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Transient Runaway in a Fixed-Bed Catalytic Reactor

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1. Introduction

Fixed-bed tubular reactors have important commercial applications in the chemical and allied process industries. A typical reactor consists of a cylindrical tube filled with solid catalyst particles and is mounted in a vertical upright position. After the reacting fluid has entered the reactor, it moves along the packed bed and reacts on the catalyst particles to produce the desired products. While fixed-bed reactors, relatively to other types of catalytic reactors, are flexible, efficient, low-cost, and require low maintenance, their most serious disadvantage is poor heat transfer with attendant poor temperature control (e.g., see Bartholemew and Farrauto, 2006).

When a highly exothermic reaction is carried out in a packed bed reactor some problems associated with the operation of the reactor may arise. In this case, if the reaction generates heat faster than it can escape the reactor, viz., through reactor walls, the temperature of the fixed bed increases. The reaction and heat generation rates then increase, which causes a further rise in the reactor temperature. This can result in thermal runaway, with the temperature increasing ever more rapidly until the reactants have been finally exhausted. Runaway, or parametric sensitivity, describes a situation in which a small change in an operating variable such as feed temperature, concentration, or flow rate induces a large change in the temperature profile of the reactor. Further, a number of runaways occur due to scale-up, inadequate procedures and training, raw material quality control, maintenance, etc (Etchells, 1997; Barton and Rogers, 1997). Runaway can promote undesired side reactions, catalyst deactivation, productivity loss, and deterioration of product selectivity. Because of the risk associated with thermal runaway, many industrial reactors are operated under overly suboptimal conditions. It should be noted that near-runaway operation is potentially advantageous to catalytic processes as it may result in optimum reactor operation, especially if energy savings can be attained. For these reasons, understanding thermal runaway is of significant industrial importance.

The concept of thermal runaway was first introduced to chemical reactor analysis in late 1950s (Bilous and Amundson, 1956). There have been many studies in the literature on the derivation of runaway criteria, e.g., in tubular and fixed-bed reactors with a single reaction (van Welsenaere and Froment 1970; McGreavy and Adderley, 1973; Morbidelli and Varma 1982, 1988; Bashir et al. 1992), in reactors with multiple reactions (Hosten and Froment, 1986; Henning and Perez, 1986; Morbidelli and Varma, 1989; van Woezik and Westerterp, 2000,

2002), with the effect of coolant temperature (Henning and Perez, 1986; Hosten and Froment, 1986; Hagan et al. 1988; Bauman et al., 1990). All of this research has focused on developing steady-state criteria for thermal runaway. Specifically, these studies have identified regions in which small changes in the reactor operating conditions lead to large steady-state changes in the reactor temperature. Because this research has been performed on steady-state systems, the results indicate only process conditions under which the system is prone to thermal runaway. Once the system parameters reach values indicating runaway, however, the parametric sensitivity results provide no indication of when thermal runaway occurs. Recently only, correlations for time to runaway for a continuous stirred tank reactor have been developed (Slaback and Riggs, 2004). In order to derive the correlations analytically, a first-order, exothermic reaction has been considered and the reactant has been assumed to be in excess to eliminate the material balance equation.

The focus of this chapter is on the investigation of the thermal behavior of a two-dimensional fixed-bed tubular reactor, and the development of a dynamic criterion indicating the onset of thermal runaway. The ability to predict the tendency towards thermal runaway over time is of valuable importance, and can help in correcting operating conditions in order to suppress runaway before it has occurred. This chapter is organized as follows: a detailed description of the reactor model is given in section 2. In section 3, the model is solved using brute force and simulation results for two inlet temperatures are discussed. In the fourth section, runaway conditions are qualitatively elucidated and the transient criterion is developed.

