Influence of Cooling Design on Fixed-Bed Reactors Dynamics

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Tubular fixed-bed reactors of the heat-exchanger type are selected commonly in industrial plants to carry out highly exothermic reactions. Due to the important heat effects involved, these units usually exhibit the well-known problems of a pronounced maximum in the axial temperature profile (hot spot), combined with high parametric sensitivity.

Ray (1972) and Jorgensen (1986) reviewed publications related to the dynamics and control of these types of reactors. Except for autothermal reactors, which will not be included in the present discussion, all their references assumed constant-coolant temperature. However, this is a strong hypothesis, valid only when boiling or perfectly mixed fluids are used at the shell side. The effect of different cooling designs on the dynamic behavior of a wall-cooled catalytic reactor has been analyzed only by Gatica et al. (1989). These authors analyzed the response of axial temperature and concentration profiles to changes in the reactant's inlet conditions for the cocurrent design. Nevertheless, more critical disturbances are changes at the inlet coolant temperature which is often used as an indirect manipulated variable for control purposes. Therefore, the influence of this variable on the reactor dynamics will be studied in the present work.

Recent steady-state simulation results have shown that the behavior of this type of reactors depends strongly on the mutual direction of the reacting fluid and coolant streams. In a recent article, three basic cooling schemes (countercurrent, cocurrent, and perfectly-mixed coolant) were analyzed (Borio et al., 1989a). Their conclusion was that for equivalent production rates, the steady-state cocurrent operation yields the lowest values for both maximum temperature and parametric sensitivity. In a second contribution, the authors defined the optimal cocurrent coolant flow rate leading to conditions of maximum attainable safety and developed a simple expression which enables to predict its value (Borio et al., 1989b).

As optimal operation conditions cannot be defined only on the basis of steady-state analysis, this work complements the above-mentioned works by means of a similar development, performed under nonsteady-state conditions. The underlying idea is that to be confirmed as the best choice, the optimal design found from steady-state analysis should demonstrate also to exhibit an acceptable dynamic performance.

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The Mathematical Model

A dynamic pseudo-homogeneous one-dimensional modeling of a wall-cooled fixed-bed reactor leads to the following set of differential equations:

$$\alpha\frac{\partial p_A}{\partial t} = -\frac{\partial p_A}{\partial z} - A\,p_A\,k_\infty \exp(-a/T) \tag{1}$$

$$\beta\frac{\partial T}{\partial t} = -\frac{\partial T}{\partial z} + B\,p_A\,k_\infty \exp(-a/T) - C(T-Tc) \tag{2}$$

$$\gamma\frac{\partial Tc}{\partial t} = -\frac{\partial Tc}{\partial z} + D(T-Tc) \tag{3}$$

The chosen reaction rate expression corresponds to a pseudo-first-order kinetics, which has been claimed to be adequate for the catalytic air oxidation of o-xylene to phthalic anhydride (van Welsenaere and Froment, 1970).

The steady-state version of this model obtained by canceling the time partial derivatives corresponds exactly to the equations utilized in previous works (Soria López et al., 1981; Borio et al., 1989a,b). This correspondence will be useful to investigate the reactor dynamics around some characteristic steady-state operations of interest. The initial and boundary conditions used to solve the proposed mathematical problem are the following:

$$p_A(0,z)=p_{AS}(z),\ T(0,z)=T_s(z),\ Tc(0,z)=Tc_s(z)$$
$$p_A(t,0)=p_{AO},\ T(t,0)=Tc(t,0),\ Tc(t,j)=Tc_j,$$
$$\text{with}\ \begin{cases} j=0 & \text{cocurrent} \\ j=L & \text{countercurrent} \end{cases} \tag{4}$$

The simplifying assumptions adopted in the model, together with the values of the parameters and the rate expression used in the calculations can be found elsewhere (Soria López et al., 1981; Gatica et al., 1989). The global heat-transfer coefficient was estimated as in a previous contribution (Borio et al., 1989a).

It is worth noting at this point that in the dynamic model of Eqs. 1 to 3, the coolant flow rate Wc affects the parameters D and γ, both related to the energy balance in the cooling medium. In the present work, a vectorial character similar to that described by Borio et al. (1989a) will be assigned to Wc.

