Design Considerations for Tubular Reactors with Highly Exothermic Reactions

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Practical guidelines are required for the design and operation of complicated catalytic packed-bed reactors. Primary among design considerations is the avoidance of operating regions of high parametric sensitivity, in which small changes in operating conditions can lead to thermal runaway in the reactor. Existing criteria for predicting these regions rely on complex mathematical formulations for differential sensitivity between input and output variables. The present work centers on the development of practical design criteria for avoiding reactor instability and temperature sensitivity in multitubular packed-bed reactors. A set of simple guidelines is proposed for the sizing of reactors and proper selection of operating conditions. The implications of these open-loop sensitivities for a controlled reactor are investigated. It is shown through simulation studies that violation of the proposed criteria leads to control problems and difficulty in operating at the design point.

Introduction

Multitubular packed-bed reactors display a rich range of steady-state and dynamic behavior. Often, there is an economic incentive to operate near a region of parametrically sensitive behavior. In these regions, small changes in inlet conditions and/or physical parameters can lead to catastrophically large excursions in the bed temperature. Clearly it is important to establish guidelines for safe reactor operation, and moreover it is advantageous to specify *a priori* the design of a reactor which avoids these dangerous operating conditions.

In this article, some simple design criteria for sizing a reactor and choosing reaction conditions are described. The usual uncertainties associated with catalyst properties and reaction kinetics suggest the formulation of conservative criteria. This conservatism might be reduced with more detailed modeling. The proposed criteria, however, are far simpler to evaluate and provide reasonable bounds.

It is possible to formulate these design guidelines in terms of three straightforward criteria:

- The reactor should have a mild radial temperature profile. This protects against self-acceleration of the reactor at the center of the reactor.
- The reactor temperature profile should not exhibit sensitivity to small perturbations in the inlet conditions and physical parameters.
- The pressure drop must be kept at an acceptable level.

Initially it is assumed that the catalyst particle has a single steady state, but, as will be shown, it is possible to modify the parametric sensitivity criterion to predict the occurrence of multiple steady states.

Of particular interest in this study is the sensitivity criterion. This property has been described in great detail for a variety of reactors (Emig et al., 1980; Morbidelli and Varma, 1985, 1986a,b; Puszynski et al., 1981; Varma, 1989). These studies focus on the formulation of normalized differential sensitivities for describing conditions of runaway behavior. However, the direct implications of this sensitivity theory for reactor design are not apparent. Alternatively, extensive simulations can be used for predicting these conditions. However, there are often large uncertainties associated with the parameters in these models. A primary goal of this work is the formulation of an incipient runaway condition in terms of *practical* design variables which are well known (diameter, length, catalyst prop-

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erties, and so on) and can be directly incorporated into an efficient design scheme.

The preceding discussion focused on open-loop (uncontrolled) packed-bed reactor operation. There are, however, several incentives for using feedback control in a chemical reactor (either tubular or continuous stirred tank):

- Stabilization of an unstable operation
- Mitigation of open-loop sensitivity
- Improvement of overall performance

Unstable operation is characterized by the presence of multiple steady states with hysteresis between ignited and quenched states. In contrast, open-loop sensitivity is characterized by a single stable steady state. In an ideal packed-bed reactor or CSTR, open-loop sensitivity is manifested at conditions where small perturbations in operating conditions lead to huge excursions in the single stable reactor temperature. In the ideal CSTR, the operating conditions can be further perturbed to lead to bifurcations in the steady-state temperature; thus introducing steady-state multiplicity and unstable modes of operation. In an *ideal* tubular reactor, the lack of a mechanism for thermal feedback within the reactor's contents precludes the possibility of multiple steady states. In practice, however, the countercurrent conduction of heat through the packing and tube wall, countercurrent cooling and feed preheating with the effluent, all contribute to thermal feedback and lead to steady-state multiplicities in the tubular reactor. From a practical perspective, it is not important whether thermal runaway occurs from high parametric sensitivity or ignition to a new steady state. The overriding concern is tight control of such temperature excursions.

A number of academic studies have demonstrated that multiplicity and open-loop sensitivity in a CSTR are easily handled with simple proportional control action. (See Aris and Amundson, 1958, for earliest work in this area.) The relatively straightforward design of industrial CSTRs suggests that these theoretical stabilizing properties should also work in practice (with proper mixing and so on).

In principle, the same stabilization theory can be invoked for a packed-bed reactor, and safe closed-loop operation can be attained in a region of open-loop sensitivity (see, for example, Seider et al., 1990). A number of experimental studies have also focused on the stabilizing effects of feedback control in a *single* tube reactor (Hansen and Jörgensen, 1976; Hoiberg et al., 1971; McDermott et al., 1986; Tremblay and Wright, 1974). The interested reader is referred to a comprehensive survey by Jörgensen (1986) for additional studies on packed-bed reactor control. These ideas, however, were never introduced into industrial practice. These theoretical and experimental studies have largely ignored some of the key industrial issues, in particular, the consideration of *multitubular* reactors. Furthermore, industrial control of tubular reactors has focused on the maintenance of cooling jacket temperatures at prescribed steady-state values. During transient operation, key variable measurements are often unavailable. In addition, manipulated variables are constrained, thus limiting their effectiveness. These two considerations often preclude the possibility of dynamic reactor stabilization.

This brings into question the validity of these theoretical stabilization studies and their relevance to industrial packed-bed reactor control. As this article will demonstrate through analysis and simulation, the only recourse for safe reactor operation is strict adherence to design guidelines of the type presented here.

Tubular Reactor Design Criteria

Radial temperature profile

In an exothermic reaction taking place in a tubular reactor, the heat evolved at the center has to be transferred to the wall. If this radial temperature difference, ΔT_r, is too large, the reactions will self-accelerate at the center of the reactor. Consequently, the first design criterion will focus on the permissible size of ΔT_r, and the translation of this specification into meaningful reactor parameters.

We will show that a good measure of the range of acceptable radial temperature difference is given by the parameter:

$$\Theta = \frac{RT_w^2}{E} \tag{1}$$

Consider a Taylor series expansion of the temperature profile from the wall to the center of the bed:

$$-\frac{1}{T_{\text{center}}} = -\frac{1}{T_w} + \frac{\Delta T_r}{T_w^2} + \cdots \tag{2}$$

Then, the reaction rate at the center can be approximated as follows:

$$r = k_0 e^{-E/RT} f(C) \sim k_0 e^{E/RT_w} e^{\Delta T_r/\Theta} f(C) \tag{3}$$

This type of approximation, referred to as the "method of expanding the exponent" (Frank-Kamenetskii, 1955), has been used in combustion studies to predict first-order effects in simple mathematical terms. From this equation, it is evident that Θ is a scaling parameter for ΔT_r. Furthermore, consider the following Taylor series expansion:

$$e^{\Delta T_r/\Theta} \sim 1 + \frac{\Delta T_r}{\Theta} + \left(\frac{\Delta T_r}{\Theta}\right)^2 \frac{1}{2!} + \cdots \tag{4}$$

A reasonable criterion for avoiding self-acceleration at the reactor center is to require that:

$$\frac{\Delta T_r}{\Theta} < 1 \tag{5}$$

This is consistent with the stationary theory of thermal explosion (Frank-Kamenetskii, 1955). From Eq. 4, it is clear that this will lead to a reaction rate that increases at most linearly with temperature along the radial direction in the bed. This criterion, however, involves the calculation of the radial temperature difference and is therefore limited as a design tool. From a pragmatic viewpoint, we require a criterion that depends on such quantities as the physical dimensions of the reactor, the catalyst properties, and the overall conversion.

Clearly, if self-acceleration at the center of the reactor is to be avoided, then the heat produced by the reaction must be quickly removed by convection and conduction (ignoring ra-

diation effects). The ratio of convective forces to conductive forces is given by the Peclet number:

$$Pe = \frac{GC_pD}{k_{eff}} \tag{6}$$

If one considers only dynamic contributions to k_{eff} (Froment and Bischoff, 1979), then the heat-transfer Peclet number can be approximated by the mass-transfer Peclet number which approaches 11 for high flow rates. Note that this yields an upper bound on the true heat-transfer Peclet number, and thus a conservative result in the following analysis. More detailed calculations can be used to refine this result as necessary.

