TABLE 4.1 Reactor Operating Parameters for Open-Loop Oscillations

Heat capacity of feed	2700 J/kg·K
Heat capacity of product	2400 J/kg·K
Density	650 kg/m^3
Feed temperature	303 K
Normalized activation energy (E_a/R)	11,000 K
Heat of reaction	−159,000 J/mol
Mass of metal wall in reactor	10,000 kg
Heat capacity of metal wall	460 J/kg·K
Heat losses	175,000 W
Steady-state temperature	182°C
Coolant temperature	163 → 170°C
Feed flowrate	3.2 kg/s
Feed concentration	7.81 mol/kg
Pre-exponential factor	9.7×10^7 s^{-1}
Heat transfer parameter (UA)	98,000 W/K
Residence time	17.7 min
Effective liquid volume	5.2 m^3

We show the reactor and jacket temperatures (T and T_c) along with C_A, the concentration of component A in the reactor. Initially, when the simulation started, the heat transfer area was sufficiently large to maintain open-loop static and dynamic stability. However, a few minutes into the simulation, we reduced UA by 20 percent. This creates dynamic instability with complex eigenvalues as in Eq. (4.25). The reactor temperature and composition start oscillating with a growing amplitude. However, the amplitude growth stops roughly 5 hours after the onset of instability and the reaction enters into a *limit cycle* of constant period and amplitude.

This type of limit cycle was actually observed in the industrial reactor described by Vleeschhouwer et al. (1992). It was also shown by these authors that stability could be regained by altering the operating point. We confirmed this in our simulation by raising the jacket temperature after roughly 7 hours of process time (see Fig. 4.9). The reactor temperature transitions to nearly 200°C where the limit cycle stops. The high temperature may not be desirable from a safety or product quality standpoint, so we reduced the jacket temperature after the reactor had stabilized. The limit cycle started up again almost instantly. We finally reduced the jacket temperature to about 155°C. The oscillations stopped but the exit concentration might have been too high for the refining train to handle.

The operational difficulties described around the CSTR could be avoided altogether by making the heat transfer area in Fig. 4.8 sufficiently large. This is easy to do when the plant is being designed but may be a costly proposition once the process is in operation. That is

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4.5.3 Parametric sensitivity

Open-loop multiplicity, instability, and oscillations are restricted to reactors with material or energy feedback. It is tempting then to think that plug-flow reactors, lacking backmixing, should be problem-free. This is far from the case. Ironically, it is the lack of backmixing that causes some of the problems. For example, while a CSTR can operate adiabatically at a reasonable temperature and conversion given a low feed temperature, a plug-flow reactor requires a minimum feed temperature to get the reactions started at the inlet of the reactor. In addition, from this point on every additional unit of heat produced along the adiabatic plug-flow reactor is locally added to what has already been produced upstream. There is no temperature averaging over the length of the reactor. This introduces a climbing temperature profile as long as there are additional reactants to convert. Since the rate of reaction increases exponentially with temperature, there is always a possibility that the reaction will go to completion, which may lead to unacceptably high temperatures. The adiabatic temperature rise ΔT_{ad} and the maximum reactor temperature $T_{\max}$ are important parameters for plug-flow systems. The adiabatic temperature rise can be estimated from the reaction heat available in the feed in relation to its capacity to absorb this heat. The maximum temperature is then the feed temperature plus the adiabatic temperature rise:

$$T_{\max} = T_0 + \Delta T_{ad} \approx T_0 + \frac{y_{A0}(-\Delta H)}{C_{P0}^m} \tag{4.26}$$

where T_0 = feed temperature
y_{A0} = mole fraction of limiting reactant in feed
ΔH = heat of reaction
C_{P0}^m = average molar heat capacity of feed

We illustrate the use of Eq. (4.26) by calculating the maximum achievable temperature in the HDA and vinyl acetate reactors discussed in this book. Both of these are gas-phase, plug-flow systems.

Starting with the HDA reactor, we find most of the needed information in Chap. 10. The feed temperature is 1150°F, the heat of reaction −21,500 Btu/lb · mol, and the mole fraction of toluene (limiting component) in the reactor feed is 0.0856. The molar heat capacity of the feed is computed from its composition and standard literature data:

$$C_{P0}^m = 0.43(7.2) + 0.48(16) + 0.09(59) = 16 \text{ Btu/lb} \cdot \text{mol} \cdot {}^\circ\text{F}$$

The estimated maximum temperature is

$$T_{\max} \approx 1150 + \frac{(0.0856)(21{,}500)}{16} = 1265°\text{F} = 685°\text{C}$$

The adiabatic temperature rise for this system is roughly $\Delta T_{\text{ad}} = 115°\text{F} = 64°\text{C}$.

The vinyl acetate process is described in Chap. 11. The reactor inlet temperature is 148.5°C and oxygen is the limiting reactant ($y_{A0} = 0.075$). The heat of reaction is −42.1 kcal/mol vinyl acetate or −84.2 kcal/mol oxygen. The average heat capacity of the feed is computed from data provided in Chap. 11:

$$C^m_{P0} = 0.59(13.3) + 0.22(14.2) + 0.11(37.5) + 0.08(7.5)$$
$$= 15.7 \text{ kcal/kmol °C}$$

When we consider the main reaction and exclude the side reaction, the estimated maximum temperature is

$$T_{\max} \approx 148.5 + \frac{(0.075)(84{,}200)}{15.7} = 551°\text{C}$$

and the adiabatic temperature rise, $\Delta T_{\text{ad}} = 402°\text{C}$.

The maximum temperature in the HDA reactor is greater than it is in the vinyl acetate reactor. Yet, only the HDA reactor can be operated adiabatically; the vinyl acetate reactor must be cooled. There are two reasons for this. The first reason is the magnitude of the adiabatic temperature rise. There is virtually no reaction system that stays selective over a 400°C temperature range. For example, the side reaction in the vinyl acetate reactor becomes significant over 200°C and dominates completely at 500°C. In addition, the catalyst in the vinyl acetate reactor would quickly deactivate over 200°C due to sintering. The HDA reactor, on the other hand, has no catalyst and the adiabatic temperature rise is under 100°C, which is a reasonable range for a reaction to remain selective.

The second reason why the vinyl acetate reactor must be cooled is sensitivity. Sensitivity S is a measure of the reaction's potential to run away from the feed temperature. This tendency is determined by two factors: the relative increase in reaction rate with temperature [Eq. (4.7)] and the feed's potential to elevate the reactor temperature, ΔT_{ad}:

$$S = \left(\frac{dr/r}{dT}\right)_{T_0} \Delta T_{\text{ad}} = \frac{E_a}{RT_0^2} \cdot \frac{y_{A0}(-\Delta H)}{C^m_{P0}}$$

The sensitivity for the HDA reactor is 10.8, which means that the reactor exit stream could react at a rate roughly 11 times faster than the feed provided the compositions remained constant throughout the reactor. This kind of amplification in rate due to temperature indicates that there is a good chance the reaction will go to completion, thereby inducing the full adiabatic temperature rise.