In this way, with the boundary conditions as given by Eq. 4 and using the proper sign for Wc, the model will be automatically adequate to cocurrent ($Wc>0$) or countercurrent ($Wc<0$) schemes. Moreover, as large (positive or negative) Wc values cancel both γ and D (which is equivalent to drop the convective term), the proposed model will also hold for the constant coolant temperature design.

To solve the dynamic model, finite differences were used to discretize in the spatial coordinate. Sixty axial grid points were used. Integration in time was performed by means of a Gear algorithm.

Selection of a Set of Initial Operation Conditions

Due to the highly nonlinear characteristics of the model, the dynamic behavior of the reactor will not necessarily be similar for different initial steady-state operating conditions. Therefore, each distinctive operating regime should be taken into account to perform an adequate dynamic analysis of the reactor.

A solution of the steady-state model yields, among other results, the temperature profile $T(z)$ along the reactor tube. Parametric sensitivity profiles can also be obtained by means of three additional equations which can be easily derived from the model. In the present work, the maximum temperature (T_m) and the absolute extreme in the axial profile of the reactants temperature sensitivity with respect to the inlet coolant temperature (S^*), will be used to characterize both profiles (Borio et al., 1989a). In Figure 1, values of T_m and S^* for steady-state operations yielding the same production rate have been plotted against Wc^{-1}. Also in Figure 1, the selected operating conditions 1 to 6 are shown. This group of profiles was chosen as representative of the different steady-state conditions because:

- Point #2 was defined as the most convenient operation from the safety standpoint, provided it exhibits the lowest value for T_m and low S^*. The coolant flow rate corresponding to this operation can be predicted analytically (Borio et al., 1989b).

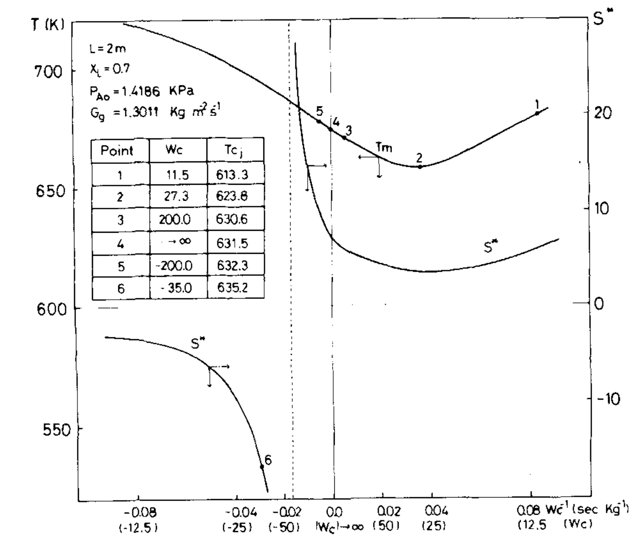

Figure 1. T_m and S^* vs. Wc^{-1} curves at conditions of constant production rate and fixed total reactor length.

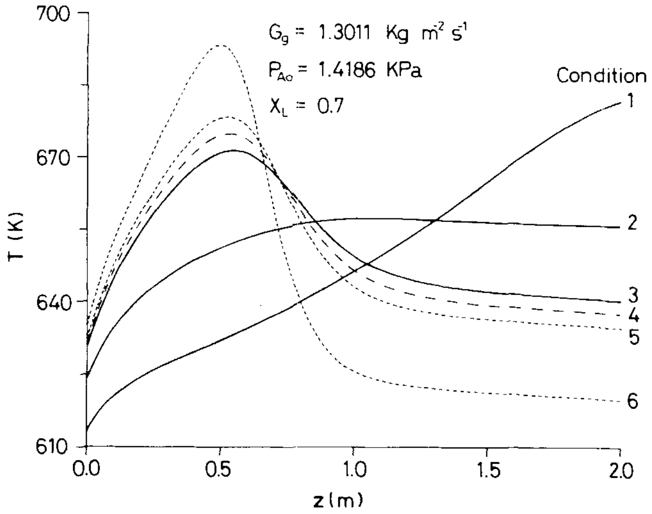

Figure 2. Axial temperature profiles for the selected steady-state operating conditions.

