

Design and Control of Gas-Phase Reactor/Recycle Processes with Reversible Exothermic Reactions

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A gas-phase reactor with reversible exothermic reactions is the classical example of the many tradeoffs between operating costs and capital costs. Because per-pass conversion is limited by chemical equilibrium, recycle is almost always needed, and this gas recycle is expensive because of the high capital cost and high energy cost of gas compression. At low temperatures, conversion is limited by small kinetic reaction rates, and large reactors are needed. At high temperatures, conversion is limited by the decrease in the chemical equilibrium constant, and large recycle flowrates are needed. Thus, the optimum design of this type of system requires finding the best set of design optimization variables: reactor temperature, reactor size, and gas recycle flowrate. In systems containing inert chemical components that must be purged from the system, an additional design optimization variable is present: purge flowrate or inert composition in the recycle gas. Higher inert compositions require larger reactors and more recycle. Lower inert compositions result in larger losses of reactant components in the purge gas. These systems present intriguing dynamic control problems because of their integrating character and control loop interaction. In this paper we consider the design and control aspects of a ternary system with the gas-phase reversible, exothermic reaction $A + B \rightleftharpoons C$ occurring in an adiabatic tubular reactor packed with solid catalyst. Reactor effluent is cooled, and the product C is removed in a separator. Gas containing unreacted A and B (and inert I in the second part of the paper) is mixed with fresh feed and recycled back to the inlet of the reactor. The optimum design of this system is obtained, and the performances of several alternative control structures are explored.

1. Introduction

The art and science of engineering offer many examples of trade-offs at the design stage. Probably the most common is the trade-off between trays (capital cost) and reflux ratio (energy cost) in distillation column design. Other examples include the following.

(1) Heat exchanger design: area (capital cost) versus pressure drop (larger heat transfer coefficients, but higher pumping or compression costs).

(2) Control valve sizing: large pressure drop (high pumping cost) versus rangeability (ability to change flowrates over a wider range).

(3) Pipe sizing: smaller pipes (lower capital cost) versus higher pressure drop (higher head pump required and more work required).

(4) Heat exchanger design: small minimum approach temperature (more heat transferred) versus heat transfer area required (higher capital investment).

The area of reactor design abounds with many trade-offs, probably more than any other area in chemical engineering. A few common examples are the following.

(1) In consecutive reactions ($A \rightarrow B \rightarrow C$) with the intermediate B as the desired product, low per-pass conversion lowers the concentration of B and reduces the production of the undesirable product C. But more recycle is required, which increases both capital and energy costs.

(2) Several small CSTRs in series can achieve the same conversion as one large CSTR, thus reducing capital investment. However, the dynamic controllability of the one large CSTR may be much better than the

series of small CSTRs, yielding significantly better control of product quality and reducing safety and environmental risks.³ Temperature control in the large CSTR is much better because of the larger heat transfer area available for the same heat removal duty.

(3) With nonequimolar reactions in which a reduction in moles occurs, higher reactor pressures reduce reactor size (lower capital investment) but require more work in feed gas compressors.

The most important trade-off in reactor design is balancing reactor size and recycle flowrate. This involves finding the optimum reactor temperature, the optimum reactor size (holdup or amount of catalyst), and the optimum recycle flowrate. Increasing temperature can either increase or decrease reactor size, depending on how close to the chemical equilibrium constraint the reactor is operating. Increasing recycle flowrate requires smaller per-pass conversion, so reactor size is decreased, but compression costs increase.

There are many important industrial reaction systems that involve reversible exothermic reactions. The production of methanol and that of ammonia are two of the largest capacity examples. Reversible exothermic reactions present fascinating design issues because of the interplay among temperature, reactor size, and recycle. At low temperatures, a large reactor is needed because reaction rates are small. Because the equilibrium constant decreases with increasing temperature, running the reactor at high-temperature reduces per-pass conversion no matter how big the reactor is. This equilibrium limit means there is an optimum temperature for a given reactor size and recycle flowrate. Then an economic balance must be made between reactor size and recycle flowrate.

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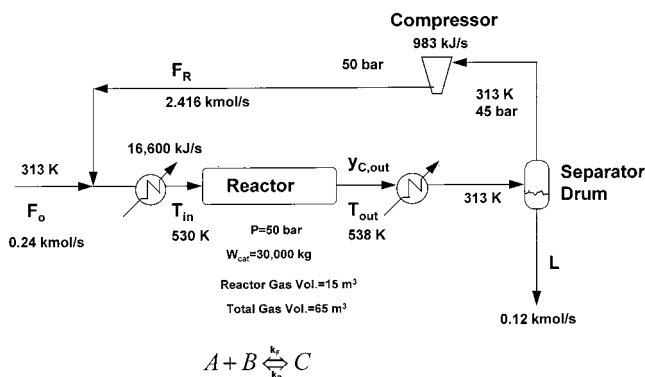


Figure 1. Steady-state conditions.

A recent paper by Jones and Wilson² presents a simplified reaction system and process flowsheet for an exothermic, reversible reaction. A very nice analysis of the problem of flexible operation of an existing plant is given. They deal with the problem of attaining the desired production rate despite slow changes in catalyst activity over time.

We use the Jones–Wilson process in this paper to study two other important issues related to this type of process: optimum design and dynamic control. In their paper, fresh feed contained only the two reactants. We consider in this paper both the case with pure reactants and the case with small amounts of an inert component in the fresh feed.

2. Process Studied (No Inerts)

Figure 1 shows the flowsheet of the process. The reversible reaction $A + B \rightleftharpoons C$ occurs in a gas-phase adiabatic tubular reactor. In this section, the feed contains only pure reactants A and B with no inerts. Following Jones and Wilson, we assume the fresh feed contains precisely stoichiometric amounts of the two reactants. In reality, this is never true because of flow measurement inaccuracy. Any practical control scheme must be able to keep track of the inventories of reactants in the system. Some type of feedback control must be used to adjust the quantities of the two reactants fed to the system. Since these reactants cannot leave the process, they must be completely reacted, and this means that every molecule of A fed requires exactly one molecule of B. This issue is discussed by Tyreus and Luyben.⁶ In this paper we assume that perfect stoichiometric amounts of reactants are fed.