2. Reactor Model

A dynamic model is used in this study to develop a criterion pointing to the onset of runaway in a two-dimensional tubular (of radius R_t) catalytic reactor (see Figure 1). The catalyst particles have a diameter d_p . The reaction system considered in this study consists of a single, irreversible, exothermic reaction. As an example, carbon monoxide conversion over copper chromite ($\text{CO} + \frac{1}{2} \text{O}_2 \rightarrow \text{CO}_2$, $\Delta H_r = -23.8 \text{ kJ/mol}$) is considered, the reaction rate of which is given by (Dekker et al., 1992; Mardanova et al., 1996; Koning and Westerterp, 1999).

$$\Re(C_s, T_s) = \frac{k_r b_{\text{CO}} p_{\text{CO}}}{1 + b_{\text{CO}} p_{\text{CO}} + b_{\text{O}_2} p_{\text{O}_2}} \quad (1)$$

where p_i ($i = \text{CO}, \text{O}_2$) is the partial pressure of i , and

$$k_r = k_r^o e^{-E_a / R_g T}, \quad b_i = b_i^o e^{\Delta H_i^{\text{ads}} / R_g T} \quad (2)$$

The dimensionless reaction rate is defined by

$$R(C_s, T_s) = \frac{\Re(C_s, T_s)}{\Re(C_s, T_s)|_{in}} \quad (3)$$

where the denominator term is the reaction rate at inlet conditions.

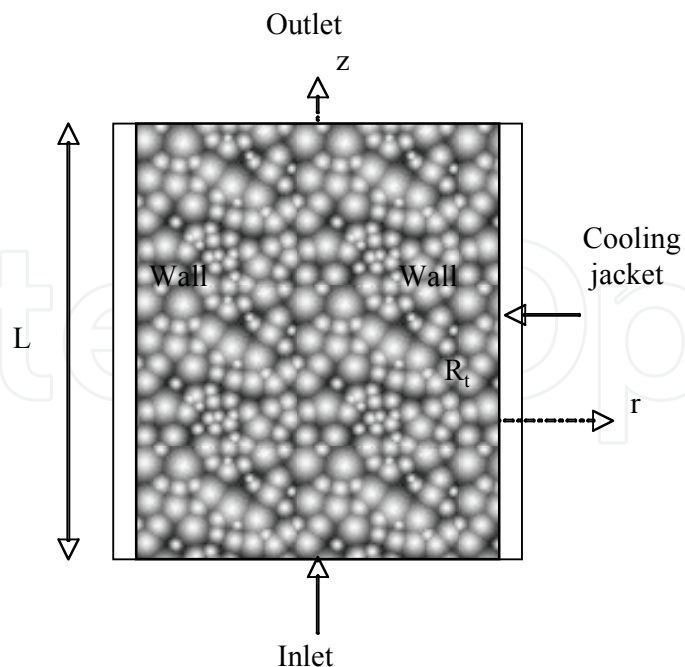


Figure 1. Schematics of the fixed bed

The porosity profile, $\psi(r)$, over a cross-section of the bed is given by (Vortmeyer and Schuster, 1983)

$$\psi(r) = \psi_{\infty} [1 + C \exp(-N[1-r])] \quad (4)$$

where $N = 2R_t/d_p$. The parameter $C = 0.5$, corresponding to a value of the porosity at the wall of 0.6 (Giese et al, 1998) and a value of the porosity at the core of the bed, ψ_{∞} , of 0.4. The radial, superficial velocity distribution has been obtained by the same authors via the solution of a modified Birkman equation (Birkman, 1947) with the porosity profile given above (equation 4). The following equation has been proposed

$$\frac{u(r)}{u_0} = K [1 - (1 - A[1-r]) \exp(B[1-r])] \quad (5)$$

where expressions for K , A , and B are given elsewhere (see Vortmeyer and Schuster, 1983; Borkink and Westerterp, 1994). The porosity and velocity distributions are shown in Figure 2, where the wall effect can be clearly discerned.

The transient material and energy balance equations are expressed using a pseudo-heterogeneous formulation as follows.