- Point #4 corresponds to the well-mixed coolant design.
- Point #6 is included to illustrate countercurrent operations with $S^*<0$.
- Point #5 represents the most common industrial conditions, that is, countercurrent scheme with high coolant flow rate, chosen to keep the total increase of coolant temperature ($Tc_o - Tc_L$) around 3 K.
- Point #3 corresponds to inlet conditions analogous to #5, but cocurrent design.
- Point #1 has been chosen to illustrate the dynamic behavior of cocurrent operations with very low coolant flow rates.

The inlet conditions defining each of the steady-state operations listed above are included in Figure 1. The steady-state temperature profiles corresponding to each of the selected operation conditions are plotted in Figure 2.

Results and Discussion

To yield a more comprehensive discussion, the results obtained from the nonsteady-state simulation will be analyzed from two different standpoints. First, the transient evolution of the whole temperature profile will be discussed. A more suitable approach for control purposes will follow, involving the analysis of the dynamic behavior of the reactor temperature at a given axial location.

Dynamic behavior of the temperature profiles

It is convenient to start the analysis from the countercurrent low coolant flow rate condition defined as point 6 in Figure 1. The evolution in time of its temperature profile, when step changes of ± 1 K enter the reactor at $t=0$ s, is shown in Figure 3. Additional simulations demonstrated that despite the size of the perturbation, the dynamic responses of the reactor starting from this initial condition were all basically the same, that is, temperature profiles evolving either to runaway (for positive inputs, $\Delta Tc_L>0$) or to cooling down ($\Delta Tc_L<0$). This behavior is known to be characteristic of unstable steady states. Therefore, this result will be used only to verify the criterion proposed

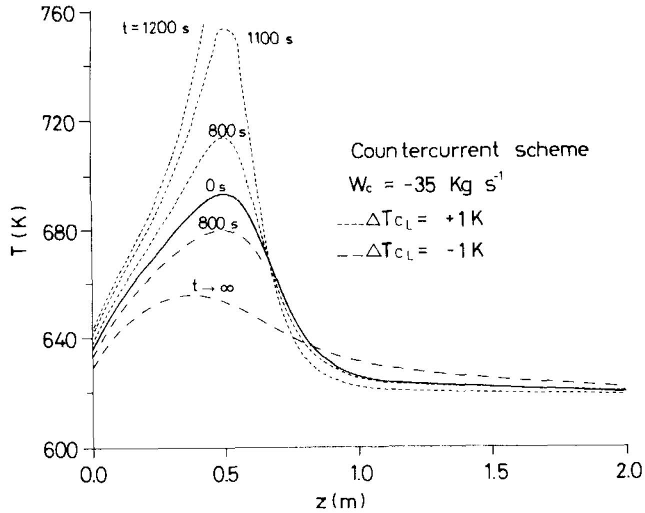

Figure 3. Dynamic evolution of the axial temperature profile for unstable condition 6.

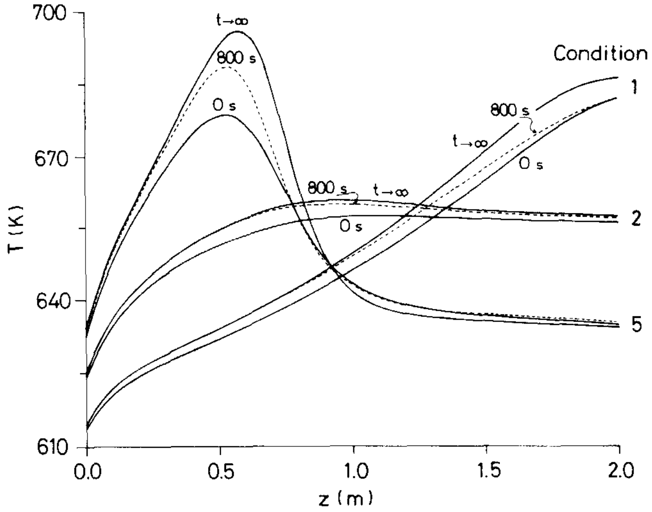

Figure 4. Dynamic evolution of the axial temperature profile for conditions 1, 2 and 5.

by Borio et al. (1989a), who claimed that unstable-state operations were characterized by the condition $S^* < 0$.