Consider a small section of the reactor tube in which no mixing occurs. Recall that an order of magnitude interpretation for a Peclet number of 11 is that for a distance of about 5.5 diameters (or 11 radii), the heat from the center does not reach the wall (that is, for diffusion across r, the heat is convected across $11r$). Thus, if we consider a bed of length $5.5D$, no heat can reach the wall and, roughly speaking, operation is adiabatic. To avoid acceleration of the reaction rate at the center of the reactor, the adiabatic temperature rise over this unmixed portion of the bed must be small. The size of the tolerable temperature drop over this portion of the bed is provided by Eq. 5 and the fact that the adiabatic temperature rise per unit length is $[\Delta H\, r(T_{ave})/(C_pG)]$. Combining these facts yields the inequality:

$$\frac{1}{\Theta}\frac{\Delta H\, r(T_{ave})}{C_pG}\frac{11D}{2} < 1 \tag{7}$$

This criterion is still not directly suitable for design calculations as it requires the computation of the rate of heat production. For preliminary design purposes, we consider the following calculations which result in a formulation of the adiabatic temperature gradient as a function of meaningful reactor design variables. If the reactor is approximately isothermal, then basic design principles (Froment and Bischoff, 1979) for a first-order reaction lead to:

$$r_{max}(T) = [-\ln(1-x)]\frac{G}{L} \tag{8}$$

The design equation (Froment and Bischoff, 1979) for an adiabatic bed yields the following expressions for the temperature and conversion gradients along the bed:

$$\frac{dT}{dz} = \frac{\Delta H}{C_p}\frac{dx}{dz} \tag{9}$$

$$\frac{dx}{dz} = \frac{r(T)}{G} \tag{10}$$

Thus the maximum temperature gradient along the bed is given by:

$$\left(\frac{dT}{dz}\right)_{max} = \frac{\Delta H}{C_p}\frac{[-\ln(1-x)]}{L} \tag{11}$$

The adiabatic temperature rise over the full length of the reactor (for a conversion of x) is given by:

$$\Delta T_{ad}(x) = \frac{x\Delta H}{C_p} \tag{12}$$

This is combined with Eq. 11 and substituted for $(\Delta Hr)/(C_pG)$ in Eq. 7 to yield the final result

$$-\frac{1}{\Theta}\frac{\Delta T_{ad}(x)}{L}\frac{\ln(1-x)}{x}\frac{11D}{2} < 1 \tag{13}$$

or

$$\frac{L}{D} > 5.5\frac{\Delta T_{ad}(x)}{\Theta}\left[\frac{-\ln(1-x)}{x}\right] \tag{14}$$

where ΔT_{ad} is the temperature rise of the total feed at conversion x. To be safe, one might want to start with an L/D at least twice as large as the term on the right side. Note that the radial temperature criterion resulted in a requirement for a minimum L/D which is independent of space velocity and catalyst activity. It is just a function of conversion and:

$$\frac{\Delta T_{ad}(x)}{\Theta} = \frac{\Delta T_{ad}(x)E}{RT_w^2} \tag{15}$$

Up to this point it was assumed that the reactions taking place in the reactor are irreversible. We can modify this for reversible reactions if we know the maximum reaction rate. For a first order reversible reaction, we can simply substitute $-\ln[1-(x/x_e)]$ for $-\ln(1-x)$. For a second order irreversible reaction we get:

$$\frac{L}{D} > \frac{5.5\Delta T_{ad}(x)}{\Theta}\frac{1}{1-x} \tag{16}$$

where again x is the fraction of reactant converted and ΔT_{ad} is the adiabatic temperature rise of the total feed at conversion x. For reversible reactions or any more complex reaction expression, Eq. 16 becomes:

$$\frac{L}{D} > \frac{5.5\Delta T_{ad}(x)}{\Theta}\frac{L}{L^*} \tag{17}$$

where L^* is the length of the reactor that would be required to obtain the conversion in an isothermal reactor if the reaction rate were at the maximum rate throughout the reactor. For practical considerations, this parameter can be measured on a microreactor or a small pilot plant.

In Figure 1 values are plotted for the quantity (D/L^*) $[5.5\Delta T_{ad}(x)]/\Theta$ vs. the L/D ratio for typical industrial reactor data (Hlavacek, 1970).

The criterion in Eq. 17 dictates that (D/L^*) $[5.5\Delta T_{ad}(x)]/\Theta$ should be less than 1 which is clearly violated for these reactors. This reveals the conservative nature of the previous calculations. A less restrictive condition on the radial temperature profile will be derived in the following section on parametric sensitivity.

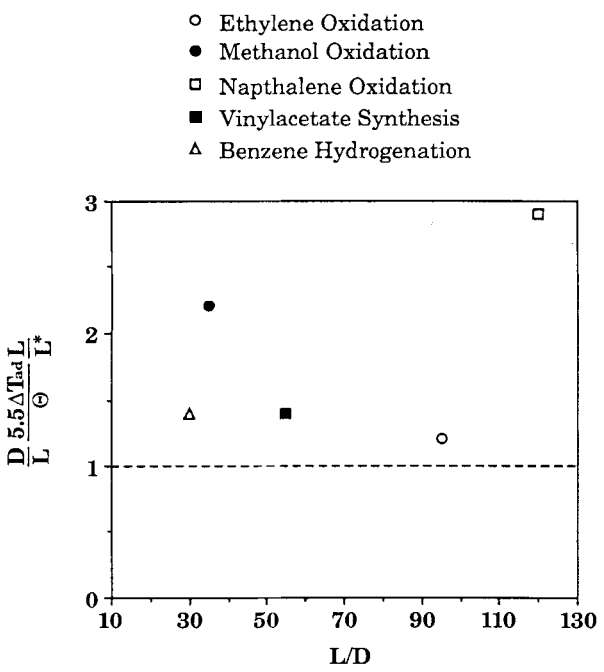


Figure 1. Verification of radial temperature criterion (data from Hlavacek, 1970).

Parametric sensitivity

Barkelew (1959) and others have formulated criteria for the temperature sensitivity of packed bed reactors to changes in operating conditions or physical parameters. However, these criteria are in terms of mathematical differential sensitivities which offer little insight for a practical design engineer. We will show that these criteria can be reduced to a very simple form involving practical design parameters.

To transfer the heat through the wall, a driving force close to the inside of the wall is required. When the reaction rate changes, the temperature close to the wall has to rise to compensate for the higher heat transfer required. Exercising good engineering judgment, we require that the difference between the average bed temperature and the coolant temperature $(T_{ave}-T_c)$ is small compared to Θ. As with the previous criterion, we can utilize the stationary theory for thermal explosions to calculate a reasonable bound on this scaled temperature difference $[(T_{ave}-T_c)/\Theta]<1$. Referring back to Eqs. 2, 3 and 4, this is understood as requiring the reaction rate to accelerate at most linearly along the radial direction. In terms of the "forces" involved, we know that in a well-mixed reactor, the rate of heat generation is balanced by the difference between the average bed temperature and the coolant temperature:

$$\frac{4h}{D}(T_{ave}-T_c)=r\Delta H \tag{18}$$

where h is the overall heat-transfer coefficient and T_{ave} is the average bed temperature. Thus our requirement becomes:

$$\frac{T_{ave}-T_c}{\Theta}=\frac{Dr\Delta H}{4h\Theta}<1 \tag{19}$$

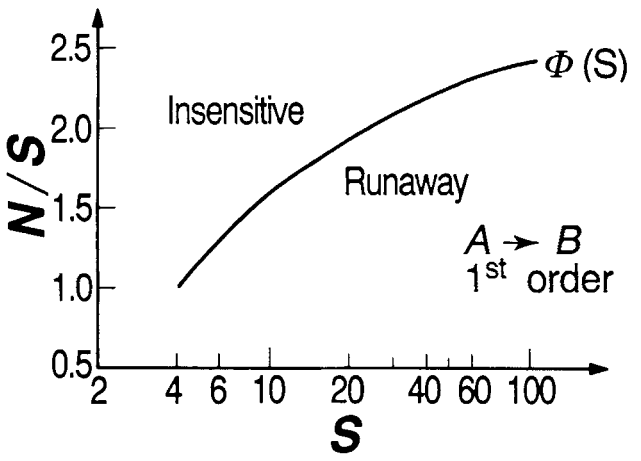


Figure 2. Barkelew's sensitivity criterion (from Barkelew, 1959).

Using the relationship in Eqs. 8 and 12:

$$r_{max}(T)\Delta H=GC_p\frac{\Delta T_{ad}(1.0)}{L}[-\ln(1-x)] \tag{20}$$

we get

$$\frac{\frac{GC_p\Delta T_{ad}(1.0)}{L}[-\ln(1-x)]}{\frac{4h}{D}\Theta}<1 \tag{21}$$

where $\Delta T_{ad}(1.0)$ is the theoretical adiabatic temperature rise for the complete feed at full conversion. While this is not a physically realizable temperature rise, it provides a useful measure for the calculation of heat effects. As before, we can substitute $x/(1-x)$ for the quantity in brackets for a second order reaction. This equation can be interpreted as a limit on the reactor diameter:

$$D<D_{max}=\frac{4}{v_s[-\ln(1-x)]}\frac{h\Theta}{C_p\Delta T_{ad}(1.0)} \tag{22}$$

where v_s is the space velocity. This suggests a plausible criterion for temperature sensitivity which will be compared with the approach used by Barkelew.