Material balance:

Fluid phase:

$$\psi a \frac{\partial C_f}{\partial t} = \frac{1}{Pe_{m,a}} \frac{\partial^2 C_f}{\partial z^2} + \frac{1}{Pe_{m,r}} \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial C_f}{\partial r} \right) - \frac{\partial C_f}{\partial z} + St_m (C_s - C_f) \quad (6)$$

Solid Phase:

$$(1 - \psi) a \frac{dC_s}{dt} = - (1 - \psi) Da R(C_s, T_s) - St_m (C_s - C_f) \quad (7)$$

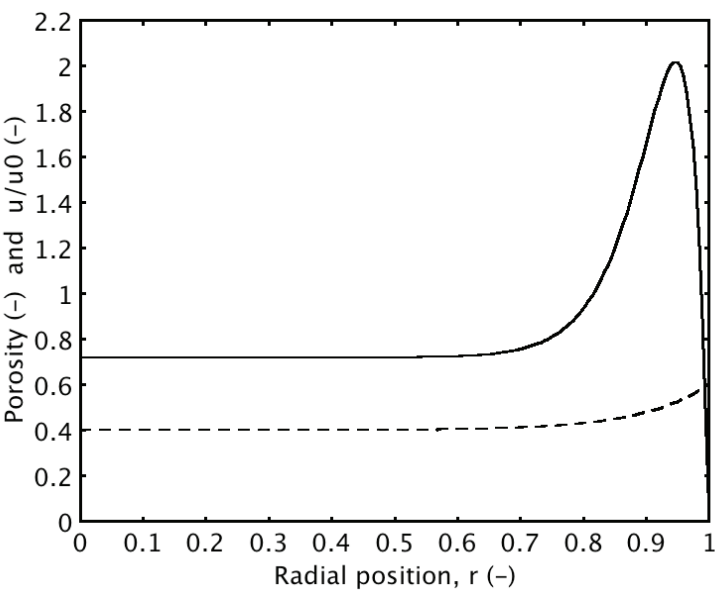


Figure 2. Velocity (—) and porosity (---) profiles across the fixed-bed reactor

Energy balance:
Fluid phase:

$$\psi a \frac{\partial T_f}{\partial t} = \frac{1}{Pe_{h,a}} \left(\frac{\partial^2 T_f}{\partial z^2} \right) + \frac{1}{Pe_{h,r}} \frac{1}{r} \frac{\partial}{\partial r} \left(r \frac{\partial T_f}{\partial r} \right) - \frac{\partial T_f}{\partial z} + St_h (T_s - T_f) \tag{8}$$

Solid Phase:

$$\frac{(1-\psi) \rho_s c_{p,s}}{\rho_f c_{p,f}} a \frac{dT_s}{dt} = (1-\psi) Da R(C_s, T_s) - St_h (T_s - T_f) \tag{9}$$

where C_f and C_s are the dimensionless concentrations of carbon monoxide in the fluid and solid phases, respectively, and T_f and T_s are the dimensionless temperatures of the fluid and solid phases, respectively. The independent variables are the dimensionless time, t , and dimensionless radial, r , and axial, z , coordinates. The variables and parameters of the model are defined as

$$\begin{aligned} r &= \frac{r^*}{R_t}, \quad z = \frac{z^*}{R_t}, \quad t = \frac{t^*}{t_o}, \quad C_f = \frac{C_f^*}{C_{in}^*}, \quad C_s = \frac{C_s^*}{C_{in}^*}, \quad T_f = \frac{T_f^* - T_{in}^*}{\Delta T_{ad}^*}, \quad T_s = \frac{T_s^* - T_{in}^*}{\Delta T_{ad}^*}, \quad a = \frac{R_t}{t_o u}, \\ \Delta T_{ad}^* &= \frac{|\Delta H_r| C_{in}^*}{\rho_f c_{p,f}}, \quad Bi = \frac{h_w R_t}{k_{r,eff}}, \quad Pe_{m,a} = \frac{u R_t}{D_{a,eff}}, \quad Pe_{m,r} = \frac{u R_t}{D_{r,eff}}, \quad Pe_{h,a} = \frac{u \rho_f c_{p,f} R_t}{k_{a,eff}}, \\ Pe_{h,r} &= \frac{u \rho_f c_{p,f} R_t}{k_{r,eff}}, \quad St_m = \frac{a_p h_m R_t}{u}, \quad St_h = \frac{a_p h_p R_t}{u \rho_f c_{p,f}}, \quad Da = \frac{\rho_s R_t}{u C_{in}^*} \Re(C_s, T_s)|_{in} \end{aligned} \tag{10}$$