The remainder-selected conditions 1 to 5 verify $S^* > 0$, which correspond to stable (industrially feasible) steady states. Therefore, in what follows they will be put together to perform a comparative analysis of the reactor dynamics at different initial operating conditions.

Inspection of the curves in Figure 2, which correspond to different stable operating conditions, permits us to distinguish at least three distinct types of initial steady-state profiles:

I) Temperature always increases with the axial coordinate (as in condition 1, which would develop a hot spot beyond the exit of the reactor).
II) Pseudo-adiabatic operation or profiles which exhibit a very flat maximum (condition 2).
III) Temperature profiles showing a well-defined hot spot (conditions 3 to 5).

Simulated dynamic responses for initial conditions 1, 2 and 5 are shown in Figure 4. All these responses correspond to a 1 K step change in Tc_j. Responses from initial conditions 3 and 4 are not included in this figure due to the dynamic similarity they exhibit with their steady-state "analogous" condition 5.

A well-known feature of the transient behavior of adiabatic fixed-bed reactors is the faster evolution of local temperatures toward their final steady-state value occurring at the inlet section. This effect, mainly due to the comparatively large thermal conductivity and heat capacity of the catalyst particles, should also be observed in the responses of wall-cooled fixed-bed reactors. When the disturbance is a step change in Tc_j, however, the heat transported in the axial direction by the coolant itself introduces a new contribution to the global dynamic response. Being of convective nature, the speed of this transport is directly related to the coolant flow rate. Therefore, for industrial conditions of type III, the coolant contribution should become more important. All this reasoning is confirmed by the evolution of the temperature profiles shown in Figure 4. Curves corresponding to initial profiles of type I are effectively "slow" to transmit the disturbances to the outlet, which is analogous to the behavior of adiabatic reactors. Contrarily, if initial profiles are of type III, a perturbation in Tc_j will introduce changes at all axial locations almost at the same time.

Finally, another characteristic dynamic aspect of these types of units is revealed from inspection of the curves corresponding to condition 5 in Figure 4. It concerns the confused information that would be obtained from temperature measurements performed downstream from the hot spot. Hot spots will always be present when the reactor is held at operation conditions of type III, which correspond to the range of current operation. This anomalous behavior, which led to the extended industrial practice of measuring the temperature before the hot spot, will be discussed in more detail in the next section.

Temporal responses at a given axial position

As the tubular reactor is a distributed-parameter system, the evolution in time of the reactant's temperature at a given axial position would correspond to the signal obtained from a sensor located at that point. Signals of this kind is usually selected as measured variables in classical schemes for the temperature control of chemical reactors.

The optimal location of temperature sensors in tubular catalytic reactors has been discussed by many authors (for example, Kumar and Seinfeld, 1978; Harris et al., 1980; Alvarez et al., 1981; Cinar, 1984). As they assumed constant-coolant temperature design, all their initial steady-state profiles presented a well-defined hot spot located near the reactor inlet. They concluded that the best location for a temperature sensor, in other words, the axial point presenting a desirable dynamic behavior, is just before the hot spot. Due to the different shape of some of the selected profiles, these results cannot be directly extended to the present case, and in what follows this problem will be analyzed in more detail.

It may be practical to start the analysis discussing the dynamic behavior of the outlet temperature, provided that the reactor outlet is the easiest place to install a thermocouple probe. For the present case, this is equivalent to locating the sensor at $z = 2$ m. The response of the outlet temperature to a step of 1 K in Tc_j, for the different initial steady-state con-