The vinyl acetate reaction is even more sensitive. With $S = 44.2$ for the side reaction alone it would be virtually impossible to try to prevent an adiabatic reactor from reaching full conversion on oxygen by mere control of the inlet temperature. Small changes in the inlet conditions would quickly amplify down the reactor, forcing complete conversion, adiabatic temperature rise, and destruction of the catalyst, if we were lucky in the best scenario. We must apply external cooling to the vinyl acetate reactor.

Given that the vinyl acetate reactor has to be cooled, we now examine the issues around cooled plug-flow systems in general. These systems often attain their maximum temperature before the exit, resulting in a temperature peak or hot spot somewhere along the length of the reactor (see Fig. 4.10). The height of the temperature peak is designed to be well under the adiabatic temperature rise. However, the adiabatic temperature rise is always lurking in the background and cooled reactors can also be quite sensitive to minor changes in the system's operating parameters. These issues were first discussed by Bilous and Amundson (1956) and were later investigated in detail by Froment and Bischoff (1979). A sensitive reactor can easily run away (but also quench) due to minor changes in the feed conditions or the coolant temperature as shown in Fig. 4.10. We emphasize that the runaway and quenched states are not caused by output multiplicity but are distinct steady states related to different, but narrowly separated, input conditions.

From a control perspective, it is important to know if a plug-flow reac-

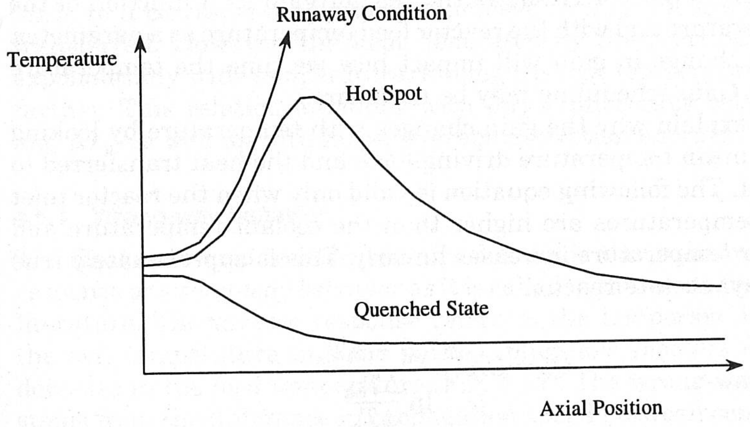

Figure 4.10 Plug-flow reactor temperature sensitivity.

tor has a hot spot that is sensitive to the operating parameters. If this is the case it might be necessary to measure and control the hot-spot temperature directly to prevent runaway. It is of course best to avoid the dynamic control problem altogether and have an insensitive reactor by design. Froment and Bischoff (1979) and Shinnar et al. (1992) discuss various design criteria for making a plug-flow reactor insensitive to parameter variations. In principle it is a matter of providing enough cooling surface per unit reactor volume in relation to the rate of heat generation per unit volume. The required ratio of surface area to heat generated increases as a function of the system sensitivity S.

The vinyl acetate reactor we use in Chap. 11 has been designed to be insensitive to parameter variations under normal operating conditions. The hot-spot temperature is only 162°C with an exit temperature of 159°C. It is adequate to control the exit temperature instead of the hot spot. Since multiplicity, open-loop stability, and sensitivity are of no concern for this reactor, we can focus our attention on the open-loop characteristics relevant to the control of exit temperature with jacket cooling.

Of prime interest for any control loop is the linearity of the process for varying operating conditions. For example, as the catalyst deactivates we have to operate at a higher exit temperature to maintain vinyl acetate production rate. A steady-state model is useful to explore what happens to the process under such conditions. The model helps us make two important observations about the reactor as the exit temperature increases. First, the hot spot moves out of the reactor and the highest temperature occurs at the exit. The reason is that the side reaction, with its high activation energy and high heat of reaction, plays an increasingly greater role at higher temperatures. The second change in reactor character is an increase in process gain. The gain is measured as the ratio of the change in exit temperature to the change in coolant temperature. Figure 4.11 shows the reactor gain as a function of the exit temperature and with the reactor feed temperature as a parameter. The large change in gain will impact how we tune the temperature controller. Gain scheduling may be necessary.

We can explain why the gain changes with temperature by looking at the log mean temperature driving force and the heat transferred to the coolant. The following equation is valid only when the reactor inlet and exit temperatures are higher than the coolant temperature and the reactor temperature increases linearly. This is approximately true for the vinyl acetate reactor.

$$Q_c = UA \frac{\Delta T_{\text{out}} - \Delta T_{\text{in}}}{\ln \dfrac{\Delta T_{\text{out}}}{\Delta T_{\text{in}}}}$$

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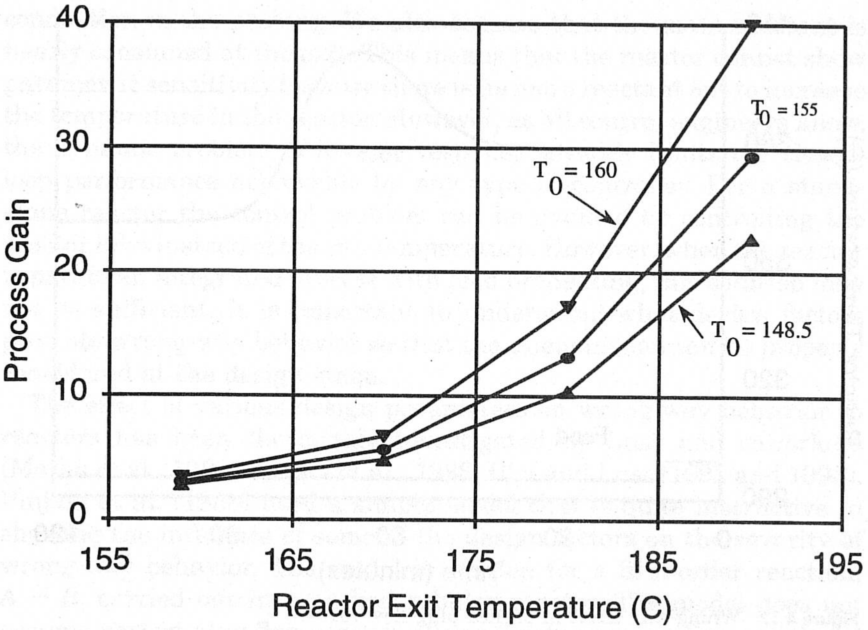

Figure 4.11 Vinyl acetate reactor gain as function of exit temperature.

where Q_c = heat transferred to coolant
$\Delta T_{\text{out}} = T - T_c$
$\Delta T_{\text{in}} = T_0 - T_c$
T = reactor exit temperature
T_0 = reactor feed temperature
T_c = coolant temperature

A decrease in ΔT_{in} (due to an increase in the coolant temperature) leads to a correspondingly larger increase in ΔT_{out} for the same heat transferred. However, the heat generated by the reaction increases exponentially with temperature, forcing the exit temperature to climb further. This relation amplifies with large differences between ΔT_{out} and ΔT_{in} as well as with large, average activation energies.