The fresh feed flowrate is F_o (kmol/s) with composition y_{oj} (mole fraction of j , $j = A, B$). In the case with inerts considered later, the fresh feed also contains y_{oi} mole fraction of I. The fresh feed temperature is 313 K. It is mixed with recycle F_R (kmol/s) with composition y_{pj} (mole fraction of j). The total reactor feed is heated to temperature T_{in} (K) in a heat exchanger (or furnace). This inlet reactor temperature is a crucial design optimization variable.

In the tubular reactor, adiabatic plugflow conditions are assumed. The reactor contains solid catalyst with a total weight of W_{cat} (kg). The gas volume in the reactor is V_{gas} (m³). The catalyst bulk density is 2000 kg/m³, and porosity is 0.5. The heat of reaction is $\lambda = -14\,000$ kJ/kmol of C produced. The per-pass conversion is low, so the temperature rise through the reactor is only about 8 K. Kinetic parameters are those used by Jones and Wilson² and are given in Table 1. The reversible

Table 1. Parameter Values

heat of reaction λ (kJ/kmol of C)	-14000
specific reaction rate of the forward reaction k_F (kmol s ⁻¹ bar ⁻²) (kg of catalyst) ⁻¹	$100e^{(-94000/8.314T)}$
specific reaction rate of reverse reaction k_B (kmol s ⁻¹ bar ⁻¹) (kg of catalyst) ⁻¹	$525000e^{(-108,000/8.314T)}$
heat capacities (kJ kmol ⁻¹ K ⁻¹)	
C_{pA}	30
C_{pB}	40
C_{pC}	70
C_{pI}	30
molecular weights (kg/kmol)	
MW_A	15
MW_B	20
MW_C	35
MW_I	15
q , catalyst activity	0.3

reaction is favored by high pressure, so the operating pressure in the reactor is 50 bar.

Effluent from the reactor is cooled in a heat exchanger and fed to a separator. In a real process, this preheating and cooling would typically involve feed-effluent heat exchange to reduce energy consumption. This aspect of the problem is not considered in this paper, but it is discussed in detail in previous work.⁴

Component C is assumed to be nonvolatile, and components A and B are assumed to be noncondensable. This means all the C produced in the reactor leaves in the liquid stream from the separator drum (L kmol/s of pure C). The gas from the drum is the recycle stream back to the reactor and contains only A and B. The pressure drop around the gas loop is 5 bar, so the compression ratio in the gas compressor is 50/45. Ideal gas behavior and 75% compressor efficiency are assumed. When inerts are added to the system in a later section, a purge gas stream is also removed from the separator drum.

Base case conditions are shown in Figure 1. Note that the recycle flowrate is about 10 times the fresh feed flowrate.

3. Steady-State Economic Optimization

3.1. Economics and Sizing. The optimization of this process involves finding the values of reactor size (W_{cat} of catalyst), recycle flowrate (F_R), and reactor inlet temperature (T_{in}). The objective function to be minimized is the total annual cost (TAC, 10⁶\$/yr), which is the sum of annual capital cost (reactor, catalyst, and compressor capital investment divided by a payback period of 3 years) and energy cost of driving the compressor.

$$TAC = \text{energy} + \frac{\text{reactor} + \text{compressor} + \text{catalyst}}{\text{payback period}} \quad (1)$$

The compressor energy cost is calculated as a function of the recycle flowrate from the equation for reversible adiabatic compression of an ideal gas (kJ/kmol).

$$W = \frac{\gamma RT_1}{\gamma - 1} \left[\left(\frac{P_2}{P_1} \right)^{(\gamma-1)/\gamma} - 1 \right] \quad (2)$$

The ratio of heat capacities γ is assumed to be 1.312. Compressor suction temperature is 313 K. Suction pressure is 45 bar and discharge pressure is 50 bar. The annual energy cost of compression, assuming \$0.07/(kW h), is

$$\text{energy (10}^6\$/\text{yr)} = 0.227F_R \quad (3)$$

where F_R is in kmol/s. The capital costs of the compressor and reactor vessel are estimated from the correlations in Douglas¹

$$\text{compressor cost (10}^6\$) = 0.345(F_R)^{0.82} \quad (4)$$

$$\text{reactor vessel cost} = 0.035(D_R)^{1.066}(L_R)^{0.802} \quad (5)$$

with reactor diameter D_R and length L_R in meters. An aspect ratio of L_R/D_R is assumed for the reactor vessel to give reasonable pressure drop, and its cost is twice that of a plain pressure vessel. The cost of the catalyst is assumed to be \$5/kg in the base case. The impact of catalyst cost on the optimum design is studied in this section.

The ordinary differential equations describing the steady-state tubular reactor are given by Jones and Wilson.² The independent variable is catalyst weight w . These equations are integrated from 0 to W_{cat} .

$$\frac{dF_C}{dw} = \mathcal{R}_C \quad (6)$$

$$\frac{dT}{dw} = \frac{-\lambda \mathcal{R}_C}{F_A c_{pA} + F_B c_{pB} + F_C c_{pC}} \quad (7)$$

$$\mathcal{R}_C = q(k_F P_A P_B - k_B P_C) \quad (8)$$

where F_j is the molar flowrate of component j at any axial position, \mathcal{R}_C is the rate of production of C per kilogram of catalyst, q is the activity of the catalyst, c_{pj} is the molar heat capacity of component j (kJ/(kmol K)), P_j is the partial pressure of component j , and k_F and k_B are the specific reaction rates of the forward and reverse reactions. See Table 1 for details of parameter values. At the inlet of the reactor the molar flowrates are F_{0A} and F_{0B} , and the temperature is T_{in} . At any axial point down the length of the reactor, the flowrates are $F_A = F_{0A} - F_C$ and $F_B = F_{0B} - F_C$. The partial pressure of component j is calculated from the total pressure ($P = 50$ bar) and the molar flowrates.

$$P_j = \frac{PF_j}{F_A + F_B + F_C} \quad (9)$$

Remember that, without inerts in the system, both the recycle and the fresh feed contain only reactants A and B, and they are in exact stoichiometric ratios, i.e., 0.50 mole fraction of each.