In his notation, the sensitivity criterion is given by:

$$\frac{DSC_pc_0k_0}{4he^{E/RT_w}}<\frac{1}{\Phi(S)} \tag{23}$$

where $\Phi(S)$ for a first order irreversible reaction is given in Figure 2 and

$$S=\frac{\Delta HE}{C_pRT_w^2} \tag{24}$$

and

$$N=\frac{4he^{E/RT_w}}{DC_pc_0k_0} \tag{25}$$

$\Phi(S)$ ranges in value from approximately 1.0 for small S to 2.5 for large S. In our notation, $S=[\Delta T_{ad}(1.0)]/\Theta$ and $c_0k_0e^{-E/RT_w}$ is simply $r(T_w)$. Thus, Barkelew's criterion (Eq. 23) becomes:

$$\frac{Dr(T_w)\Delta H}{4h\Theta}<\frac{1}{\Phi(S)} \tag{26}$$

Following the same manipulations as before, the final design criterion can be expressed as a maximum diameter for the tube:

$$D<D_{max}=\frac{1}{\Phi\left(\dfrac{\Delta T_{ad}(1.0)}{\Theta}\right)}\frac{4}{v_s[-\ln(1-x)]}\frac{h\Theta}{C_p\Delta T_{ad}(1.0)} \tag{27}$$

$\Phi(S)$ can be taken from Figure 2 and it changes from 1 to 2.5.

This result is very similar to our earlier result (Eq. 22), which was based on simple engineering rules of thumb. This confirms that Eq. 27 is a plausible form for the sensitivity criterion. Furthermore, Eq. 27 incorporates the additional correction term $\Phi(S)$ which, for large values of $\Phi(S)$, makes it more stringent than Eq. 22. The result in Eq. 27 should be made more conservative by introducing a safety coefficient as we do not know our parameters very accurately and they may change during operation. A safety factor of 2 is reasonable (that is, $D=0.5D_{max}$).

Instead of a look-up table approach to solving for $\Phi(S)$, it is possible to actually calculate an explicit value from the following correlation proposed by Morbidelli and Varma (1985):

$$\Phi(S)=2.718\Gamma\left[1-\left(\frac{S_0}{S}\right)^{2/3}\right] \tag{28}$$

where Γ is an empirical parameter given by:

$$\Gamma=\frac{8.7}{7.66+n^{0.6}} \tag{29}$$

and for n (reaction order) equal to 1, the expression for S_0 is:

$$S_0=\frac{4\gamma}{\gamma-4} \tag{30}$$

where γ is the dimensionless activation energy, E/RT_w. The advantage of such an approach is clear, one can trivially rederive the critical $\Phi(S)$ curve for different reaction orders, feed temperatures, and activation energies. The correlation can be readily incorporated into Eq. 27 and thus eliminate the use of Figure 2.

The accuracy of the two approaches (Barkelew's look-up table and the correlation in Eq. 28) are examined against experimental data in the literature for vinyl-acetate synthesis (Emig et al., 1980) and for a university methanation reactor (Webb, 1989). In Figure 3, we plot D/D_{max} vs. $[\Delta T_{ad}(1.0)]/\Theta$, where D_{max} is taken from Eq. 27.

Four sets of data are plotted in the diagram:

- One datum point for the Caltech methanation reactor, not parametrically sensitive
- Data for Emig's vinyl-acetate synthesis reactor, which displayed temperature sensitivity
- Data for Emig's vinyl-acetate synthesis reactor, which did not display temperature sensitivity
- One datum point for an industrial vinyl-acetate synthesis reactor, not parametrically sensitive.

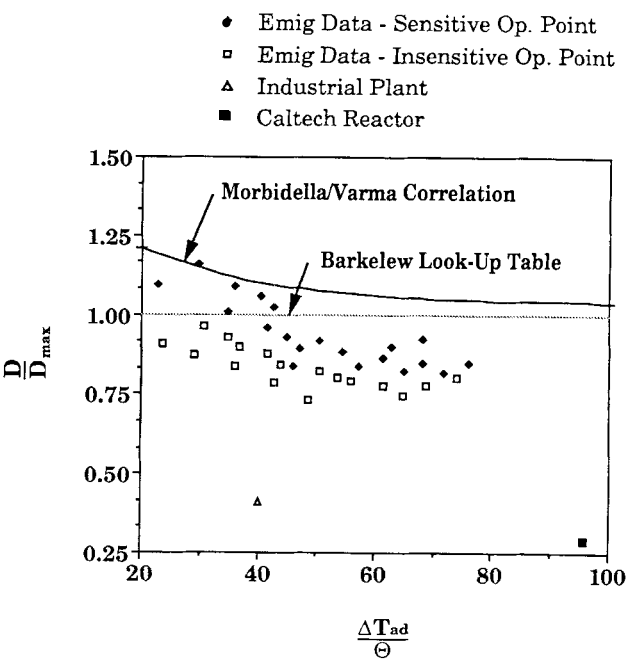


Figure 3. Verification of sensitivity criterion (data from Emig et al., 1980; Webb, 1989).

In addition, two loci are shown:

- The design criterion in Eq. 27 using Barkelew's look-up for Φ
- The design criterion in Eq. 27 using the Morbidelli/Varma correlation in Eq. 28 for Φ.

It can be seen that the agreement is quite good between the predicted regions for temperature sensitivity and the experimental data. The agreement for the Φ correlation from Eq. 28 is better qualitatively than the one based on Barkelew's look-up for Φ, although the former is slightly more conservative. The quantitative error could be attributed to the fact that the true order for the vinyl-acetate synthesis has been reported to lie between zero and one. The utility of the correlation in Eq. 28 is that such changes in reaction order (or activation energy or inlet temperature) can be readily incorporated in the computation of Φ. As before, a safety factor of 2 should be incorporated ($D=0.5D_{max}$).

Finally, it should be pointed out that both the Caltech methanation reactor and the industrial vinyl-acetate synthesis reactor have been properly designed (or even overdesigned) and exhibit no temperature sensitivity.

More involved calculations are possible for $\Phi(S)$, depending on the desired complexity. For instance, the same criterion (Eq. 27) can be used with a Φ which has been modified to incorporate particle effects. The resulting criterion can be used as in Morbidelli and Varma (1986b) to predict the occurrence of multiple steady states in the reactor. The drawback of this

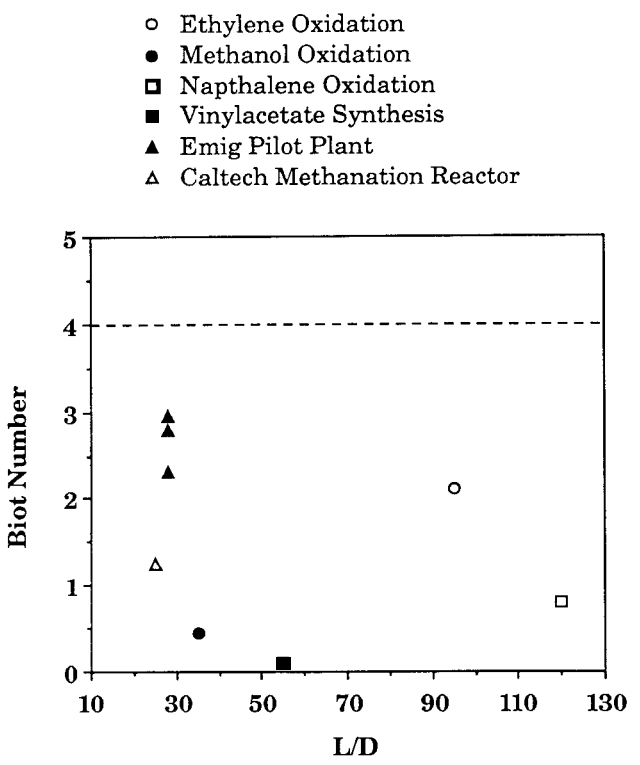


Figure 4. Verification of Biot number specification (data from Emig et al., 1980; Hlavacek, 1970; Webb, 1989).

latter approach is that it is more computationally intensive than even the Barkelew calculations.

The criterion in Eq. 27 has been derived by assuming a simple homogeneous model with only axial heat conduction. A mean heat-transfer coefficient can be calculated for such a model by considering the sum of the resistances to heat transfer in the bed and at the wall (as in Froment and Bischoff, 1979):

$$\frac{1}{h}=\frac{1}{h_w}+\frac{D}{8k_e} \tag{31}$$

where k_e is the radial conductivity and h_w is the heat-transfer coefficient between the inside reactor wall and the cooling jacket. A quadratic temperature profile has been assumed. The critical assumption in one-dimensional models for parametric sensitivity calculations is that the first term on the righthand side in the Eq. 31 is larger than the second term. In other words, one assumes that the resistance to heat conduction from the inside reactor wall to the cooling jacket is much larger than the internal resistance to radial heat conduction in the bed.

$$\frac{h_w D}{2k_e}=Bi<4 \tag{32}$$

where Bi is the dimensionless Biot number. Some typical values for the Biot number are plotted in Figure 4 for several typical industrial and academic laboratory reactors (Emig et al., 1980; Hlavacek, 1970; Webb, 1989).