4.5.4 Wrong-way behavior

The final open-loop reactor issue we discuss is the problem of inverse response or *wrong-way behavior* as it is called in the reactor engineering literature. The inverse response refers to the temporary increase in the exit temperature in some packed, plug-flow reactors following a decrease in the feed temperature (Fig. 4.12). The wrong-way behavior stems from the difference in propagation speed between concentration

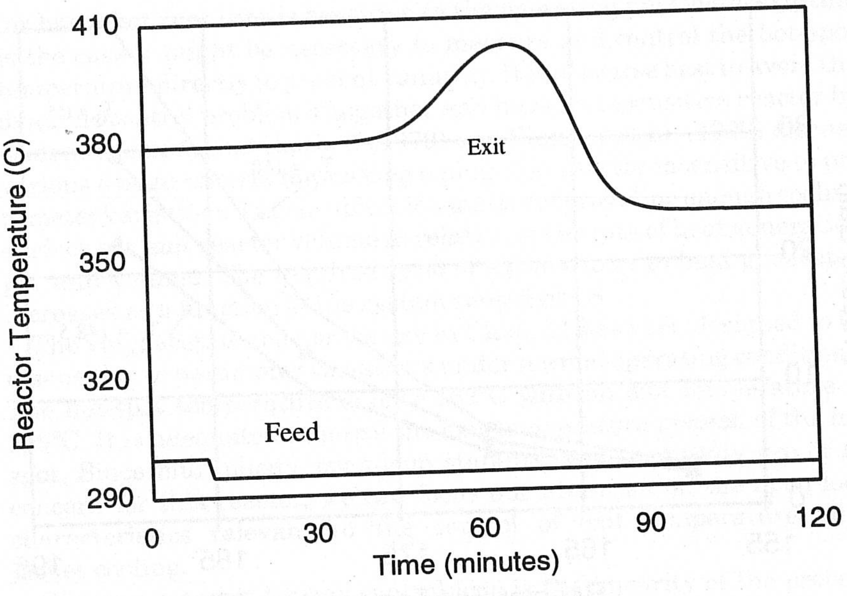

Figure 4.12 Wrong-way effect in packed plug-flow reactor.

and temperature disturbances. For example, when the feed temperature decreases, the conversion in the front end of the reactor decreases. When the higher concentration of reactants reaches the middle and back end of the reactor, where the packing is still hot, the reaction takes off and produces a transient temperature increase. Eventually the lower feed temperature will produce a new steady state with a lower exit temperature than before.

Inverse response creates control difficulties. Assume, for example, that we wish to control the exit temperature of an adiabatic plug-flow reactor by manipulating the inlet temperature as shown in Fig. 4.13. From a steady-state viewpoint this is a perfectly reasonable thing to consider, since there are no issues of output multiplicity or open-loop instability, assuming the fluid is in perfect plug flow and there is no

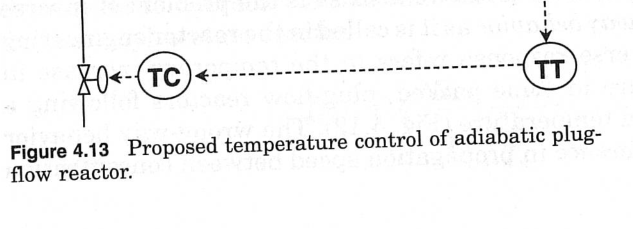

Figure 4.13 Proposed temperature control of adiabatic plug-flow reactor.

conduction in the packing. We also assume that the main reactant is nearly consumed at the exit. This means that the reactor cannot show parametric sensitivity because there is no more reactant left to increase the temperature in the reactor. However, as all control engineers know, the dynamic problem of inverse response severely limits the closed-loop performance achievable by any type of controller. For a stand-alone reactor the control problem can be avoided by controlling the reactor inlet instead of the exit temperature. However, when the reactor is part of an integrated process with feed preheating, this solution may not be sufficient. It is important to understand what design factors promote wrong-way behavior so that the phenomenon can be properly considered at the design stage.

The effect of various design parameters on wrong-way behavior in reactors has been thoroughly investigated by Luss and coworkers (Metha et al., 1981; Pinjala et al., 1988; Il'in and Luss, 1992 and 1993). Pinjala et al. (1988) used a simple model that is quite instructive in showing the influence of some of the design factors on the severity of wrong-way behavior. The model is written for a first-order reaction, $A \rightarrow B$, carried out in a packed tubular reactor. The model does not assume perfect plug flow since it allows for dispersion of material and energy. A single component balance and the energy balance take the following forms when written in dimensionless variables.

$$\frac{1}{\mathrm{Le}}\frac{\partial x}{\partial t} = \frac{1}{\mathrm{Pe}_m}\frac{\partial^2 x}{\partial z^2} - \frac{\partial x}{\partial z} - \mathrm{Da}\cdot e^{-(1/y)}\cdot x$$

$$\frac{\partial y}{\partial t} = \frac{1}{\mathrm{Pe}_h}\frac{\partial^2 y}{\partial z^2} - \frac{\partial y}{\partial z} + \beta\cdot\mathrm{Da}\cdot e^{-(1/y)}\cdot x + U(y_w - y)$$

where x = dimensionless composition variable, C_A/C_{A0}
y = dimensionless temperature, RT/E_a
t = dimensionless time, $(ut'/\epsilon L)/\mathrm{Le}$
z = dimensionless axial position, z'/L

The other parameter groups in the model are defined as follows:

Lewis number (ratio of thermal time constant to material time constant due to fluid flow):

$$\mathrm{Le} = 1 + \frac{(1-\epsilon)\rho_s C_{Ps}}{\epsilon\rho_0 C_{P0}}$$

Peclet number for mass (ratio of linear flowrate to diffusivity):

$$\mathrm{Pe}_m = \frac{Lu}{D_e}$$

Peclet number for heat (ratio of fluid heat flow to conductivity):

$$\mathrm{Pe}_h = \frac{Lu\rho_0 C_{P0}}{k_e}$$

Damköhler number (ratio of residence time to reaction time):

$$\mathrm{Da} = \frac{L}{u} A_f$$

Normalized adiabatic temperature rise (ratio of available reaction heat to feed heat capacity normalized by the activation energy):

$$\beta = \frac{R}{E_a} \frac{(-\Delta H)C_{A0}}{\rho_0 C_{P0}}$$

Dimensionless heat transfer parameter:

$$U = \frac{2hL}{ru\rho_0 C_{P0}}$$

where D_e = effective dispersion coefficient for mass
h = overall heat transfer coefficient
k_e = effective thermal conductivity
L = reactor length
r = radius of reactor
t' = time
u = superficial velocity
z' = axial position coordinate
ϵ = void fraction of bed
ρ_0 = feed density
ρ_s = packing density
C_{P0} = heat capacity of feed
C_{Ps} = heat capacity of packing