The steps in the optimization procedure are summarized below.

- (1) The system pressure is fixed at 50 bar. The desired production rate is specified ($(F_C)^{\text{spec}}$).
- (2) A value for the amount of catalyst is picked (W_{cat}).
- (3) A value for the recycle flowrate F_R is guessed.
- (4) The reactor inlet temperature T_{in} is varied over a range of temperatures, and the temperature that gives the maximum production rate is found at the given recycle flowrate. See Figure 2.

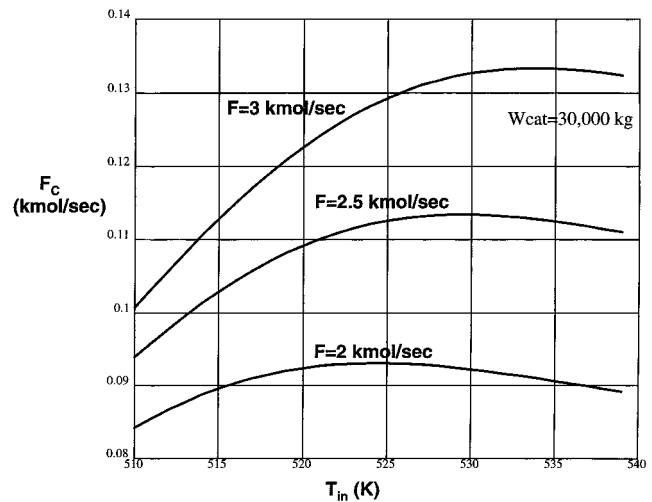


Figure 2. Effect of the inlet temperature and flowrate on the production rate.

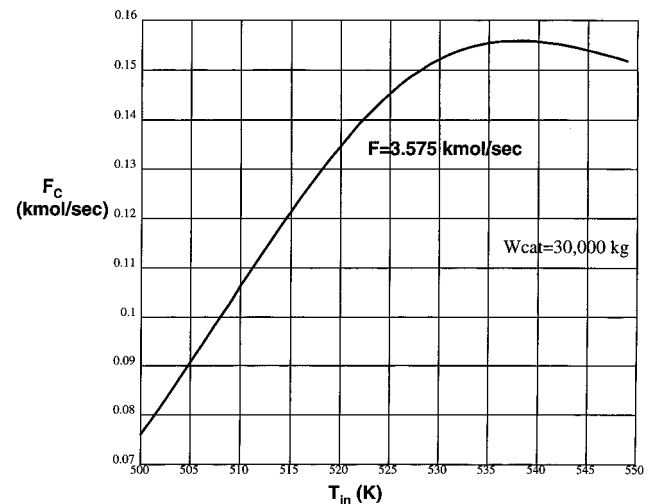


Figure 3. Effect of the inlet temperature and flowrate on the production rate at higher flowrate.

(5) Then an iterative convergence method (interval halving) is used to find the value of F_R that produces the desired $(F_C)^{\text{spec}}$. The inlet temperature that gives the maximum conversion is used in each case.

(6) Capital and energy costs are calculated at this recycle flowrate and reactor inlet temperature. The objective function of TAC is evaluated.

(7) Finally the amount of catalyst is varied over a range, and the catalyst holdup giving the minimum TAC is found.

3.2. Steady-State Design Results. Figure 2 illustrates the effect of inlet reactor temperature on the production of product C. These results are for a reactor containing 30 000 kg of catalyst. For a given flowrate of gas fed to the reactor F , there is an optimum inlet temperature that maximizes conversion. This optimum temperature increases slightly with increases in flowrate, but changes in the flowrate have more of an effect on the production rate. This figure shows that increasing reactor inlet temperature can either increase or decrease production rate, depending on where the process is operating with respect to the balance between reaction rates (kinetically limited) and chemical equilibrium (thermodynamically limited).

Figure 3 shows that to achieve a production rate of 0.15 kmol/s we would need to have a flowrate of about

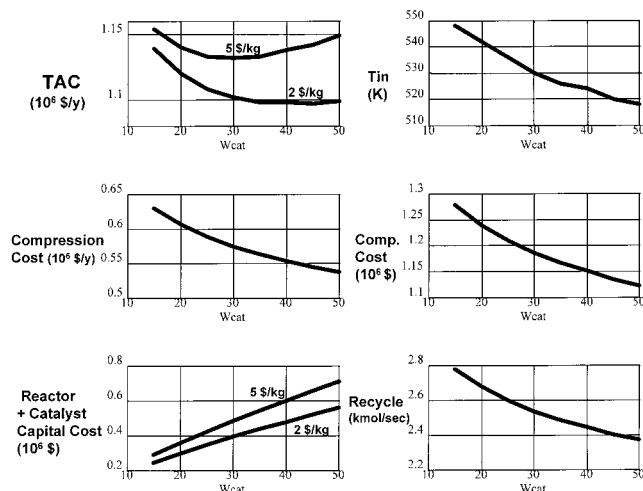


Figure 4. Optimum design at 0.12 kmol/s production rate.

3.5 kmol/s. If we only wanted a production rate of 0.12 kmol/s, we could reduce the flowrate or we could reduce the inlet temperature. We return to this case later when we discuss control structure alternatives.

Figure 4 summarizes optimum design results when a production rate of 0.12 kmol/s is specified. As the reactor is increased in size, the capital costs of the reactor and the catalyst increase, but the capital cost of the compressor and the compression energy cost both decrease because the recycle flowrate decreases. There are two curves shown for two different catalyst costs: \$2/kg and \$5/kg. As expected, increasing catalyst cost decreases the optimum size of the reactor and increases the optimum recycle flowrate.

Note that the capital cost of the compressor is high (about \$1 200 000), and the compressor energy cost is more than half the total annual cost.

4. Dynamics and Control

The conditions given in Figure 1 are used as a base case. For a production rate of 0.12 kmol/s, the optimum reactor contains 30 000 kg of catalyst and operates with a recycle flowrate $F_R = 2.416$ kmol/s. A dynamic model of the process is developed, and several alternative control structures are evaluated.