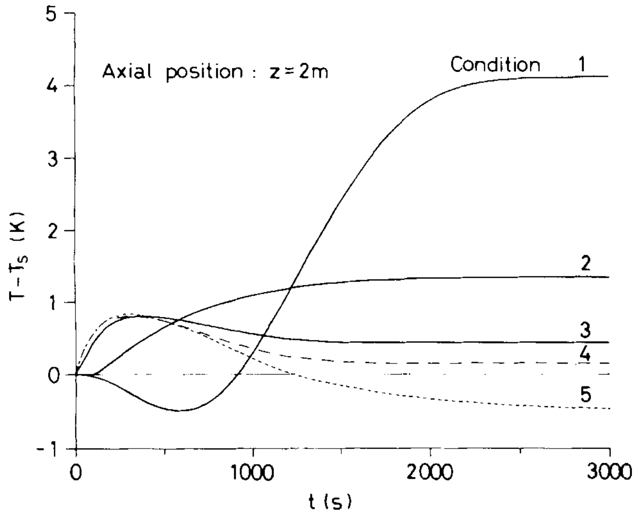

Figure 5. Dynamic behavior of the deviation temperature at $z=2$ m.

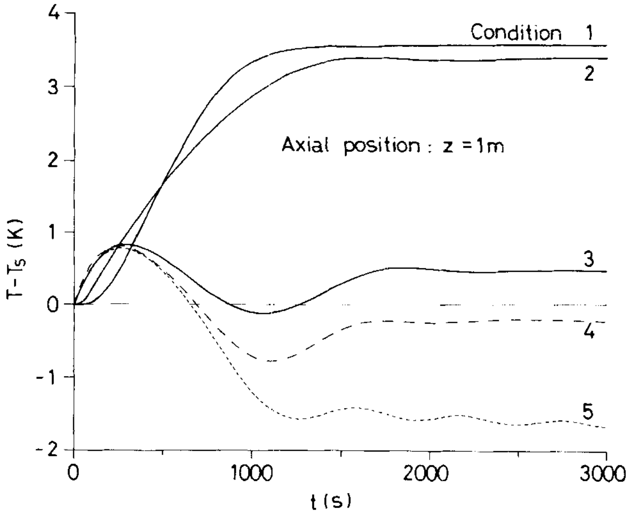

Figure 6. Dynamic behavior of the deviation temperature at $z=1$ m.

ditions 1 to 5 under study, is shown in Figure 5. A primary observation of these curves indicates that for current industrial designs (conditions 3 to 5), the selection of T_L as the measured variable may not be an adequate choice. In fact, the responses obtained illustrate the low sensitivity of this variable to step changes at the reactor inlet. Nevertheless, the better response among these three designs is given by the cocurrent scheme (condition 3). Conversely, the worst open-loop behavior corresponds to the countercurrent case, which exhibits a significant inverse response. Low flow rates of cocurrent schemes (as condition 1) lead to the highest sensitivities. However, an important inverse response also appears, which would necessarily lead to undesirable control problems. From a control standpoint, condition 2 offers the best of all responses. The sensitivity of the outlet temperature, while still low, is higher than that of the industrial cases, and T grows monotonically after a small pure delay. This behavior can therefore be conveniently described through a linear, overdamped, second-order dynamic model.

The second variable to analyze is the temperature at $z=1$ m. The different open-loop responses of the temperature at the middle of the reactor tube to a step change in Tc_j are shown in Figure 6. As can be clearly seen, this location can only be a reasonable choice for a practical control structure in the case that the reactor is to be operated around conditions similar to 1 or 2. Indeed, for current industrial conditions 3 to 5, the inadequate dynamic behavior of the temperature would make the choice of this signal unacceptable as a measured variable.

The final location to analyze corresponds to $z=0.4$ m. The evolution of the temperature corresponding to different initial steady-states under study is drawn at Figure 7. At this location, all responses show to be fast and to increase monotonically with time. Therefore, they can be easily approximated through linear dynamic models of low complexity. A similar behavior was found at all axial locations between the reactor inlet and the hot spot site.

Once the dynamics of the local temperature has been described at three different axial locations and for the five selected initial conditions, the problem of the temperature-sensor location can be re-examined. Results shown at Figure 5 prove that high flow rates of cocurrent or countercurrent schemes have a dynamic behavior similar to the constant-coolant temperature design. As can be expected, for all current industrial designs, the best sensor location is just before the hot spot. However, even for these designs, the previous considerations do not constitute a satisfactory solution to the complex problem of selecting the measured variable for control purposes. In fact, the mechanical difficulties related with the insertion of the thermocouple probe in a precise point of the reactor tubes must be added to the uncertainty in defining the hot spot location. Moreover, in many cases the hot spot travels toward the outlet due to catalyst deactivation phenomena.