where the asterisk denotes variables with absolute values, and subscript *in* denotes inlet conditions. Parameter ΔT_{ad}^* is the adiabatic rise, i.e., the highest possible temperature rise in the reactor under adiabatic operation and full conversion of the reactant. In equation 10, $k_{a,eff}$ ($k_{r,eff}$) is the effective axial (radial) thermal conductivity, $D_{a,eff}$ ($D_{r,eff}$) is the effective axial (radial) mass diffusivity, ρ_f (ρ_s) is the fluid (solid) density, $c_{p,f}$ ($c_{p,s}$) is the fluid (solid) heat capacity, h_p (h_m) is the inter-phase heat (mass) transfer coefficient, h_w is the wall heat transfer coefficient, and a_p is the specific area of the solid. The dimensionless numbers appearing in the second and third rows of equation 10 are defined in the usual way. Equations 1-10 are solved subject to the following initial and boundary conditions: IC:

$$t = 0, C_f = C_s = 0, T_f = T_s = 0. \tag{11}$$

BCs:

$$\begin{aligned} z = 0, \forall r, -\frac{\partial C_f}{\partial z} &= -Pe_{m,a}(C_f - C_{in}), -\frac{\partial T_f}{\partial z} = -Pe_{h,a}(T_f - T_{in}) \\ z = L/R_t, \forall r, -\frac{\partial C_f}{\partial z} &= 0, -\frac{\partial T_f}{\partial z} = 0 \\ r = 0, \forall z, -\frac{\partial C_f}{\partial r} &= 0, -\frac{\partial T_f}{\partial r} = 0 \\ r = 1, \forall z, -\frac{\partial C_f}{\partial r} &= 0, -\frac{\partial T_f}{\partial r} = Bi(T_f - T_w) \end{aligned}$$

The material and energy balance equations 6-10, along with equations 1-5 and initial/boundary conditions 11, have been solved using a finite-element scheme under various process parameters and operating conditions. The latter and other process parameters are given in Table 1.

L	1 m	ρ_f	31.1 mol/m ³	ΔH_{co}^{ads}	30.60 kJ/mol
R_t	0.02655 m	cp_f	29.45 J/mol/K	b_{o2}^o	3.08 10 ⁻⁵ Pa ⁻¹
C_{in}^*	0.75 mol/m ³	cp_s	149.32 J/mol/K	ΔH_{o2}^{ads}	24.44 kJ/mol
u_o	0.1 m/s	k_r^o	39.7 kmol/kg/s	b_{co}^o	6.81 10 ⁻⁸ Pa ⁻¹
T_{in}^*	335 K, 380 K	ΔH_r	-23.8 kJ/mol	ΔH_{co}^{ads}	30.60 kJ/mol
T_w^*	335 K, 380 K	E_a	57.29 kJ/mol	Bi	0.24
ρ_s	10.6 kmol/m ³	b_{co}^o	6.81 10 ⁻⁸ Pa ⁻¹	Re	683

Table 1. Process parameters and conditions used in the simulations

Materials properties and effective transport parameters have been estimated using a number of widely known correlations (see Agnew and Potter, 1970; Bauer and Schlunder, 1978; Poling et al., 2000; Elsari and Hughes, 2002; Incropera and DeWitt, 2002; Klemme and van Miltenburg, 2002).