4.1. Dynamic Model. The tubular reactor is rigorously modeled using partial differential equations. A lumped-model approximation is used to capture the important dynamics with a minimum of complexity. There are no sharp temperature or composition gradients in the reactor because of the low per-pass conversion and high recycle flowrate. Therefore a 10-lump model is used. The steady-state temperature and composition profiles in the lumped model are very close to those calculated from the rigorous steady-state distributed model discussed earlier.

The thermal capacitance of the gas in the reactor is assumed negligible compared to that of the solid catalyst. Therefore, a single dynamic energy balance is used for each lump, and the gas temperature is assumed to be equal to the catalyst temperature.

The energy balance for the n th lump is

$$c_{p,cat} W_{cat,n} (dT_n/dt) = F_{n-1} c_{p,n-1} T_{n-1} - F_n c_{p,n} T_n - \lambda R_{C,n} \quad (10)$$

where $W_{cat,n} = W_{cat}/NR$, NR = number of lumps. The

heat capacity of the catalyst is 0.837 kJ/(kg K). The reaction rate is given by

$$R_{C,n} W_{cat,n} q(k_{F,n} P_{n,A} P_{n,B} - k_{B,n} P_{n,C}) \quad (11)$$

where $P_{n,j} = P y_{nj}$ and y_{nj} is the mole fraction of j in lump n .

The dynamic component balances on the gas phase in each lump assume a constant average molar density of $\rho_{av} = 1.124$ kmol/m³ (at 535 K and 50 bar). The gas holdup in each of the 10 lumps is 1.5 m³. With about 2.65 kmol/s flowing through the reactor, the gas holdup time per lump is about 0.5 s.

Component balances for the n th lump ($j = A, B$) are

$$V_{gas} \rho_{av} (dy_{nj}/dt) = F_{n-1} y_{n-1,j} - F_n y_{nj} - R_{C,n} \quad (12)$$

Component balances for n th lump ($j = C$) are

$$V_{gas} \rho_{av} (dy_{nj}/dt) = F_{n-1} y_{n-1,j} - F_n y_{nj} + R_{C,n} \quad (13)$$

where ρ_{av} is the average molar density. Note that the reaction is nonequimolar, so the total flowrate decreases down the length of the reactor.

The total pressure in the gas loop is calculated from an approximate dynamic total mass balance. Gas enters in the fresh feed, and gas is converted into liquid component C in the heat exchanger following the reactor. An average density is used assuming an average molecular weight of 17.5 kg/kmol and an average temperature of 500 K. The total volume in the reactor V_{gas} and in the separator drum V_{drum} is assumed to be 65 m³.

The total mass balance on the gas loop is

$$\frac{(17.5)(V_{gas} + V_{drum})}{(0.08314)(500)} \frac{dP}{dt} = F_o(17.5) - (35)F_{NR,C} \quad (14)$$

The total mass balance on the drum is

$$dM_D/dt = F_{NR,C} - L \quad (15)$$

where M_D is the molar liquid holdup in the separator and L is the liquid flowrate (kmol/s).

4.2. Control Structures. The process has four variables that must be controlled: pressure, production rate, separator drum liquid level, and separator inlet temperature. Pressure is normally held slightly below the maximum equipment design pressure because high pressure favors the forward reaction. We must be able to directly or indirectly set the production rate. The separator liquid level is held by a proportional controller manipulating liquid product flowrate L in all control structures considered below. The separator inlet temperature should be as low as possible. This accomplishes two objectives. First, it minimizes compressor work because the work of compression is directly related to suction temperature T_1 . Second, it makes sure all the product C is condensed and removed from the system. In all the simulation studies we assume drum temperature is perfectly controlled at 313 K.

In addition to the four controlled variables mentioned above, the reactor inlet temperature also should be controlled. However, the value of this temperature could be changed if it is desirable to do so for some reason. The inlet temperature is controlled by manipulating heat transfer in the preheater Q_H (high-pressure steam

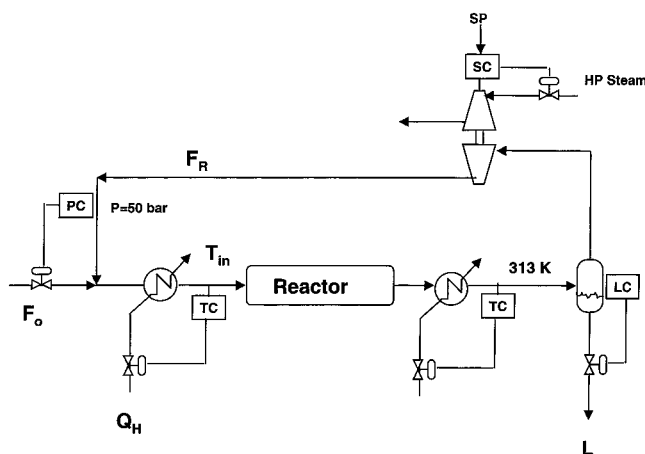


Figure 5. CS1: pressure controlled by fresh feed; recycle fixed.

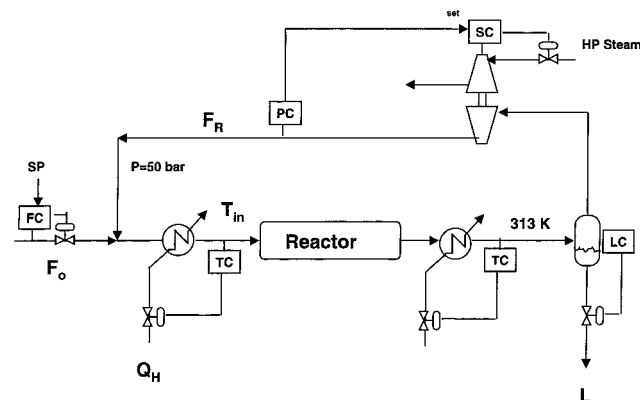


Figure 6. CS2: pressure controlled by recycle; fresh feed fixed.

flowrate or fuel to a furnace). In one of the control structures considered below (CS3), we use the reactor inlet temperature to indirectly set the production rate.

There are potentially five manipulated variables. Two of them, condenser duty and liquid product, are used as described above. The remaining three are fresh feed F_o , recycle flowrate F_R (compressor speed), and pre-heater duty Q_H . The control structures discussed below use different pairings of these variables.