The Lewis number has been defined by Ray and Hastings (1980) as the ratio of two time constants, τ_h and τ_c. The thermal time constant τ_h is the ratio of the total heat capacity of a unit reactor volume V to the heat capacity of the fluids flowing through this volume. For an unpacked reactor this time constant equals the residence time, $\tau = V/F$. For a packed reactor the thermal time constant can be considerably longer than the residence time. Similarly, the material time constant τ_c is the ratio of material holdup in a unit reactor volume to the flow of material through this volume. Again, for an unpacked reactor, $\tau_c = \tau$, whereas a packed reactor could have the material time constant much shorter than the overall holdup time due to the space occupied by the packing. Pinjala et al. (1988) point out that only packed reactors can exhibit wrong-way behavior. They also show that the maximum temperature peak is independent of the Lewis number when $\mathrm{Le} > 100$.

As an example of how to compute the Lewis number we use the vinyl acetate reactor described in Chap. 11. The catalyst bulk density, $(1 - \epsilon)\rho_s$, is given as 385 g/L and the heat capacity of the packing, C_{Ps}, is 0.23 cal/g · °C. We previously calculated the fluid heat capacity to be 15.7 cal/g · mol · °C. The feed molar density is

$$\frac{P}{RT} = \frac{(128/14.7)}{0.08205(273.15 + 148.5)} = 0.25 \text{ g} \cdot \text{mol/L}$$

Porosity data is given for the catalyst as $\epsilon = 0.8$. The Lewis number for the vinyl acetate reactor is

$$\mathrm{Le} = 1 + \frac{(1 - \epsilon)\rho_s C_{Ps}}{\epsilon \rho_0 C_{P0}} = 1 + \frac{385(0.23)}{0.8(0.25)(15.7)} = 29$$

This means that it takes the fluid stream 29 times longer to cause a temperature change in the reactor than it takes to change the reactor composition. This is a sufficient difference in the propagation speeds to induce wrong-way behavior. However, a large Lewis number is not sufficient to create a problem. Another important requirement is that the reaction should be near completion at the exit of the reactor. This requirement is not met for the vinyl acetate reaction which has only a 36 percent conversion in oxygen.

For a reactor system with a significant Lewis number and a reaction that is near completion at the reactor exit, Pinjala et al. (1988) showed that the maximum peak of the temperature transient y^* is an increasing function of $\mathrm{Pe}_h\beta/\mathrm{Da}$:

$$y^* \propto \frac{\mathrm{Pe}_h \beta}{\mathrm{Da}} \tag{4.27}$$

Equation (4.27) predicts that the temperature peak will be severe when the adiabatic temperature rise is large and when there is little thermal conductivity in the system ($\mathrm{Pe}_h \gg 1$). By allowing the reactor to have significant dispersion of heat (through conduction or backmixing) it is possible to reduce the height of the temperature peak. However, as we allow for dispersion we introduce thermal feedback and create the potential for output multiplicity and instability. For example, it is possible that the entire reactor can switch to a second hot steady state following a decrease in feed temperature. This phenomenon starts as a temperature wave, initiated by the wrong-way behavior, moving slowly backward up the reactor until it reaches the feed point. Here, the feed mixture "ignites" and the entire reactor quickly switches to the high-temperature steady state. This ignition can have a disastrous impact on the packing or the integrity of the reactor.

4.6 Unit Control

4.6.1 Heat management

We noted earlier in this chapter that many reactions in the chemical industries are exothermic and require heat removal. A simple way of meeting this objective is to design an adiabatic reactor. The reaction heat is then automatically exported with the hot exit stream. No control system is required, making this a preferred way of designing the process. However, adiabatic operation may not always be feasible. In plug-flow systems the exit temperature may be too hot due to a minimum inlet temperature and the adiabatic temperature rise. Systems with backmixing suffer from other problems in that they face the awkward possibilities of multiplicity and open-loop instability. The net result is that we need external cooling on many industrial reactors. This also carries with it a control system to ensure that the correct amount of heat is removed at all times.

The control system must manipulate heat removal from the reactor, but what should be the measured (and controlled) variable? Temperature is a good choice because it is easy to measure and it has a close thermodynamic relation to heat. For a CSTR, temperature control is particularly attractive since there is only one temperature to consider and it is directly related to the heat content of the reactor. However, in a spatially distributed system like a plug-flow reactor the choice of measured variable is not so clear. A single temperature is hardly a unique reflection of the excess heat content in the reactor. We may select a temperature where the heat effects have the most impact on the operation. This could be the hot spot or the exit temperature depending upon the design of the reactor and its normal operating profile.

To summarize, we find that heat management is required for many industrial reactors and that this task can be accomplished by controlling a reactor temperature. We start our discussion of unit control by reviewing several reactor temperature control schemes.

Continuous stirred tank reactors. The simplest method of cooling a CSTR is shown in Fig. 4.14. Here we measure the reactor temperature and manipulate the flow of cooling water to the jacket. Using a jacket for cooling has two advantages. First, it minimizes the risk of leaks and thereby cross contamination between the cooling system and the pro-

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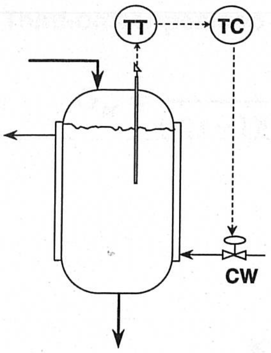

Figure 4.14 Simple CSTR temperature control.

cess. Second, there are no internals to obstruct an agitator from providing effective mixing. The main disadvantage of using jacketed cooling is the limitation on heat transfer area due to reactor geometry.

Direct supply of cooling water is a simple, reliable, and inexpensive method for cooling the reactor. However, it has some serious shortcomings. First, the water flow at low rates may be inadequate to maintain a good heat transfer coefficient. Second, the gain between the coolant flow and the amount of heat transferred varies nonlinearly with load, thereby making controller tuning difficult. Third, there can be a significant temperature gradient on the cooling water side that could create local hot spots on the process side.

