^{-1}$
$C_p$ = average heat capacity of reacting liquid, J · $kg^{-1}$ · $K^{-1}$
$C_{pf}$ = heat capacity of feed, J · $kg^{-1}$ · $K^{-1}$
$C_{pW}$ = heat capacity of solid parts of reactor, J · $kg^{-1}$ · $K^{-1}$
$D$ = asymptotic condition parameter, Eq. 19, $s^{-2}$
$\Delta H$ = reaction enthalpy, J · $mol^{-1}$
$k_c$ = first-order reaction rate constant, $s^{-1}$
$k$ = second-order frequency factor, kg · $mol^{-1}$ · $s^{-1}$
$m$ = mass of reaction liquid in reactor, kg
$m_W$ = mass of solid parts of reactor, kg
$n$ = stirrer speed, $s^{-1}$
$\Delta Q$ = heat production due to mixing and stirring, W
$r$ = reaction rate, mol · $s^{-1}$
$S$ = heat transfer surface area of cooling coil, $m^2$
$t$ = time, s
$T$ = temperature of reacting system in reactor, K
$T_A$ = activation temperature, K
$T_c$ = inlet temperature of coolant, K
$T_f$ = temperature of feed, K
$T_r$ = reference temperature, Eq. 6, K
$T_s$ = temperature of reacting system under steady-state conditions, K
$\Delta_a T$ = adiabatic temperature rise, Eq. 6, K
$\Delta_m T$ = maximum temperature rise, Eq. 6, K
$\Delta_s T$ = steady-state temperature rise, Eq. 11, K
$U$ = overall heat transfer coefficient of cooling coil, W · $m^{-2}$ · $K^{-1}$

### *Greek letters*

$\gamma$ = parameter, Eq. 6, 1
$\lambda$ = parameter, Eq. 16, $s^{-1}$
$\tau$ = residence time, $\tau = m/\Phi$, s
$\tau_h$ = heat release time, $\tau_h = mC_{pf}/(US)$, s
$\Phi$ = mass flow rate, kg · $s^{-1}$

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