(1) CS1: Figure 5 gives a control structure in which fresh feed controls pressure. The reactor inlet temperature is fixed. The recycle flowrate is used to indirectly set the production rate. If more throughput is desired, compressor speed is increased. This increases the conversion of reactant to product C. The pressure in the system drops because all the C is condensed. The pressure controller then pulls in more fresh feed to maintain gas loop pressure.

(2) CS2: Figure 6 shows a control structure in which pressure is controlled by recycle flowrate and the production rate is directly set by the fresh feed flowrate.

(3) CS3: Figure 7 gives a control structure where the recycle flowrate is fixed, pressure is controlled by the fresh feed, and the production rate is indirectly set by changing the reactor inlet temperature.

The first two structures are straightforward. The last is not so simple because it requires a changing reactor inlet temperature. If the system is operating at the peak of the conversion–temperature curve (see Figure 2), throughput will *decrease* if we make *any* change in inlet temperature. In this situation there is no way to increase the production rate without changing the recycle flowrate. Therefore, for this control structure to

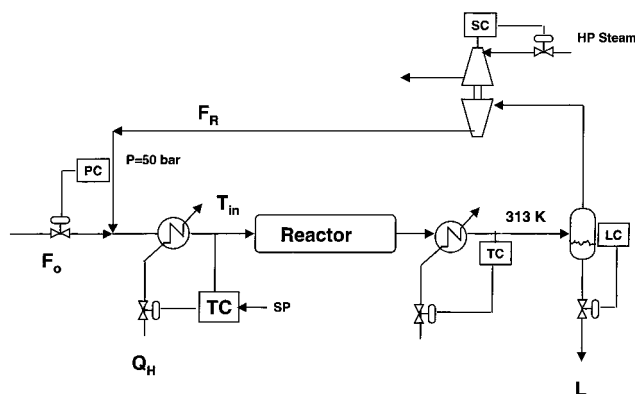


Figure 7. CS3: pressure controlled by fresh feed; recycle fixed; production rate set by the reactor inlet temperature.

Table 2. Controller Tuning Parameters^a

structure	controlled variable	manipulated variable	τ_m or D (s)	K_u	P_u	K_c	τ_I
CS1	P	F_o	30	8.0	180	1.2	400
CS2	P	F_R	60	6.4	320	1.0	700
CS3	T_{in}	Q_H	30	3.2	120	0.8	260
CS4	P	F_o	30	25	196	4.0	430
	yPI	F_P	180	250	750	75	1560

^a Pressure transmitter span 10 bar; composition transmitter span 0.50 mole fraction; temperature transmitter span 50 K. Control valves sized for twice the steady-state value.

be operable, the normal steady-state reactor inlet temperature must be below the peak. This means that the system must be operating with a recycle flowrate that is higher than the base case.

The results shown in Figures 2 and 3 illustrate this point. Suppose the normal operating throughput is 0.12 kmol/s of product and the reactor contains 30 000 kg of catalyst. Figure 2 shows that to achieve this capacity a flowrate (recycle plus fresh feed) of about 2.55 kmol/s is needed, using an inlet temperature of about 530 K. To reduce throughput with the recycle flowrate fixed, we can reduce the reactor inlet temperature. But we cannot increase throughput. To have rangeability in both directions, a higher recycle flowrate (such as shown in Figure 3) must be used. Now production rate can be manipulated by changing the reactor inlet temperature, up to the limit of about 0.15 kmol/s at a temperature of 530 K. Beyond this point an increase in T_{in} produces a reduction in capacity. Thus, the CS3 control structure requires higher recycle flowrates under nominal conditions. This implies higher energy costs.

4.3. Dynamic Results. The loops are tuned by inserting reasonable temperature and pressure lags in the loops and running a relay-feedback test to obtain the ultimate gain and ultimate period. The Tyreus–Luyben settings⁷ are used with some empirical reduction in controller gains. See Table 2 for details. Note that the pressure loop lags are assumed to be smaller (0.5 min) than the compressor speed lags (1 min) since rapid changes in compressor speed are undesirable for mechanical reasons.

Figures 8–13 give results for these three control structures with a variety of disturbances. In Figure 8 CS1 and CS2 are compared for a step increase in the reactor inlet temperature of 20 K. Since the reactor started at the optimum inlet temperature, an increase in temperature results in a drop in the production rate with the recycle flowrate fixed (CS1). Figure 8 shows that, after an initial drop in pressure, pressure begins

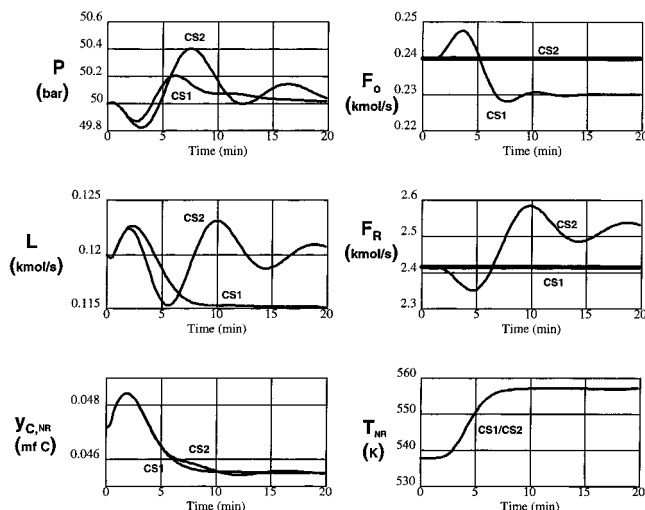


Figure 8. CS1 and CS2: +20 K inlet temperature.