Fortunately we can readily solve many of the problems associated with a direct supply of cooling water. For example, in Fig. 4.15 we have provided a water recirculation loop to maintain a large constant flow of water through the jacket. Fresh cooling water is added to the loop to maintain the desired reactor temperature. This arrangement keeps

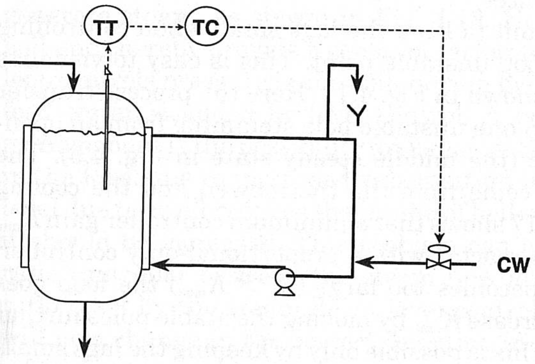

Figure 4.15 Circulating cooling water temperature control.

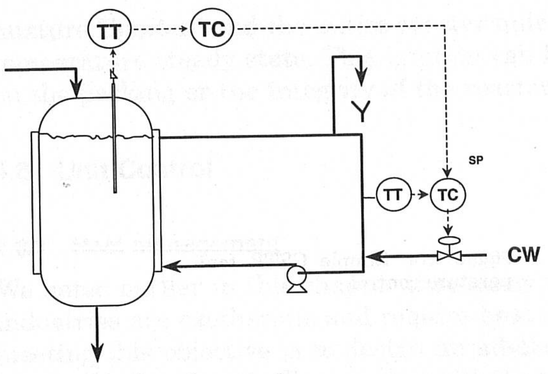

Figure 4.16 Cascade control.

the heat transfer coefficient constant, it linearizes the process gain, and it maintains a uniform temperature in the jacket. The only potential drawback with the arrangement in Fig. 4.15 is the dynamics added by the cooling water loop. Backmixing in the jacket loop averages out changes made in the cooling water supply. The entire jacket loop has to change temperature before there is a change in heat transfer from the reactor. This temperature lag is in series with the thermocouple lag and the thermal lag of the reactor content. With a low controller gain the reactor temperature could change slowly, causing sluggish control. With a high controller gain the jacket temperature may overshoot the correct value and cause the reactor temperature loop to be underdamped. The solution is to provide a cascade arrangement as shown in Fig. 4.16. The secondary controller maintains a target jacket temperature and the reactor temperature controller provides the setpoint. The jacket temperature controller can be tuned tightly, which effectively shortens the secondary lag caused by the cooling jacket dynamics. This makes it possible to tune the reactor temperature controller for better performance.

It is particularly important to keep the lags small when controlling a CSTR around an open-loop unstable point. This is easy to visualize in the root locus diagram shown in Fig. 4.17. Here the process transfer function is third order with one unstable pole stemming from an open-loop unstable steady state (the middle steady state in Fig. 4.6). The other two time constants come from the thermowell and the cooling jacket dynamics. Figure 4.17 shows that a minimum controller gain $K_{\min}$ is required to stabilize the reactor with a proportional-only controller. However, when the gain becomes too large ($K_c > K_{\max}$) the loop goes unstable again. We can increase $K_{\max}$ by moving the stable poles further to the left in the diagram. This is possible only by keeping the lags small.

The scheme shown in Fig. 4.16 can be extended to high-temperature

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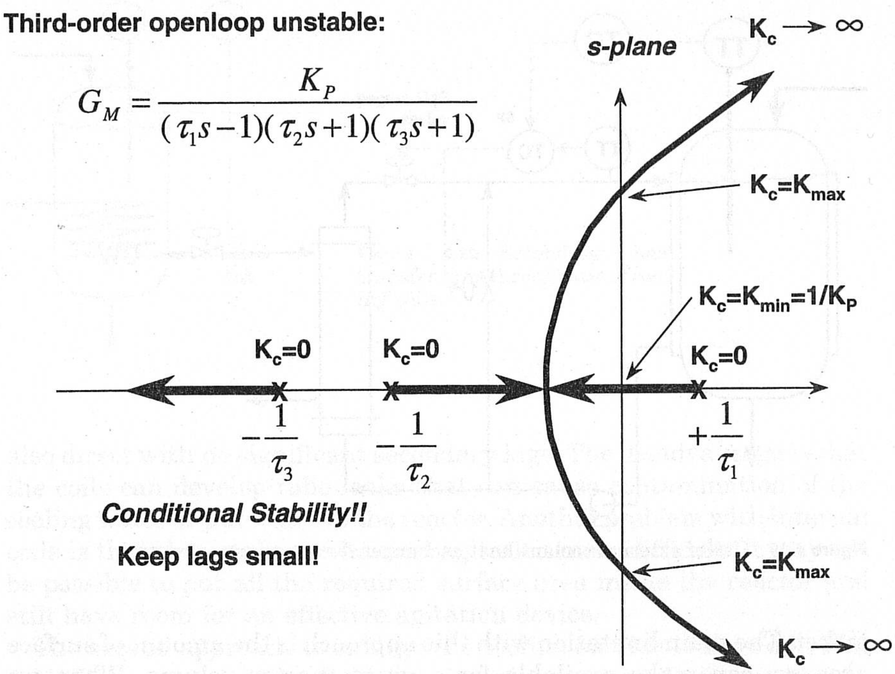


Figure 4.17 Root-locus plot of open-loop unstable CSTR.

applications. For example, in Fig. 4.18 we have closed the circulation loop thus allowing for higher pressure on the jacket side or a different coolant than water. The external heat exchanger still uses cooling water. To obtain faster temperature control and to avoid nonlinearities in response to cooling water flow, we provide a bypass around the external exchanger. The cooling water flow can now be kept constant while the bypass provides a rapid response in jacket temperature.

Another method for dealing with high reactor temperatures is to generate steam, as shown in Fig. 4.19. Here we allow the coolant to boil and thereby provide a constant jacket temperature. The secondary loop controls pressure in the boiler drum by venting steam. Fresh boiler feed water is added by level control. A potential problem with this arrangement is the possibility for boiler swell that results in an increase in the level due to increased vaporization in the jacket. The increased level due to swell reduces the intake of boiler feed water when in reality it should be increased. This problem can be overcome by providing a ratio controller between the steam flow and the feed water with the ratio reset by the steam drum level controller. Boiler feed water flow will now change in the correct direction in response to load.