3. Simulation

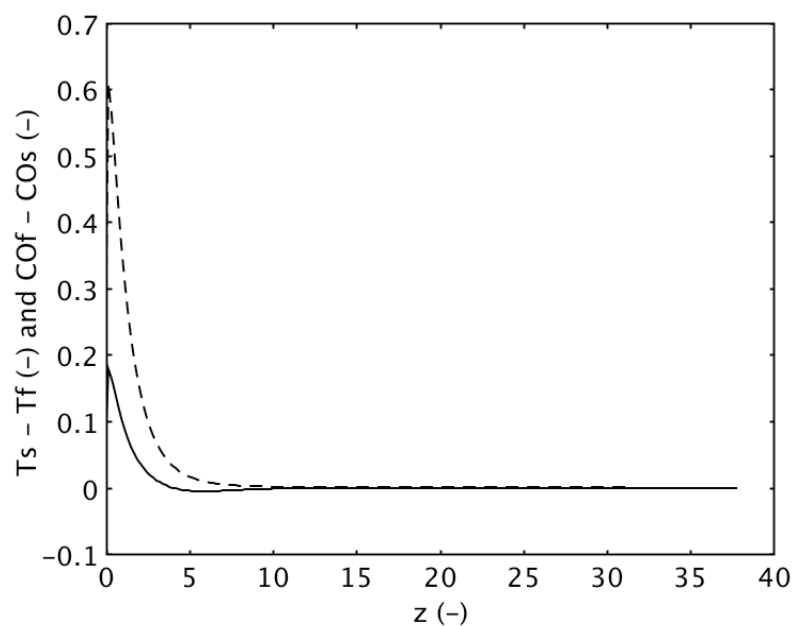


Figure 3: Inter-phase temperature (—) and concentration (——) differences along the bed centerline and at $t = 1$

Typical process conditions and parameters along with phase properties and kinetic data are listed in Table 1. For all simulation runs, the value of the temperature of the reactor wall, T_w^* , coincides with the value of the initial reactor temperature as well as the value of the reactor inlet temperature, T_{in}^* . The Reynolds number corresponds to the value of the velocity at the reactor core, i.e., $u(r) = u_0$. The simulation results have been obtained at two values of $T_{in}^* = T_w^*$, starting from 380 K at fixed inlet concentration, C_{in}^* , and velocity profile. The other value of the temperature has been identified so as to have the reactor under runaway conditions by keeping all other parameters constant.

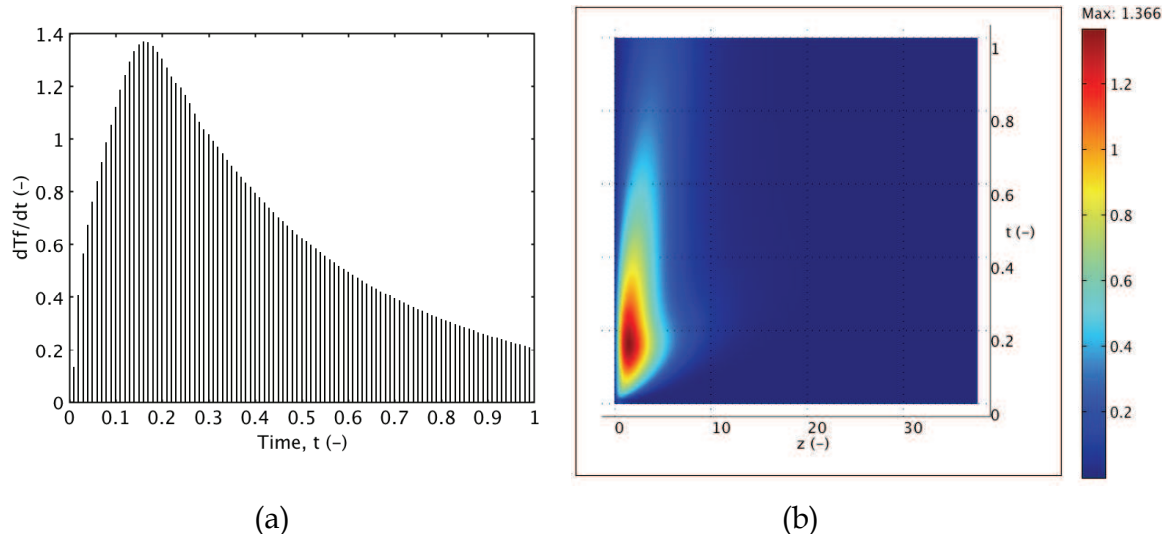


Figure 4. Time derivative of fluid temperature versus time along the bed centerline (a), and versus time and axial direction (b)

3.1 Inlet temperature = 380 K

Figure 3 depicts the difference in temperature and concentration between the solid and fluid phases. As it can be noticed from the figure, the model is clearly two dimensional with the largest inter-phase temperature and concentration differences just downstream the fixed-bed inlet. In this region of the bed, the temperature reaches a maximum value (referred to as a hot spot) as shown in Figure 4, where the inter-phase temperature difference reaches ~40 K in absolute value. The time dependence of the derivative of the fluid temperature, dT_f/dz , throughout the reactor and along the bed centerline ($r = 0$) is illustrated in Figure 4 (a). In this figure, each vertical line represents the entire axial direction envelope. The time derivative of the fluid temperature increases to reach a maximum value then decreases over time. Figure 4 (b) is a surface map of the derivative of the fluid temperature as a function of the axial position and time. As time goes by, the change in the fluid temperature increases to reach a maximum value corresponding to time $t \cong 0.2$ (see Figure 4, b), followed with a monotonous decrease with time thereafter.