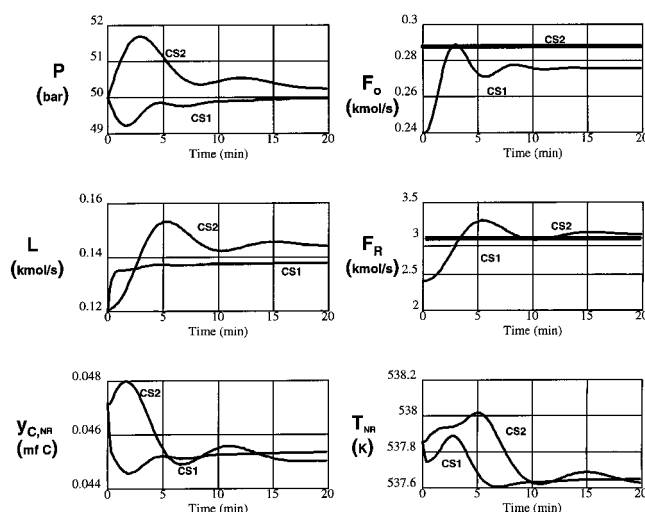


Figure 9. CS1 and CS2: +20% fresh feed or recycle.

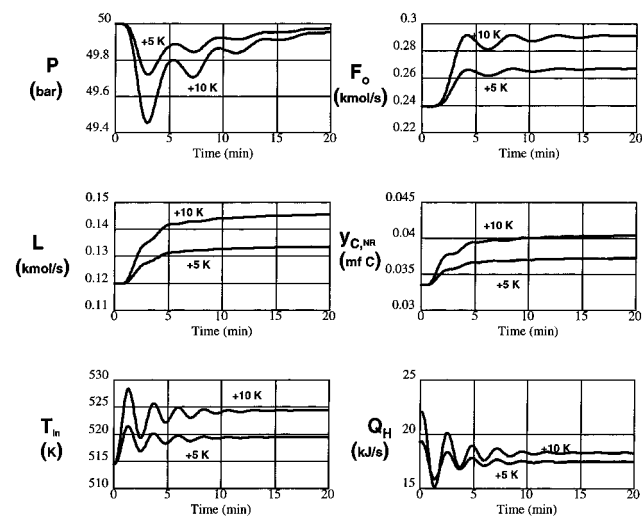


Figure 10. CS3: +5 and +10 K inlet temperature.

to increase, which cuts fresh feed in CS1. The product flowrate L gradually decreases from 0.12 to 0.115 kmol/s, which corresponds to the drop in fresh feed F_o from 0.24 to 0.23 kmol/s. In CS2 the fresh feed is fixed, and the increase in pressure results in an increase in recycle flowrate, which maintains the production rate at 0.12 kmol/s. Notice that in both structures there is a decrease

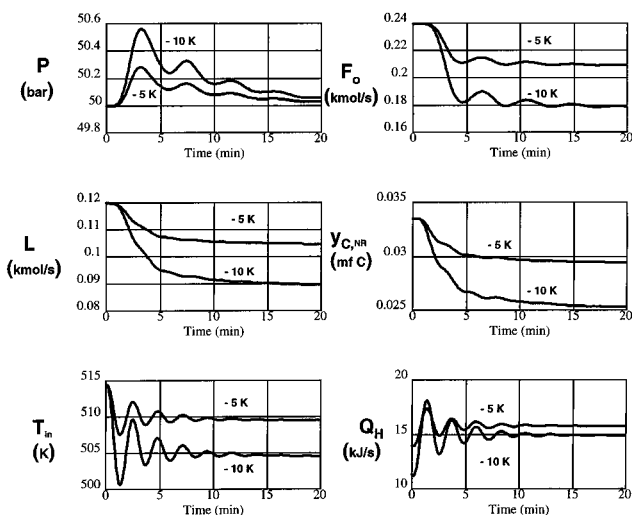


Figure 11. CS3: -5 and -10 K inlet temperature.

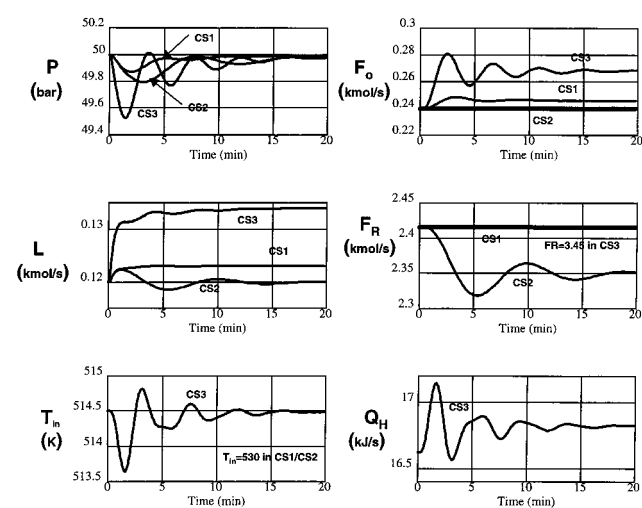


Figure 12. +20% change in catalyst activity.

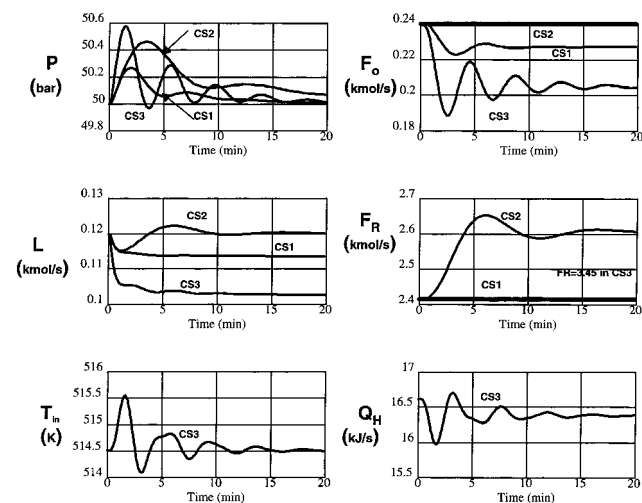


Figure 13. -20% change in catalyst activity.

in the concentration of C leaving the reactor ($y_{C,NR}$), due to the reduction in the net reaction rate at the higher temperatures in the reactor.

In Figure 9 the fresh feed or recycle flowrates are increased 20%. The production rate increases in both structures, but CS1 is somewhat faster because the pressure-to-feed loop can be tuned faster than the

pressure-to-compressor speed loop due to the larger dynamic lags.