The heat removal schemes we have examined so far all use a cooling

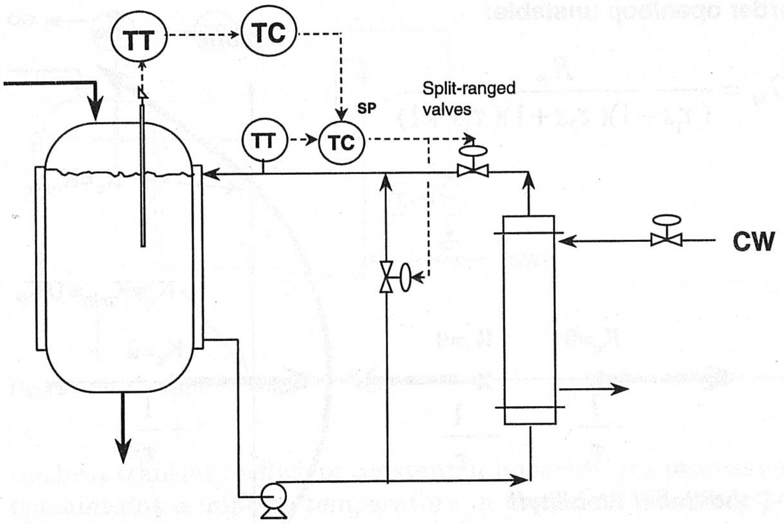


Figure 4.18 Use of external coolant heat exchanger for temperature control.

jacket. The main limitation with this approach is the amount of surface area we can make available for a given reactor volume. When we examine the open-loop characteristics of the reactor we may find that we need a larger surface area to avoid multiplicity and open-loop oscillations. Let us examine a few control approaches for systems with a large heat transfer area.

The first approach is to insert cooling coils in the reactor as shown in Fig. 4.20. The advantage with this method is that we don't have to handle the reactor content outside the reactor. The heat transfer is

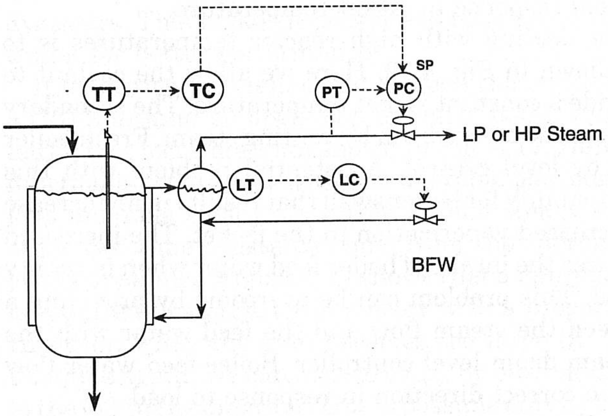


Figure 4.19 Temperature control through steam generation.

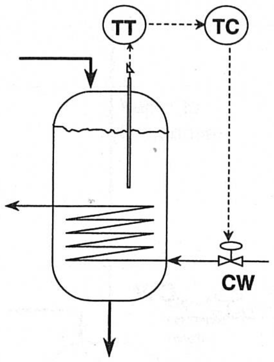


Figure 4.20 Extending heat transfer area through use of cooling coils.

also direct with no significant secondary lags. The disadvantage is that the coils can develop tube leaks that can cause contamination of the cooling water or put water in the reactor. Another problem with internal coils is that they make mechanical agitation more difficult. It may not be possible to put all the required surface area inside the reactor and still have room for an effective agitation device.

The second approach to enlarge the cooling surface is shown in Fig. 4.21. Here we bring the fluid reactor contents outside the reactor to cool the stream in an external heat exchanger. The industrial reactor shown in Fig. 4.8 used this principle. Also some fluidized-bed reactors, where the excess recycled gaseous reactants provide fluidization, commonly have these designs. There are two advantages with this method. First, the heat transfer area is completely independent of reactor size and geometry. Second, the circulation helps promote effective backmix-

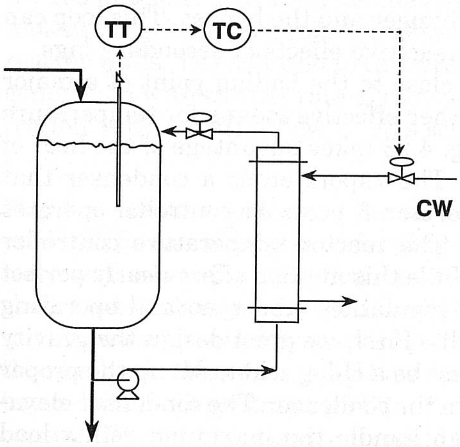


Figure 4.21 Circulation of reactor content through external heat exchanger.

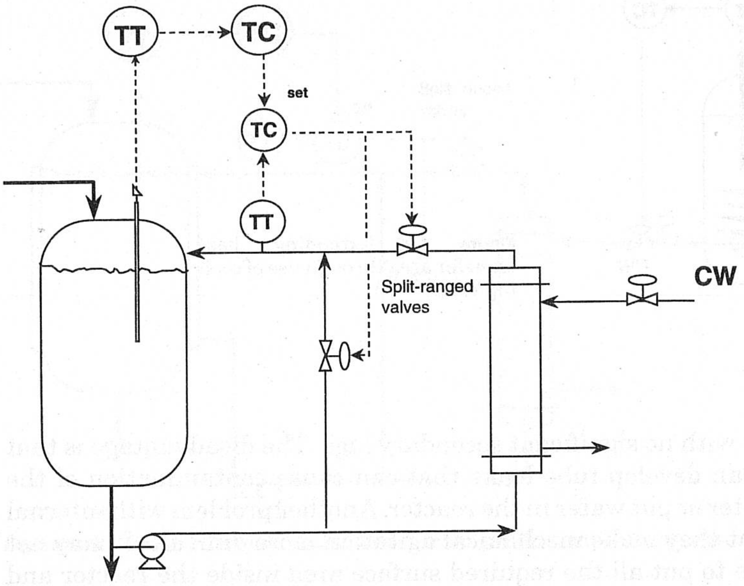


Figure 4.22 Bypass of circulating reactor content around external heat exchanger.

ing in the reactor. The main disadvantage is that we must be able to pump the reaction mass for liquid-filled reactors or compress the gases for fluidized beds.

The external heat exchanger shown in Fig. 4.21 introduces an undesirable and potentially troublesome secondary lag. To reduce the time constant due to the capacitance of the metal in the external exchanger, we can keep the coolant flow constant and bypass the exchanger as shown in Fig. 4.22. The secondary loop controls the mixed stream temperature coming from the exchanger and the bypass. This loop can be made fast, which reduces the negative effects of secondary lags.