3.2 Inlet temperature = 335 K

In this case, the inlet temperature of the fluid has been decreased from 380 K until it has reached the value of 335 K at which runaway occurs (this is elucidated in the next section).

The change in the fluid temperature as a function of time along the reactor centerline is shown in Figure 5 (a). The temperature derivative increases with time initially then it relatively levels off before it increases over time again. Over the entire time interval, this increase is monotonous though. The surface map of the time derivative as a function of time and along the bed centerline is depicted in Figure 5 (b). This figure illustrates the appreciable increase in the time derivative of the fluid temperature towards large values of time downstream the fixed-bed inlet.

4. Runaway Criterion

The runaway condition has been defined in various ways in the classical parametric sensitivity literature (Chambré, 1956; Barkelew, 1959). In both studies the use of isoclines in the determination of the runaway criterion was introduced. This approach provides a graphical method for determining runaway conditions in a reactor. Several other studies have used modifications of this approach, defining the runaway condition by locating maxima in some parameter of the reaction system. All of this work, however, was performed using a steady-state analysis of the system. Of special relevance to the present work is the criterion whereby thermal runaway occurs in steady-state tubular reactors when the second derivative of the temperature is larger than zero, i.e., runaway is identified by the presence of an inflection point in the temperature profile ahead of the hot spot locus (developed by Bashir et al., 1992).

In the present work, a particular characteristic of the dimensionless temperature profile is advanced to define runaway for the transient system under investigation. Using the inflection point criterion above, we have noticed that the latter coincides with a special trend in the variation of the time derivative of the fixed-bed temperature. For both cases of the previous section, Figure 6 depicts the fluid temperature and the corresponding space second derivative, dT_f^2/dz^2 , along the centerline of the fixed-bed reactor. From Figure 6 (a), corresponding to $T_{in}^* = T_w^* = 380$ K, there is no indication of reactor runaway as the second

derivative approaches the zero line from the negative side but does not cross it before the hot spot locus. At $T_{in}^* = T_w^* = 355$ K on Figure 6 (b), however, the second derivative breaks through the zero line before the hot spot locus and takes on positive values: runaway occurs. This result has been compared to a few variations of the temperature of the bed, and found to coincide with a trend whereby the first time derivative of the temperature increases monotonously over time throughout the reactor (see Figure 5). In other terms, for runaway to occur dT_f^2/dt^2 must be positive over the entire time interval. This finding holds for other runaway conditions as we have changed the value of the inlet concentration for $T_{in}^* = T_w^* = 355$ K.