In Figure 10 the response of structure CS3 is shown for step changes in the setpoint of the inlet reactor temperature controller. Further increases in the reactor inlet temperature would not produce much of an increase in the production rate because the equilibrium constraint is approached. Figure 11 shows that throughput can be easily reduced by dropping the reactor inlet temperature.

Figures 12 and 13 show how all three structures respond for changes in catalyst reactivity. The parameter q is changed $\pm 20\%$ from its nominal value of $q = 0.3$. The production rate in CS2 is maintained constant despite these changes by varying the recycle flowrate. Both CS1 and CS3 experience changes in throughput, but CS3 is more sensitive to catalyst activity.

All three of these alternative control structures are stable and workable. However, on balance the CS1 control structure has several advantages over the others. The compressor speed is held constant, which is often desirable because of mechanical problems. Maximum capacity is easily achieved by simply maximizing the recycle flowrate and setting the reactor inlet temperature at the peak value on the production-temperature curve.

Structure CS2 has the advantage of having a direct handle on the production rate, but manipulation of compressor throughput is required and control is slower and not as tight as with CS1. In this paper compressor speed is used to set the recycle flowrate. Naturally there are other alternatives such as suction throttling and spillback that can be used for a constant-speed compressor. These techniques use more energy at flowrates less than maximum. Structure CS3 also wastes compression power at less than maximum load. In addition manipulation of the heat added to the reactor feed can sometimes be slow if a fired-furnace is used.

5. Process with Inerts

Up to this point, the fresh feed to the process has contained only reactants A and B. In most industrial cases the fresh feed streams are not completely pure but contain small amounts of impurities. This occurs because it may be very expensive to purify these streams beyond certain limits. In this section we consider such a situation. The fresh feed now has a composition y_{oi} where $j=A, B$, and I . The inert component I is assumed to be low-boiling, so it cannot leave the process in the liquid product stream leaving the bottom of the separator drum. It will, therefore, build up in the system unless a purge stream is added.

Now the design problem has the added dimension of establishing the optimum purge flowrate, or equivalently, the composition of the inert component in the purge and recycle streams y_{PI} . The modified flowsheet is sketched in Figure 14. One additional manipulated variable has been added (purge flowrate F_P), but one additional controlled variable has also been added (y_{PI}).

We explore this type of process in the following sections from the standpoint of both design and control.

5.1. Optimum Design. The optimum design procedure must be slightly revised to find the optimum purge gas inert composition y_{PI} . An additional step is added to the procedure outlined above. For a given value of inert concentration in the fresh feed y_{oi} , a range of values for y_{PI} is covered, using the original procedure

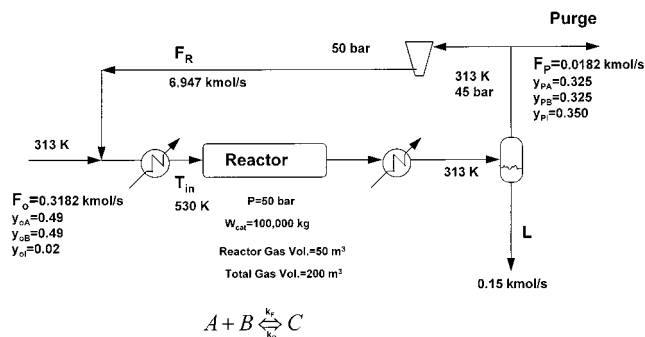


Figure 14. Steady-state conditions with inert components in the feed ($y_{oi} = 0.02$).

at each y_{PI} value. The result is the minimum TAC design (catalyst, recycle flow, and inlet temperature) for each purge gas inert concentration considered. Then the y_{PI} giving the smallest TAC is selected as the optimum case.

One important difference between the original problem without inerts and this problem with inerts is accounting for the loss of reactants in the purge stream. This is handled by assuming that the total production rate of C is fixed at 0.15 kmol/s for all inert values. As the purge flowrate increases (y_{PI} decreases), more fresh feed must be fed into the system because some A and B are lost in the purge. A value of \$5/kmol is used for the reactants lost in the purge.

For given fresh-feed and purge gas inert impurity levels (y_{oi} and y_{PI} , respectively) and the desired production rate of C (F_C), the flowrates of the purge F_P and the fresh feed F_o can be found from inert component balances around the system. First F_o is calculated from

$$F_o = \frac{2F_C}{1 - y_{oi} - y_{oi}(1 - y_{PI})/y_{PI}} \quad (16)$$

Then the purge flowrate (F_P) can be calculated.

$$F_P = F_o y_{oi} / y_{PI} \quad (17)$$

The amount of reactant lost in the purge is $F_P(1 - y_{PI})$. At \$5/kmol the annual cost of reactant losses is

$$\text{cost losses (10}^6 \text{ \$ / yr)} = 160 F_P (1 - y_{PI}) \quad (18)$$

This cost is added directly to the energy and annual capital costs to find the TAC in eq 1.

Results for several values of feed inert compositions are given in Table 3 and Figure 15. The very significant economic impact of feed impurity is clearly shown in these results. Having only 2% inert component in the feed triples the TAC and almost doubles the reactor size. As the feed impurity increases, the optimum reactor size, recycle flowrate, and purge gas inert composition all increase.

For a given feed impurity level, increasing the purge gas impurity concentration results in more recycle flow, less purge flow, slightly less fresh feed, higher compressor energy consumption, lower reactant losses in the purge and higher capital costs of the reactor, catalyst, and compressor.

5.2. Dynamics and Control. 5.2.1. Revised Model. The dynamic model must be modified to account for the effect of the inert component in the reactor and recycle

Table 3. Effect of the Inert Component on Optimum Design^a

inert component in feed y_{oi}	0	0.02	0.04	0.06
(mole fraction of I)				
catalyst W_{cat} (10^3 kg)	55	100	120	200
recycle F_R (kmol/s)	2.89	6.95	8.08	11.3
inert component in the purge	n.a.	0.35	0.40	0.50
y_{PI} (mole fraction of I)				
purge flowrate (kmol/s)	0	0.0182	0.0333	0.0407
inlet temp (K)	518	530	528	526
capital costs (10^6 \$)				
reactor vessel	0.49	0.78	0.80	1.09
catalyst	0.11	0.50	0.60	1.00
compressor	1.37	2.71	3.07	4.04
energy cost (10^6 \$/yr)	0.684	1.58	1.83	2.56
purge losses (10^6 \$/yr)	0	1.87	3.16	3.23
TAC (10^6 \$/yr)	1.34	4.75	6.48	7.84

^a Production rate 0.15 kmol/s.

stream. The partial pressure of component j is

$$P_j = \frac{Py_j}{y_A + y_B + y_C + y_I} \quad (19)$$

A dynamic component balance for the inert component is added.