Liquid-filled CSTRs operating close to the boiling point of a major component or a solvent offer another effective means for temperature control. The method shown in Fig. 4.23 takes advantage of the heat of vaporization to cool the reactor. The vapors enter a condenser that provides cold reflux back to the reactor. A pressure controller operates on the condenser cooling valve. The reactor temperature controller provides the pressure setpoint. While this method offers nearly perfect temperature control (due to self-regulation) under normal operating conditions, it also has some pitfalls. First, we must design the gravity return system properly. There must be a U-leg seal to set up the proper pressure balance for correct flow in the condenser. The condenser elevation must also be high enough to handle the maximum reflux load without having liquid back up in the condenser and reduce the heat

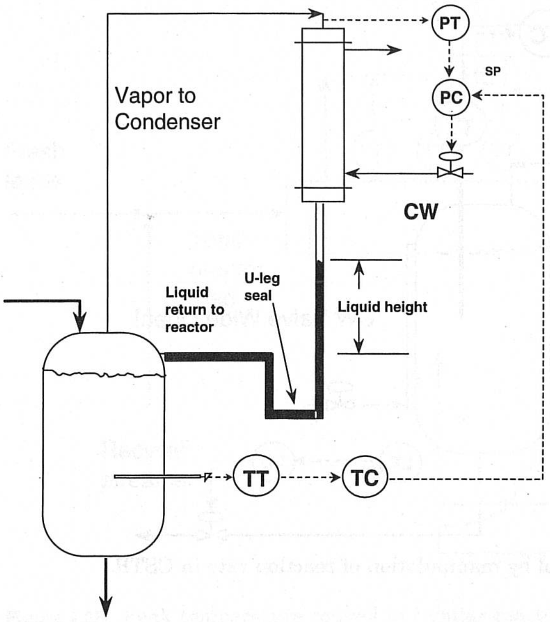


Figure 4.23 Cooling through vaporization of reactor content.

transfer surface. Second, we must provide a vent valve to remove noncondensables that could accumulate and blanket the condenser. Finally, it may be advantageous to provide a jacket with cooling water to help bring down the reactor temperature after a shutdown when the reaction has stopped and there is little vaporization.

In some situations the dynamics of the cooling system may be such that effective temperature control cannot be accomplished by manipulation of the coolant side. This could be the situation for fluidized beds using air coolers to cool the recirculating gases or for jacketed CSTRs with thick reactor walls. The solution to this problem is to balance the rate of heat generation with the net rate of removal by adjusting a reactant concentration or the catalyst flow. Such a scheme is shown in Fig. 4.24.

Plug-flow reactors. Many of the techniques discussed for cooling jacketed CSTRs can also be used for plug-flow systems. This works particularly well when the plug-flow reactor is implemented as a tube-and-shell heat exchanger. In such configurations the reaction takes place in the tubes (frequently packed with catalyst) and the coolant is on the shell side. However, plug-flow systems introduce some new issues beyond those for jacketed CSTRs. First, which temperature should we control? The exit temperature would be most convenient for a multitubular reactor since it avoids having to insert thermocouples inside

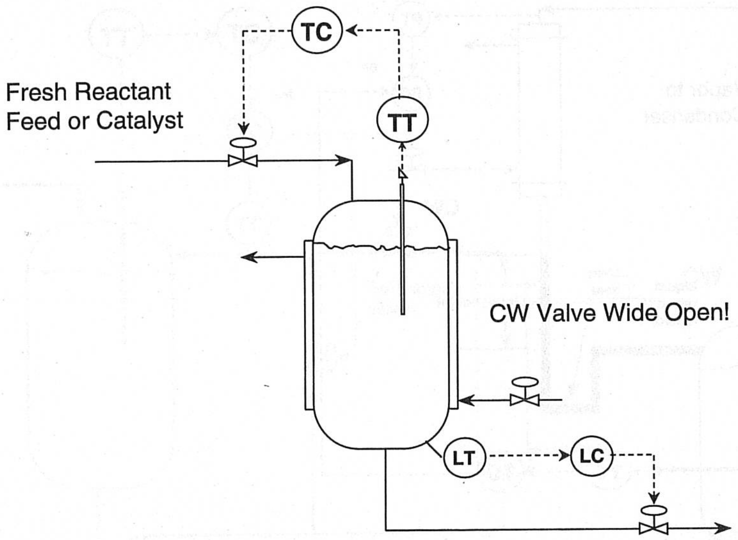


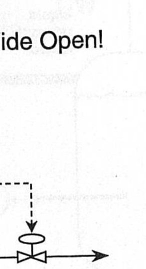
Figure 4.24 Temperature control by manipulation of reaction rate in CSTR.

narrow tubes with catalyst. On the other hand, if the hot spot is sensitive to operating conditions, we may have to control the peak temperature in the reactor. Since the hot spot frequently moves, depending upon the state of the catalyst and other operating parameters, it may be necessary to provide several measurements along the length of the reactor. The peak temperature can then be selected by using a high selector as shown in Fig. 4.25.

The second issue for cooled tubular reactors is how to introduce the coolant. One option is to provide a large flowrate of nearly constant temperature, as in a recirculation loop for a jacketed CSTR. Another option is to use a moderate coolant flowrate in countercurrent operation as in a regular heat exchanger. A third choice is to introduce the coolant cocurrently with the reacting fluids (Borio et al., 1989). This option has some definite benefits for control as shown by Bucalá et al. (1992). One of the reasons cocurrent flow is advantageous is that it does not introduce thermal feedback through the coolant. It is always good to avoid positive feedback since it creates nonmonotonic exit temperature responses and the possibility for open-loop unstable steady states.

The vinyl acetate reactor discussed in Chap. 11 uses boiling water on the shell side of the reactor. This arrangement is similar to that shown in Fig. 4.19.

It is sometimes necessary to achieve better control of the reactor temperature profile than can be accomplished in a cooled multitubular arrangement. For those cases we can arrange a series of short packed-



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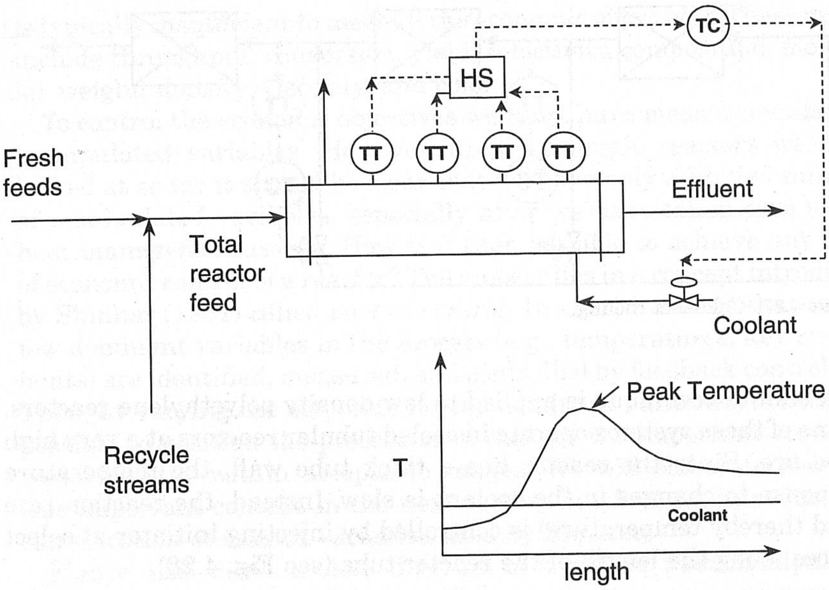


Figure 4.25 Peak temperature control in tubular reactor.

bed reactors with intermediate cooling (see Fig. 4.26). Each bed is operated adiabatically with an inlet temperature low enough to prevent complete conversion. This arrangement is also useful for reversible, exothermic reactions where the adiabatic temperature rise causes the reaction to reach equilibrium. Conversion stops at equilibrium and does not resume until the reaction mixture has been cooled in the intermediate heat exchangers. Ammonia and sulfur trioxide reactors are operated in this fashion.