It is clear that the radial profiles are fairly mild for the presented industrial data and the Caltech reactor. However, in the case of the data from Emig et al. (1980) in which they were attempting to induce parametric sensitivity, the radial profiles were very steep and suggest that radial effects play a significant role in the critically stable behavior observed in this case.

In light of these observations, we recommend that the requirement on the Biot number (<4) serve as a verification that the parametric sensitivity analysis is correct. Violation of this specification suggests the need for a more involved sensitivity criterion which incorporates radial effects.

Adjustments to criteria

In the preceding analysis, it has been assumed that there is no recycle or dilution; however, it is straightforward to account for these effects. In general, the recycled stream is the product of a separation process and contains unconverted feed components and to a smaller extent intermediates and reactor products. If the recycle is wholly unconverted feed components, then it has no impact on Eqs. 14 and 27 as long as one remembers that v_s is based on total feed to the reactor. v_s is often given in terms of fresh feed only, which is designated as v_s^0. In this case, v_s has to be adjusted accordingly

$$v_s=v_s^0(1+R) \tag{33}$$

where R is mass recycle per mass feed. If the recycle contains a diluent, then this affects ΔT_{ad}. This requires iteration, but it is straightforward as long as we always use ΔT_{ad} and v_s based on total feed in Eqs. 14 and 27.

There is another practical caveat. Most catalytic reactions are neither first nor second order due to the influence of adsorption rates on the overall rate law. If they are pseudo-first-order (first order in mole fraction, with a more complex dependence on total pressure, Wei and Prater, 1962), one can still use a first order approximation. It is, however, safer to directly measure r_{max} and the apparent activation energy in the pilot plant. As both measurements are affected by mass transfer to the catalyst particle and inside it, it is preferable to confirm the estimates in a pilot plant using a single tube with the same length as the final multitubular reactor.

Acceptable pressure drop

The foregoing calculations are only concerned with the stability of the reactor and its sensitivity to disturbances. There is another important consideration, namely limiting the pressure drop to an acceptable value.

For turbulent flow, which is a reasonable assumption for most tubular reactors, the total pressure drop is given by the Ergun Equation (Froment and Bischoff, 1979)

$$\Delta P=L\frac{\alpha}{D_p}\frac{G^2}{\rho}=\frac{v_s^2 L^3}{\rho}\frac{\alpha}{D_p} \tag{34}$$

where the friction factor, α, is equal to:

$$\alpha=\frac{(1-\epsilon)}{\epsilon^3}\left[\frac{150(1-\epsilon)}{Re_p}+1.75\right] \tag{35}$$

α can be directly measured in a pilot plant and experimental values are preferred.

The friction factor correlation in Eq. 35 holds for spherical catalyst particles with $d_p/D<5$.

Design procedure

Summarizing the results of the previous sections, we have the following specifications on the reactor dimensions:

$$L>L_{\min}=5.5D\frac{\Delta T_{ad}(x)}{\Theta}\left[\frac{-\ln(1-x)}{x}\right] \tag{36}$$

$$D<D_{\max}=\frac{1}{\Phi\left(\frac{\Delta T_{ad}(1.0)}{\Theta}\right)}\frac{4}{v_s[-\ln(1-x)]}\frac{h\Theta}{C_p\Delta T_{ad}(1.0)} \tag{37}$$

$$L<L_{\max}=\sqrt[3]{\frac{\Delta P_{\max}\rho D_p}{\alpha v_s^2}} \tag{38}$$

The assumptions used in the derivation of these guidelines are as follows:

1. $Pe=11$
2. First-order, irreversible, isothermal reaction
3. Spherical catalyst pellets.

Equations 36 and 37 give a reasonable first estimate for the physical dimensions of the reactor in terms of the parameters which are convenient for design purposes. The temperature scaling parameter Θ depends on the activation energy and is thus determined by the nature of the reaction and the catalyst. The parameters which can be easily adjusted include ΔT_{ad}, v_s, L, and D. ΔT_{ad} can be lowered by adding a diluent to the feed. v_s is normally adjusted in coordination with the catalyst activity. Catalyst activity can often be adjusted using an inert zeolite matrix, in which the matrix acts as a binder. If the activity is lowered, the maximum reaction rate is lowered which requires a higher residence time in the reactor. The original conversion can be maintained by lowering v_s. A related idea is to profile the catalyst activity in a controlled manner to avoid thermal runaway and the subsequent (poorly controlled) catalyst deactivation. It has been demonstrated (Pirkle and Wachs, 1987) that if less active catalyst is employed in the region of the reactor where the reaction driving force is greatest, then the parametric sensitivity of the bed is greatly reduced. In practice, this selective deactivation may occur unintentionally and lead to reduced sensitivity in a bed which may have originally been highly sensitive.

In terms of the diameter of the reactor, Eqs. 36 and 37 indicate that a smaller diameter yields better thermal stability in the bed. However, from an economic perspective, it is cheaper to produce tubes with larger diameters. Economic considerations also favor lower dilution rates and higher space velocities. Therefore, solving for an economically optimal set of conditions which also satisfy Eqs. 36 and 37 requires an iterative procedure. For instance, for fixed D, a reduction in v_s is traded off (via Eq. 37) against less diluent and a higher ΔT_{ad}. Similarly, for fixed ΔT_{ad}, the economic penalty of reducing v_s is traded against an economically favorable larger tube diameter (via. Eq. 37).

Let us now outline an iterative design procedure based on these equations. Consider an undiluted feed and fixed conversion. This fixes ΔT_{ad} and we get L/D from Eq. 36. Turning to Eq. 37, we can adjust v_s to get an acceptable diameter. These two results yield a reactor length, which can be verified against the acceptable pressure drop in Eq. 38. If at the desired condition, both Eqs. 36 and 38 give satisfactory answers, then there is no problem. If either L or ΔP are not acceptable, we have to continue our iteration as long as Eq. 36 conflicts with Eq. 38.

Note that we have a separate criterion for $L_{\min}$ and for $L_{\max}$. Each of these contain both independent and joint parameters. Since $L_{\min}$ increases with D, and $L_{\max}$ is independent of D, we can change their ratio by adjusting D. A reasonable guideline requires that $L_{\max}$ exceed $L_{\min}$ by a factor of 2. Otherwise, we have to modify other parameters. These modifications invariably affect the cost. For example, we can reduce $L_{\min}$ by lowering x (equivalently, increasing L^*). This is achieved at the expense of heat losses in recycling the unconverted feed.

Another way to modify the conditions is by diluting the inlet stream or by recycling the product stream as was discussed earlier. Assume we do it by keeping partial pressure constant. If the amount of diluent added per gram fresh feed is R, and C_p is constant, then:

$$\Delta T_{ad}=\frac{\Delta T_{ad}^0}{1+R} \tag{39}$$

$L_{\min}$ in Eq. 36 will decrease in proportion to $1+R$, but $D_{\max}$ in Eq. 37 will not be affected as $\Delta T_{ad}(1.0)v_s$ is independent of $1+R$. If we increase the pressure to keep partial pressure constant, then criterion 38 has the following dependence on R:

$$\frac{\rho}{v_s^2}=\frac{\rho^0(1+R)}{(v_s^0)^2(1+R)^2} \tag{40}$$

and $L_{\max}$ is proportional to $1/^3\sqrt{1+R}$. Thus:

$$\frac{L_{\max}}{L_{\min}}=(1+R)^{2/3} \tag{41}$$

If we keep total pressure constant

$$\frac{\rho}{v_s^2}=\frac{\rho^0}{(v_s^0)^2(1+R)^2} \tag{42}$$

and $L_{\max}$ is proportional to $1/^3\sqrt{(1+R)^2}$. In this case:

$$\frac{L_{\max}}{L_{\min}}=(1+R)^{1/3} \tag{43}$$

Therefore, in both cases, increasing R with a recycle unit will improve the design feasibility by increasing the ratio of $L_{\max}$ to $L_{\min}$.

In many cases, the pressure drop for a reactor of 40 ft (12 m) or even 60 ft (18 m) is acceptable. Thus, the pressure drop constraint applies only to reactors with very high flow rates, such as in the case of high recycle rates. There are options to reduce pressure drop by choosing catalyst shapes with inherently lower pressure drops (Froment and Bischoff, 1979).

Equation 35 gives the friction factor for spherical catalyst particles. It is possible to select a differently shaped particle with better pressure drop properties but with nearly equivalent thermal properties (for example, ring shaped catalyst pellets).

We should also note that the care one has to apply to keep the reactor temperature uniform varies from case to case (that is, the penalty for large temperature excursions is different for each case). Higher temperatures may deactivate the catalyst, may cause side reactions, and may cause metallurgical problems. But in most cases it is advisable to look for conditions where temperature variations are small. This is discussed in more detail in Eigenberger (1985), Eigenberger and Ruppel (1986), and Eigenberger and Schuler (1989).