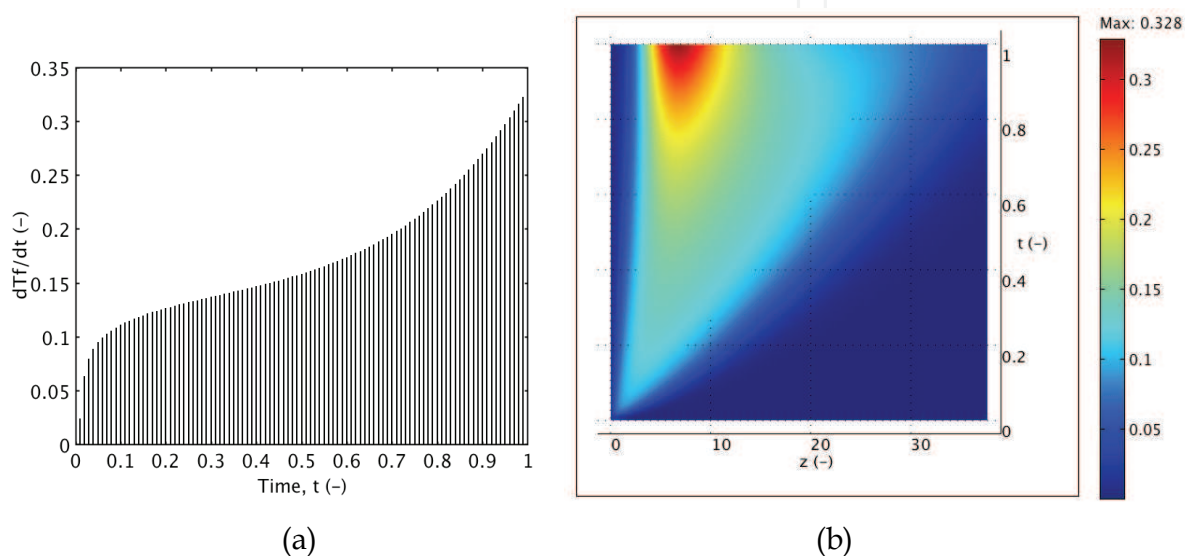


Figure 5. Time derivative of fluid temperature versus time along the bed centerline (a), and versus time and axial direction (b)

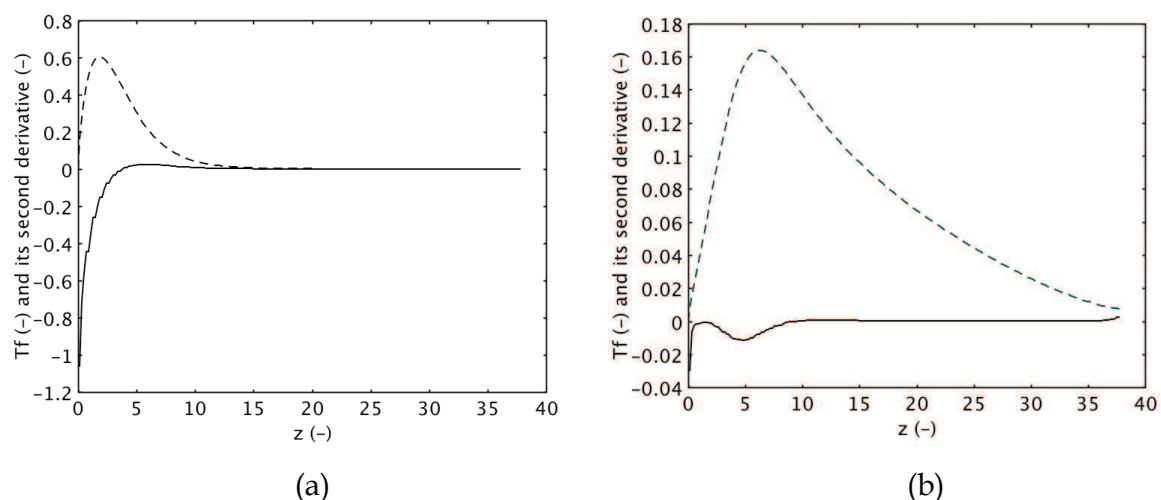


Figure 6. Fluid temperature (—) and its space second derivative (---) versus axial direction along the centerline at $t = 1$ for $T_{in}^* = T_w^* = 380$ K (a) and 335 K (b)

5. Conclusions

In this chapter we have discussed the thermal behavior of a fixed-bed tubular reactor with an exothermic chemical reaction. The work performed in the present study has focused on developing a transient criterion for thermal runaway. A criterion for the onset of thermal runaway has been advanced whereby the second time derivative of the bed temperature must be positive over the entire time interval. The proposed criterion, although qualitative in nature, is simple and can potentially be used to preclude the occurrence of runaway in fixed-bed tubular reactors. For practical purposes, a quantitative expression of the transient criterion needs to be developed in the future.

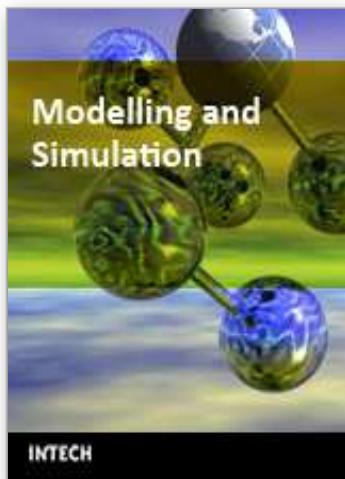
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