Figure 4.27 shows another method of controlling plug-flow systems. Instead of cooling the effluents from each adiabatic step in a heat exchanger, we introduce a cold shot of fresh feeds. The cold shot technique increases the concentration of reactant in all the segments. Mixing the cold feed with the reactor effluent lowers the inlet temperature to the next reactor.

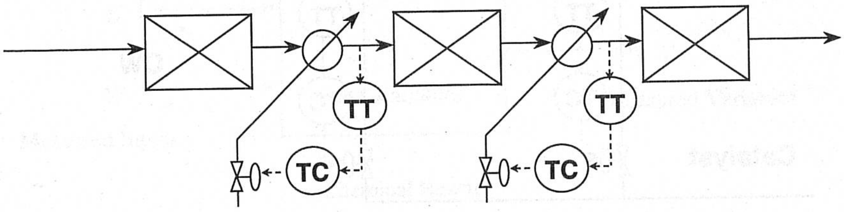


Figure 4.26 Intermediate cooling in sequence of packed adiabatic reactors.

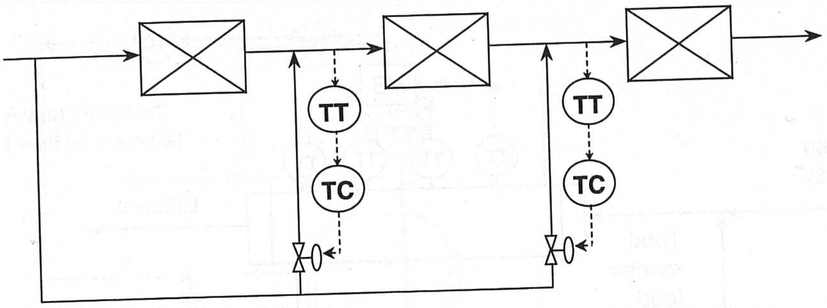


Figure 4.27 Cold-shot cooling.

A similar technique is applied to low-density polyethylene reactors. Some of these systems operate in cooled tubular reactors at a very high pressure. Since the reactor has a thick tube wall, the temperature response to changes in the coolant is slow. Instead, the reaction rate (and thereby temperature) is controlled by injecting initiator at select places along the length of the reactor tube (see Fig. 4.28).

4.6.2 Economic control objectives

Management of the heat removal in chemical reactors is required to meet thermodynamic constraints. In a sense, this is no different than satisfying mass, energy, or component balances. However, what makes heat management especially important is the close connection between heat and temperature. Temperature affects the reaction rates, which in turn control the rate of heat generation. Furthermore, in reversible reaction systems high temperature limits conversion. Proper heat management (through temperature control) becomes essential for safe, stable, and economic operation.

While safe operation is a necessary requirement to run a process it

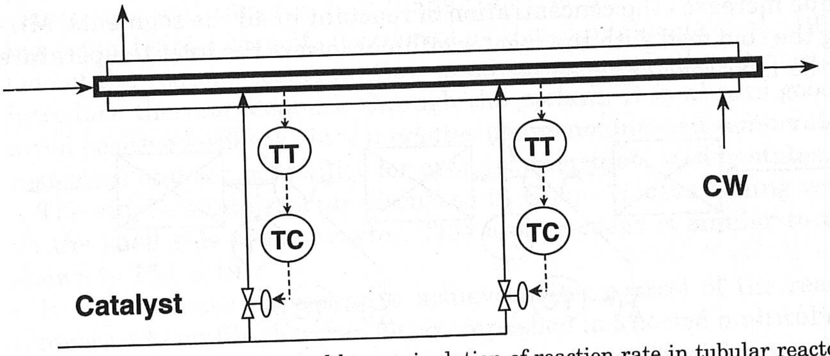
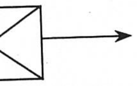


Figure 4.28 Temperature control by manipulation of reaction rate in tubular reactor.



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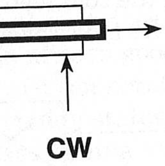

e in tubular reactor.

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is typically insufficient to meet all the economic objectives. These might include throughput, conversion, yield, selectivity, composition, molecular weight, density, viscosity, and color.

To control the economic objectives we must have measurements and manipulated variables. However, in the example reactors we have looked at so far it should be clear that we have only a limited number of manipulated variables, especially after we have taken care of the heat management issues. How is it then possible to achieve any level of economic control of a reactor? The answer lies in a concept introduced by Shinnar (1981) called *partial control*. In short it means that only a few dominant variables in the process (e.g., temperatures, key components) are identified, measured, and controlled by feedback controllers. Then, by varying the setpoints for the dominant variables, it becomes possible to position the process such that all the important economic variables stay within acceptable ranges. We will elaborate more on this important concept in the next section but first we introduce the classification of reactor variables used by Shinnar.

Shinnar (1981) does not use the term partial control

Figure 4.29 shows a block diagram of a reactor with manipulated inputs *U*, other measured inputs *W*, and unknown or unmeasured inputs *N*. We may assume that this reactor is more complicated than a simple plug-flow reactor or a CSTR. It may be more along the lines of the fluidized catalytic cracker that we showed in Fig. 4.4. The reactor can be described by a set of nonlinear differential equations as we have previously demonstrated. This results in a set of dynamic state variables *X*. The state vector is often of high dimension and we normally only measure a subset of all the states. *Y* is the vector of all measurements made on the system.

For the discussions around partial control it is convenient to classify the measurements further. For control purposes we pay particular attention to the variables Y_d (*d* for dynamic) that are measured continu-

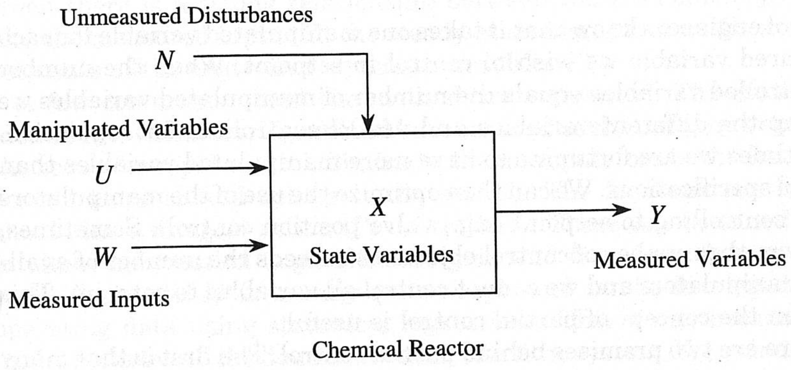

Figure 4.29 Reactor block diagram.