Hopefully, inspection of Figures 5 to 7 suggest that a global improvement to the problem can be achieved by simply adopting the cocurrent scheme and choosing an adequate value for

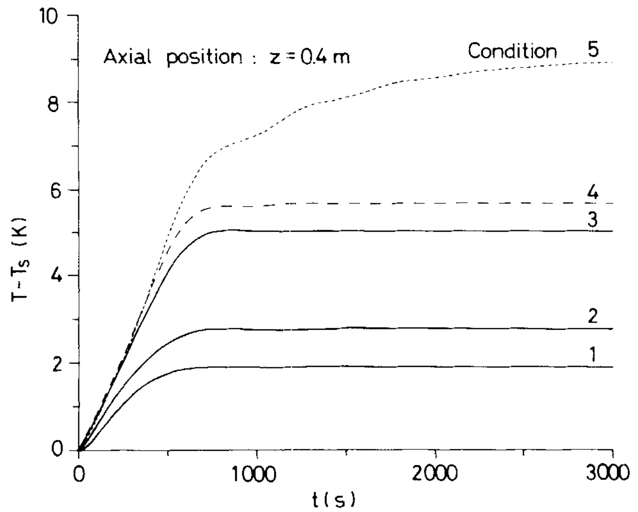

Figure 7. Dynamic behavior of the deviation temperature at $z=0.4$ m.

the coolant flow rate. In fact, operating the reactor around condition 2, an acceptable open-loop dynamic behavior will be found at all axial locations, which simplifies the measurement variable selection problem.

All the above results confirm that the optimal steady-state operation previously defined by Borio et al. (1989b) is also a good choice from the dynamic standpoint.

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Notation

a = E/R
A = $(\rho_b P M p_{Bo})/(G_g)$
B = $[\rho_b(-\Delta H) p_{Bo}]/(G_g Cp_g)$
C = $(4U)/(d_t G_g Cp_g)$
Cp = specific heat, $kJ \cdot kg^{-1} \cdot K^{-1}$
ds = shell diameter, m
dt = tube diameter, m
D = $(\pi\, dt\, N\, U)/(Wc\, Cp_c)$
E = activation energy, $kJ \cdot kmol^{-1}$
G = specific mass flow rate, $kg \cdot m^{-2} \cdot s^{-1}$
k_∞ = pre-exponential factor, $kmol \cdot s^{-1} \cdot kg\ cat^{-1} \cdot kPa^{-2}$
L = reactor length, m
M = molecular weight of gaseous mixture, $kg \cdot kmol^{-1}$
N = number of reactor tubes
p = partial pressure, kPa
P = total pressure, kPa
R = universal gas constant, $kJ \cdot kmol^{-1} \cdot K^{-1}$
S^* = absolute extreme (max, min) of $\partial T(z)/\partial Tc_j$ curves, K/K
t = time, s
T = reactor temperature, K
Tc = coolant temperature, K
U = overall heat-transfer coefficient, $kW \cdot m^{-2} \cdot K^{-1}$
Wc = coolant flow rate, $kg \cdot s^{-1}$
x = conversion
z = axial coordinate, m

Greek letters

α = $(\epsilon_b M P)/(G_g R T)$
β = $(\epsilon_b \rho_g Cp_g + \rho_b Cp_{cat})/(Cp_g G_g)$
ΔH = heat of reaction, $kJ \cdot kmol^{-1}$
ϵ = porosity, m^3/m^3
γ = $[\rho_c \pi (ds^2 - dt^2 N)]/(4\, Wc)$
ρ = density, $kg \cdot m^{-3}$

Subscripts

A = o-xylene
b = bed
B = second component
c = coolant
cat = catalyst
f = final
g = gas
j = at coolant inlet axial position
L = at axial position, $z = L$
m = maximum
o = at axial position, $z = 0$
s = steady state

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