At last we should point out that, in the vast majority of cases, the design modifications required to achieve a very robust design with modest data are not expensive. Building a longer reactor (within the limits mentioned) has typically a very small impact on cost. If one has to severely limit productivity, this is an obvious penalty. But in the experience of one of the authors (Reuel Shinnar), this happens because the designer has not fully taken into account all the measures that ensure robust performance.

Practical considerations

There are some practical trade-offs involved in sizing both L and D. Normally, the smallest tube diameter for large scale reactors is one inch. Smaller diameter tubes would be prohibitively expensive to manufacture. There are limitations placed on L by both shipping and transportation as well as the physical support of the tubes in the reactor. In addition, the tolerable pressure drop across the reactor limits the acceptable length (as was shown in the previous section). On the other hand, it is seldom economical to have a tube length smaller than 40 ft (12 m), unless the catalyst volume required is very small. A larger L will give a larger maximum tube diameter and fewer tubes. For very large reactors, this can be achieved by connecting two reactors in series.

There is another practical aspect of the sensitivity criteria which is often overlooked. The criteria in the previous section relate to fixed operating conditions. In practice we have two deviations related to flow variations and maldistributions.

First, operating conditions rarely remain fixed at their designed settings. A reactor has to be able to operate at lower flow rates or throughputs than originally designed for. It will also be operated at higher throughputs, but there is often objection to provide for this in the design. An experienced designer does so but hides it in safety coefficients.

Second, the flow rates vary from tube to tube. We therefore have to look at the effect of variations in throughput on our criteria. If the reaction is not mass-transfer-controlled, r_{max} is independent of linear velocity and only depends on inlet composition and reactor temperature. Flow rate will impact the heat-transfer coefficient in Eq. 27 and the value of the mass-flow rate G in Eq. 7. In both cases, thermal sensitivity is reduced by increasing the flow rate. Therefore, the criteria must be evaluated at the lower bound on flow rates through the tubes. In this respect, Eq. 7 is more sensitive than Eq. 27, as the criterion in Eq. 7 is inversely proportional to flow rate, whereas in Eq. 27 it is inversely proportional to a fractional power of the flow rate (typically $h = h_0 G^{\lambda}$ where $\lambda = 0.5 - 0.8$). If the criteria are conservative enough, they will protect against thermal sensitivity arising from flow variations.

In general, if the throughput is reduced, r_{max} should be reduced accordingly.

Control Issues for Tubular Reactor Design

The sensitivity criterion (Eq. 27) described earlier is appropriate for open-loop reactor operations. However, the application of a control law will in general significantly alter the dynamic properties of the system. This is the case for a simple CSTR in which open-loop bifurcations and pathologies are effectively eliminated by simple proportional control (Aris and Amundson, 1958). Similarly, theoretical studies suggest that accurate nonlinear model-based control can lead to safe operation of packed bed reactors near regions of parametric sensitivity (Seider et al., 1990). However, as we will see in this section, there are certain complicating features of an industrial packed-bed reactor which do not allow even an advanced control scheme to eradicate the sensitive open-loop behavior. We will focus on the particular control problems which arise when we try to operate the reactor near the critical values given in the design criteria.

Controlled operation of a multitubular packed-bed reactor is complicated by the temporal and spatial variations inherent in a distributed parameter system. Additional difficulty is caused by flow variations between individual tubes. The combination of these effects pose a formidable task in the placement of sensors for measuring reactor conditions. An additional complication in control design is introduced by the presence of large and often uncertain time delays in the system.

These effects are considered in more detail in the subsequent analysis. In particular, their impact upon the proposed design criteria is investigated. In the final sections, a reduced order nonlinear model of a tubular reactor is presented for simulation purposes. The resultant case studies demonstrate the controllability problems which can arise when the reactor design criteria are violated.

Multitube flow variations

The flow dynamics of a multitubular reactor will, in general, be quite complicated. However, it is reasonably straightforward to show that maldistributions in the bulk feed flow can cause significant variations in the flow rates between the various tubes. In addition, it is likely that the packing in the tubes will be far from homogeneous, leading to varying resistances to flow in the individual tubes. A value of $\pm 15\%$ has been reported in the literature recently (McGreavy and Maciel, 1989), and will be used in the subsequent analysis to predict the peak variation from tube to tube. In some cases, all tubes are individually tested after filling, which can reduce variability of flow to $\pm 10\%$.

Consider again the second criterion (Eq. 27) for temperature sensitivity. In the sensitivity diagram given in Figure 2, the ordinate (N/S) depends on the flow rate via the heat-transfer coefficient between the wall and the bed (Eq. 26). This heat transfer is generally dominated by resistance inside the tube and a reasonable empirical correlation (for high Re) is given in Froment and Bischoff (1979):

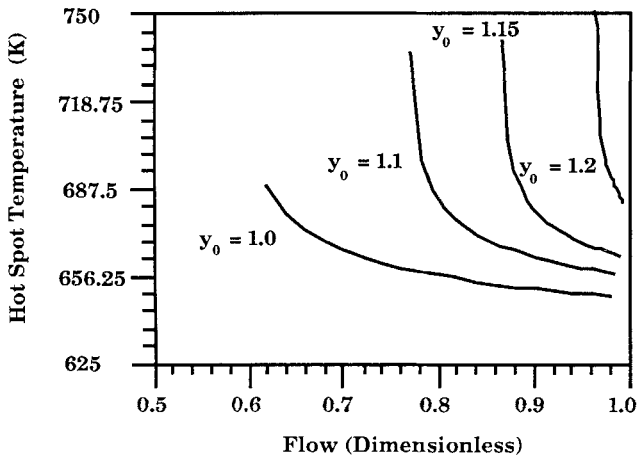


Figure 5. Steady-state dependence on hot-spot temperature on flow rate and inlet concentration (y_0).

$$h = 3.50\left(\frac{k_e}{D}\right)\left(\frac{d_p G}{\mu}\right)^{0.7} e^{-4.6 d_p/D} \tag{44}$$

This expression gives a 0.7 power law dependence of h on flow rate (G). Consequently, flow variations in the tubes on the order of 30% will give rise to N/S values which differ by as much as 20%. From Figure 2, it is clear that if our margin of safety in the reactor design is less than 20% of N/S, this flow perturbation will lead to a crossing of the sensitivity curve [$\Phi(S)$]. Consequently, if the bulk of the tubes are at a higher flow rate (large N/S) and lower temperature, then the tubes with restricted flow (small N/S) may "ignite" and undergo thermal runaway.

The effect of this change in flow rate on the hot spot temperature is depicted in Figure 5. Plotted here are the values of the hot spot temperature for a condition of high parametric sensitivity (Van Welsenaere and Froment, 1970). The particular values of the physical parameters used here will be introduced with the closed-loop simulations and are given in Table 3. It can be seen in the diagram that as the flow rate is reduced, there is poorer heat transfer and, consequently, the hot spot temperature increases. The condition in Figure 5 corresponding to an extremely steep slope (for fixed inlet concentration and flow rate) corresponds to a point along the sensitivity curve in Figure 2 (incipient runaway). As expected, higher inlet concentration (y_0) leads to greater sensitivity as the reaction rate accelerates.

Measurement set

For practical reasons, it is often necessary to use secondary measurements in the control of a tubular packed-bed reactor to improve the performance of the system. For instance, temperature sensors located axially along the reactor can be used to "infer" the values of the other reactor variables. For multitubular reactors, one is not only concerned with a reasonable number of accurate measurements along the length of the tube, but one must collect measurements from enough tubes to assure an accurate measurement of the true bulk temperature. There will be between 1,000 to 5,000 tubes in a multitubular reactor (Froment and Bischoff, 1979), but for control purposes, we want our measurement set to be much smaller. The practical implications are immediately clear: it will be impossible to monitor every tube in the reactor bundle. As was indicated earlier, this can be very dangerous when one operates near the cutoffs given for parametric sensitivity. There may be a few tubes which are not measured, have reduced flow rates, and consequently, ignite without being observed. Even if a measurement is made of a bulk property, such as the effluent temperature of the tubes, the diluted effect of the ignited behavior may still go unnoticed.

Nonminimum-phase characteristics

An additional problem in packed-bed reactor control is the presence of large, often poorly characterized, time delays in the system. Their causes include propagation rates in the bed, finite time required for actuator movement, and lags in measurement devices (for example, composition analyzers). The presence of these time delays will have a detrimental impact on the closed-loop performance. As will be demonstrated in the subsequent case studies, there is a *dynamic sensitivity* associated with operation in regions of high parametric sensitivity. This also places certain limitations on the achievable performance of multitubular reactors operating in these regions. The combined effect of these two limitations can lead to unacceptable closed-loop behavior, such as large temperature transients.

Additional nonminimum-phase (NMP) characteristics are associated with certain choices of the manipulated variable for reactor control. These include the inlet flow rate and inlet temperature. The selection of these variables for control inputs leads to right half plane zeros which are manifested in inverse-response behavior. This places severe limitations on the achievable system performance. It also precludes the application of advanced nonlinear techniques like input/output feedback linearization (Kravaris and Kantor, 1990). In the subsequent case studies, the performance of a flow controlled reactor (NMP) and a jacket temperature controlled reactor (MP) will be investigated for conditions of both high and low parametric sensitivity.

Case Studies

Nonlinear reduced-order model

For simulation purposes, a simple pseudohomogeneous model is employed where all heat- and mass-transfer resistances between fluid and catalyst phase are neglected. In dimensionless form (see Table 1), the mass and heat balances are given by:

$$\frac{\partial y}{\partial t'} + \frac{\partial y}{\partial z'} = -\mathcal{D}a\, r(x,y) \tag{45}$$

$$\frac{\partial x}{\partial t'} + \frac{\partial x}{\partial z'} = \mathbf{B}(x - x_w) - q\,\mathcal{D}a\, r(x,y) \tag{46}$$

$$y(0,t) = y_0 \quad x(0,t) = x_0 \quad \left(\frac{\partial x}{\partial z'}\right)_{z'=1} = 0 \tag{47}$$

where x is the dimensionless temperature and y is the dimensionless concentration.

Table 1. Dimensionless Variables for Reactor Model

z'	$\frac{z}{L}$
y	$\frac{c}{c_{ref}}$
x	$\frac{T}{T_{ref}}$
x_w	$\frac{T_w}{T_{ref}}$
t'	$\frac{t}{t_{ref}}$
c_{ref}	c_0
T_{ref}	T_0
t_{ref}	$\frac{L}{u}\frac{(\rho C_p)^*}{(\rho C_p)}$
$\mathcal{D}a$	$\frac{Lk_0(1-\epsilon)}{u}$
$\mathbf{B}$	$\frac{4hL}{u\rho C_p D}\left(\frac{F}{F_0}\right)^{\lambda}$
q	$\frac{-\Delta H c_0}{\rho C_p T_{ref}}$
γ	$\frac{E}{RT_{ref}}$

In developing a low-order model for this system, it is of critical importance to retain the *essential* nonlinear dynamics. By this, we refer to the relevant dynamics which accurately and succinctly reflect the stability of the reactor and the relationship between the control inputs, outputs, and the reactor's dynamic states. In this manner, a control algorithm based on this model will provide suitable closed-loop performance. The approach adopted in this work is a treatment of transport mechanisms in the reactor as a nonlinear wave which propagates up and down the bed in response to changes in the operating conditions (Marquardt, 1990). The model will be summarized briefly here; the interested reader is referred to Doyle et al. (1990) and Doyle (1991) for the full details of the model derivation.

The reactor bed is divided into two zones: an ignition zone and a reaction zone. Processes in the ignition zone are assumed to occur instantaneously compared to the relatively slower reaction zone. The front between these two zones propagates in response to changes in various reactor conditions. The dynamics of the front are considered to be the *essential* nonlinear dynamics of a packed bed reactor operating near ignition. The position of this wavefront is determined by a critical energy balance. Specifically, this condition requires that the heat removal line for the ignition zone is tangent to the heat production curve. Consequently, this zone is always at the "edge" of ignition. In our simplified model where conduction effects have been neglected, the presence of ignited states is, strictly speaking, not possible. However the calculation of an ignition position is the same, except the two curves do not intersect at the ignition point but have equal slopes. As discussed in Doyle et al. (1990), this approach yields an acceptably accurate low order approximation to the actual reactor behavior.

This treatment leads to a simple second-order nonlinear model for the bed dynamics. The two states are the filtered input and the wavefront position. The former is a fast lag applied to the true manipulated variable, and the latter equation is given by the critical stability condition. Various choices are possible for the single manipulated variable: inlet concentration, inlet temperature, jacket temperature, and inlet flow rate. In the following case studies, we consider closed loop operation of a reactor with flow manipulation, and separately, with jacket temperature manipulation. The logical choice for a controlled output is the peak temperature along the axial profile, the so-called hot spot temperature. As discussed earlier, this will minimize unwanted side reactions and catalyst deactivation which result from thermal runaway. The resultant SISO model structure is:

Table 2. Reactor Reduced Model Variables

x_1	Filtered Input
x_2	Ignition Position
u	Manipulated Variable (Flow or Jacket Temp.)
y	Hot Spot Temperature
w	Wave Propagation Velocity
τ	Lag on Input

Table 3. Physical Parameters for Sensitive Simulations

$\mathcal{D}a$	7.059*E*8
B	11.38
q	1.0
γ	21.82
λ	0.5
$r(x,y)$	$ye^{-\gamma/x}$

$$\begin{aligned} \tau\dot{x}_1 &= -x_1 + u \\ \dot{x}_2 &= w(x_1, x_2) \\ y &= h(x_1, x_2) \end{aligned} \tag{48}$$

The meaning of these variables is summarized in Table 2.

The numerical computations involve iterating on the ignition position until the critical energy balance is satisfied at a given time interval. To accomplish the energy calculations, it is necessary to perform an integration of the axial temperature profile. Once the ignition position has been determined, the wavefront velocity is calculated from the change in ignition position from the previous time interval.

Simulations

For the next four case studies, a set of operating conditions is selected which violates the proposed sensitivity criterion (Eq. 27). In particular, the diameter exceeds the recommended value (Eq. 37) by 50% ($D = 1.5D_{max}$). In this manner, we will investigate the control problems associated with a parametrically sensitive packed bed. The particular choices of physical parameters are shown in Table 3; they correspond to parametrically sensitive behavior as studied by Van Welsenaere and Froment (1970). Note that the reaction under consideration has an irreversible first-order rate law and is exothermic.

Case Study 1 (Steady-State Sensitivity, Nonminimum-Phase System). The first phenomenon studied is the effect of multitubular flow variations. As was described earlier, flow rates from tube to tube will vary by as much as 30%. These variations affect the sensitivity through a power law dependence in the

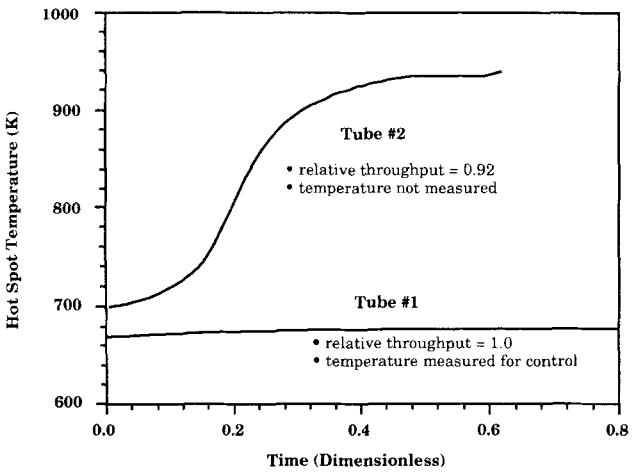


Figure 6. Closed-loop response for two tubes with different throughputs to a set point change in the hot-spot temperature for tube 1 using flow rate as a manipulated variable.

a. Tube 1 (throughput = 1.0); b. tube 2 (throughput = 0.92).

heat-transfer coefficient. For this study, a nominal value of 0.5 is chosen for this coefficient.

In Figures 6a and 6b are shown the responses of the hot-spot temperature for two reactor tubes to a step change in the set point of the bulk hot-spot temperature. The particular simulation conditions are shown in Table 4.

Note that the second tube has a flow restriction and thus has a throughput which is 8% lower than the first tube. The temperature in tube 1 is used as a measurement for a PI controller in an effort to control the hot-spot temperature in the bed. This is accomplished by manipulating the bulk flow rate through the tubes. The control parameters ($K_p = 0.5$, $\tau_I = 0.05$) are selected for a reasonable speed of response. As the figures show, tube 1 behaves as expected with an overdamped second-order response while tube 2 "ignites," reaching temperatures in excess of 900 K. It is clear that the restricted flow through the second tube leads to an unreasonably large excursion in the hot spot temperature. Using the information in Table 4 and Figure 5, it can be seen that the new steady-state position of the second tube is in a region of ignition. Thus, we observe a *steady-state sensitivity* with respect to operating conditions.

Note that the behavior depicted in Figures 6a and 6b is independent of the controller parameters. In fact, the response is independent of the controller employed (nonlinear, IMC, and so on) and all closed-loop systems will exhibit the same steady-state sensitivity for these simulation conditions.

Table 4. Simulation Conditions (Case Study 1)

Study Inlet Concentration (y_0)	1.1	
Flow Rate (F/F_0)	0.85	
Inlet Temperature (x_0)	1.00	
Jacket Temperature (x_w)	1.00	
Manipulated Variable (u)	Flow Rate	
Controlled Variable (y)	Hot-Spot Temperature	
Disturbance	None	
Set Point Change	8 K	
	Tube 1	Tube 2
Relative Throughput	1.0	0.92

Table 5. Simulation Conditions (Case Study 2)

Study Inlet Concentration (y_0)	1.1	
Flow Rate (F/F_0)	0.85	
Inlet Temperature (x_0)	1.00	
Jacket Temperature (x_w)	1.00	
Manipulated Variable (u)	Flow Rate	
Controlled Variable (y)	Hot-Spot Temperature	
Disturbance	+ 10% Step in y_0	
Set Point Change	0 K	
	Tube 1	Tube 2
Relative Throughput	1.0	0.92

Case Study 2 (Dynamic Sensitivity, Nonminimum-Phase System). Further insights are gained if a controlled response to a disturbance in inlet concentration is investigated. The simulation conditions for this example are displayed in Table 5. As before, the control configuration involves the manipulation of bulk flow rate to regulate the hot-spot temperature. Measurements are only available from tube 1 and the controller is a PI controller with settings: $K_p = 0.5$, $\tau_I = 0.05$. Now the controlled system is subjected to a 10% step increase in the inlet concentration. From the steady-state values at the end of the simulations in Figures 7a and 7b, we can see that both tubes have initial and final points which may be considered tolerable for the designed reactor.

However, a large transient excursion of the temperature in tube 2 is observed for the disturbance in concentration. The peak hot-spot temperature observed is in excess of 150 K above the final steady-state level. Such phenomenon could be accurately described as *dynamic sensitivity*.

In this case study, the controlled response is strongly dependent on the controller parameters. As the sensitivity exhibited in this case study is dynamic in nature, we can expect to minimize the size of the temperature excursion by optimizing the control parameters. Note, however, that the magnitude of the flow restriction also strongly affects this transient response.

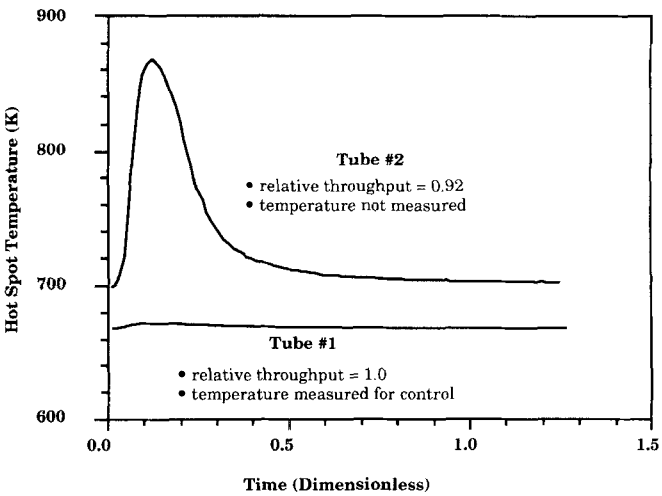


Figure 7. Closed-loop response for two tubes with different throughputs to a step disturbance in the inlet concentration in both tubes using flow rate as a manipulated variable.

a. Tube 1 (throughput = 1.0); b. tube 2 (throughput = 0.92).

Table 6. Simulation Conditions (Case Study 3)

Inlet Concentration (y_0)	1.1	
Flow Rate (F/F_0)	1.0	
Inlet Temperature (x_0)	1.0	
Jacket Temperature (x_w)	1.0	
Manipulated Variable (u)	Jacket Temperature	
Controlled Variable (y)	Hot-Spot Temperature	
Disturbance	-10% Step in F/F_0	
Set Point Change	0 K	
	Tube 1	Tube 2
Relative Throughput	1.0	0.8

Case Study 3 (Dynamic Sensitivity, Minimum-Phase System). The same sensitivities are evident in the closed-loop for a *minimum-phase* system. The control configuration is as before, with the replacement of bulk flow by jacket temperature as the manipulated variable. And as before, we only measure the temperature in tube 1 in an effort to control the hot-spot temperature in the reactor. This effectively renders tube 2 off-line and no control action will be taken to handle disturbances in this tube. The control settings are as before ($K_p=0.5$, $\tau_I=0.05$). The remaining conditions for the simulation are shown in Table 6. In this case, we consider a disturbance of a 10% step decrease in the flow rate for two tubes. The tubes chosen for this study differ by 20% in their flow throughput. The controlled response to this disturbance is depicted in Figure 8.

Again, it is observed that there is a *dynamic sensitivity* associated with this operating condition as tube 2 experiences a 300 K overshoot in its response.

As in case study 2, the size of this dynamic temperature excursion is a strong function of the control law and the magnitude of the flow restriction.

Remark on Case Studies 1, 2 and 3. As has been emphasized in the above three case studies, it is implicitly assumed that the flow-restricted tube (#2) represented an unobserved tube from the perspective of sensor measurement. Thus, it is effectively rendered off-line and no control action is taken to mitigate the effect of the disturbances.

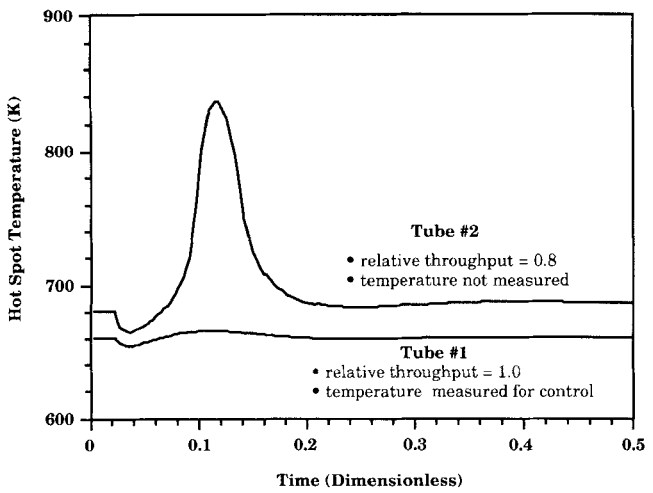


Figure 8. Dynamic sensitivity.

Closed-loop response for two tubes with different throughputs to a step disturbance in the flow rate in both tubes using jacket temperature as a manipulated variable.

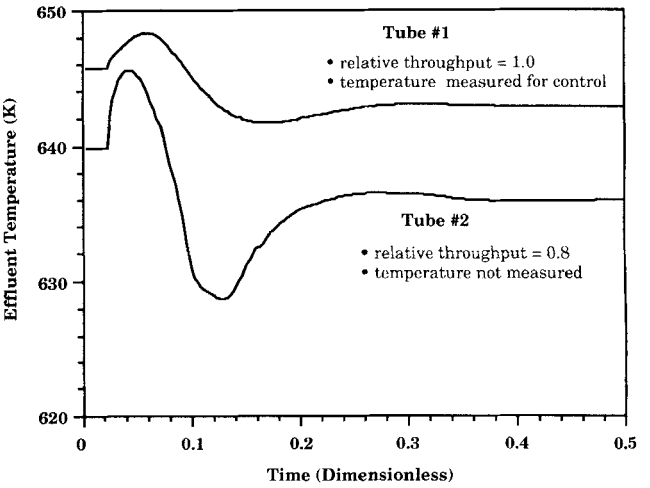


Figure 9. Dynamic sensitivity.

Closed-loop response for two tubes with different throughputs to a step disturbance in the flow rate in both tubes using jacket temperature as a manipulated variable.

Case Study 4 (Measurement Selection, Minimum-Phase System). An alternative sensor placement set might include some bulk measurements, such as the reactor effluent temperature. However, one can envision a reasonably representative flow distribution where tube 1 represents 99% of the tubes and 1% of the tubes are restricted to the flow levels given by tube 2. Recall the conditions in case study 3, and now consider the effect of the disturbance on the effluent streams from the two tubes. These results are shown in Figure 9.

It is clear that the large hot-spot temperature excursion is extremely localized and is only represented by a 15 deg K range in the effluent temperature. Recalling our assumption that tube 1 represents 99% of the total flow, it is clear that the effect of the "hot tube" (#2) would be altogether lost by dilution effects in the mixed effluent stream. In this case, a mixed stream would show a profile which is indistinguishable from the profile exhibited by tube 1.

Case Study 5 (Insensitive Operation, Minimum-Phase System). Case studies 3 and 4 are repeated with a new reactor design in which the diameter is 50% smaller than that recommended by Eq. 27 ($D=0.5D_{max}$). Now we have a reactor which is designed "safely" in accordance with the guidelines prescribed in this article. The physical parameters which lead to this condition are shown in Table 7. As before, we consider the manipulation of jacket temperature (using a PI controller with parameters: $K_p=0.5$, $\tau_I=0.05$) to regulate the hot spot temperature in the bed. We look at two different tubes, one of which has restricted flow rate and is not observed by the controller. Additional conditions for this simulation are shown in Table 8. The results for closed-loop operation are depicted in Figures 10a and 10b.

Table 7. Physical Parameters for Insensitive Simulations

$\mathcal{D}a$	7.059E8
B	34.14
q	1.0
γ	21.82
λ	0.5
$r(x,y)$	$ye^{-\gamma/x}$

Table 8. Simulation Conditions (Case Study 5)

Inlet Concentration (y_0)	1.0	
Flow Rate (F/F_0)	1.0	
Inlet Temperature (x_0)	1.0	
Jacket Temperature (x_w)	1.006	
Manipulated Variable (u)	Jacket Temperature	
Controlled Variable (y)	Hot-Spot Temperature	
Disturbance	-10% Step in F/F_0	
Set Point Change	0 K	
	Tube 1	Tube 2
Relative Throughput	1.0	0.8

We observe the effect of a 10% decrease in the feed-flow rate on the temperatures in the two tubes, which differ by 20% in their throughput. Figure 10a shows the response of the hot-spot temperatures; Figure 10b shows the response of the effluent temperatures. Clearly, the high sensitivity to operating conditions has been diminished, and the difference in temperature levels between the two tubes is negligible.

By designing a reactor which satisfies the proposed sensitivity criterion, we have considerably improved the controllability of the resultant system.

Practical Control of Tubular Reactors

The previous simulations demonstrate that it is advisable to design multitubular reactors such that they are inherently robust and insensitive to perturbations. While it is possible to operate a single CSTR by feedback stabilization, this is not advisable for a multitube reactor. The problem of designing a system such that it is robust and easy to control vs. relying on advanced control methods for stabilization is one that often appears in practice (Shinnar, 1986; Silverstein and Shinnar, 1982). Regretfully, very often the problem of the impact of design on controllability is not sufficiently recognized both in practice and in the literature. If aware of the problem, most users will prefer an inherently robust design over one relying on control.

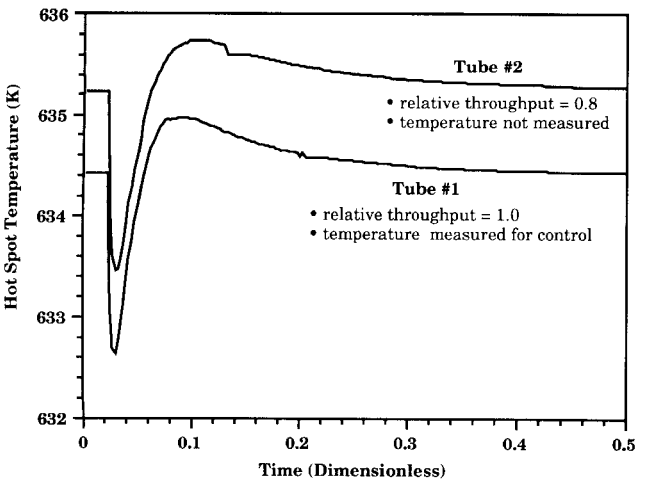
In the aerospace industries, there are cases where design of inherently unstable (or sensitive) systems stabilized by control have demonstrated advantages. One clear example of this is the X-29 high-performance aircraft (swept forward wing fighter, Ames Drysden Flight Research Facility, 1987). The design of this plane makes possible certain high performance combat flight maneuvers; however, its open-loop instability would lead to catastrophic circumstances in milliseconds if the feedback loop is turned off. There are also cases in process industries where such stabilization is essential (Lei et al., 1971; Shinnar, 1986; Silverstein and Shinnar, 1986), although the problem has not received sufficient attention.

For the specific case of the packed bed reactor, the design criteria proposed should allow safe robust design in most cases. Detailed computer modeling may sometimes allow relaxing these criteria provided detailed and reliable kinetic data are available. However, the cost of obtaining such data has to be weighed relative to the potential savings.

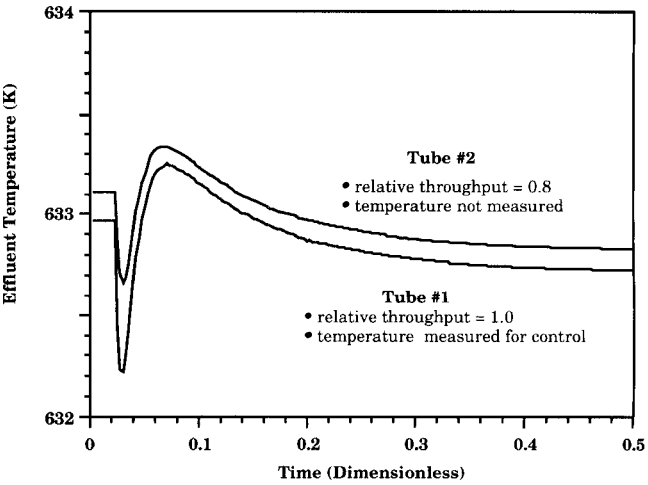
The proposed criteria can also be effectively utilized to update reactor operating conditions in response to a deactivating catalyst. The normal way to do so is to increase the temperature of the cooling bath. Several set of thermocouples along the length of a tube allow the measurement of the maximum temperature difference between the bath and the tube. If this difference is maintained at the level prescribed by the sensitivity criterion (by adjusting cooling bath temperature), then the overall productivity of the reactor can be safely optimized. The problem is sometimes much more complex due to selective poisoning and deactivation of the front section in the reactor. Here, the temperature profiles can be used to diagnose the state of the catalyst. This allows one to predict in advance the need for catalyst regeneration.

Summary

This article presents a set of *simple* and *practical* guidelines for the design of complex catalytic packed bed reactors. The design criteria are derived from requirements on the radial temperature profile, temperature sensitivity, and tolerable



a



b

Figure 10. Insensitive operation.

Closed-loop response for two tubes with different throughputs to a step disturbance in the flow rate in both tubes using jacket temperature as a manipulated variable.

a. Hot-spot temperature; b. Effluent temperature.

pressure drop. The specifications are formulated in terms of the practical reactor parameters of length and diameter, and illustrate the various tradeoffs involved in satisfying the three requirements on the temperature and pressure drop. Thus, the proposed criteria provide direct insights for the practical design engineer.

The stabilization of industrial multitubular packed-bed reactors by feedback control is addressed. It is shown that multitubular variations and the lack of proper measurement signals precludes the dynamic stabilization of the reactor in regions of parametric sensitivity. Thus, stable closed-loop operation can only be accomplished with reactors which adhere to the proposed guidelines. These ideas are demonstrated by closed-loop simulations with a reduced order nonlinear packed-bed reactor model. Operation of a reactor which violates the design criteria is shown to lead to steady state and dynamic sensitivity. This behavior is manifested as thermal runaway in reactor tubes with slightly restricted flow rates. It is also shown that for a reactor designed in accordance with the presented specifications, the behavior is stable, even for tubes with restricted flow rates. Thus, violation of the proposed criteria leads to unacceptable closed-loop performance.

Acknowledgment

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Notation

A = area $[L^2]$
Bi = Biot number $(h_w D/2k_e)$
c = concentration
C_p = specific heat $[L^2/t^2T]$
d = diameter $[L]$
D = tube diameter $[L]$
E = activation energy $[ML^2/t^2]$
F = flow rate $[M/t]$
G = superficial mass flow velocity $[M/L^2t]$
h = overall heat-transfer coefficient $[M/t^3T]$
H = specific enthalpy $[L^2/t^2]$
k_e = radial thermal conductivity $[(ML)/(t^3T)]$
k_{eff} = effective thermal conductivity $[(ML)/(t^3T)]$
k_0 = frequency factor
L = reactor length $[L]$
n = reaction order
N = $(4he^{E/RT_w})/DC_pc_0k_0)$
P = pressure $[M/Lt^2]$
Pr = Prandtl number $[(C_p/\mu)/k]$
r = reaction rate $[M/(tL^3)]$
R = gas constant or dilution/recycle factor
Re = Reynolds number $[(\rho VD)/\mu]$
S = $(\Delta T_{\text{ad}})/\Theta$
t = time $[t]$
T = temperature $[T]$
u = manipulated variable
v_s = space velocity $[M/(L^3t)]$
V = volume $[L^3]$
x = fractional conversion or dimensionless temperature
y = system output or dimensionless concentration

Greek letters

α = friction factor
Δ = difference
ϵ = fractional void space
γ = $E/(rT_w)$
λ = flow coefficient for heat transfer
μ = viscosity $[M/Lt]$
ψ = catalyst particle shape factor
ρ = density $[M/L^3]$
Θ = $[(RT_w^2)/E]$ $[T]$
τ = residence time, time lag [t]

Subscript, superscripts

$*$ = solid
0 = initial nominal
ad = adiabatic
ave = average
b = bulk
eff = effective
eq = equilibrium
f = fluid
max = maximum
min = minimum
p = particle
r = radial
w = wall

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