Component balances for the n th lump ($j = I$) are

$$V_{gas}\rho_{av}(dy_{ij}/dt) = F_{n-1}y_{n-1,j} - F_n y_{nj} \quad (20)$$

The total mass balance on the gas loop includes the removal of material in the purge stream F_p .

$$\frac{MW_P(V_{gas} + V_{drum})}{(0.08314)(500)} \frac{dP}{dt} = F_0 MW_0 - (35)F_{NR}V_{NR,C} - F_P MW_P \quad (21)$$

where MW_n is the average molecular weight of stream n .

5.2.2. Control Structure CS4. The additional control loop added is the control of the composition of the inert component in the recycle and purge gas y_{PI} . The manipulated variable is the purge gas flowrate F_p . Figure 16 shows the control scheme. The purge flowrate is quite small compared to the total volume in the system, so its use for pressure control is ineffective.

A 3 min deadtime is assumed for the composition measurement of y_{PI} . The case with 0.02 mol % inert component in the fresh feed is studied. Steady-state parameter values are given in Figure 14. Note that the reactor is 3 times the size of that in the zero-inert case. The pressure and composition loops are tuned by using the relay feedback test and Tyreus–Luyben tuning. The pressure controller gain had to be reduced to get less underdamped performance. See Table 2.

The tuning of the y_{PI} composition controller requires a large reset time. The system is a pure integrator with respect to inert composition. The 3 min deadtime with an integrator gives an ultimate period ($P_u = 2\pi/\omega_u$) of $4D$, where D is the deadtime.

$$\arg G = \arg\left(\frac{e^{-Ds}}{s}\right) = -\frac{\pi}{2} - \omega D = -\pi @ \omega = \omega_u \quad (22)$$

For a 3 min deadtime, the Tyreus–Luyben reset time is 26 min. This gives very slow control of purge gas composition, but it is adequate for basic regulatory control.

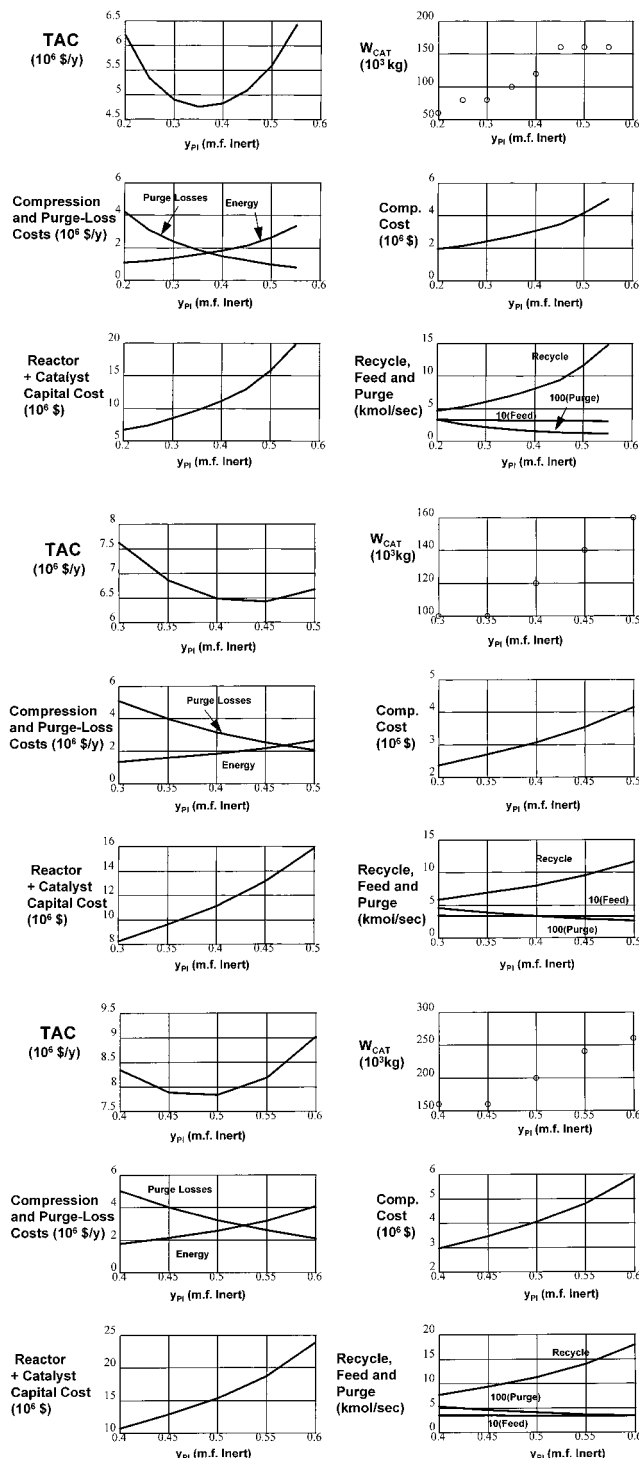


Figure 15. (A, top three rows) Optimum design with inert components in the feed ($y_{oi} = 0.02$). (B, middle three rows) Optimum design with inert components in the feed ($y_{oi} = 0.04$). (C, three bottom rows) Optimum design with inert components in the feed ($y_{oi} = 0.06$).

5.2.3. Dynamic Results. Figures 17–19 give results for disturbances in recycle the flowrate ($\pm 20\%$), reactor inlet temperature (± 20 K) and feed inert composition (y_{oi} changing from 0.02 to 0.03 and 0.01 mole fraction of I). Stable control is achieved for all these large disturbances.

The effect of changing the reactor inlet temperature is interesting (Figure 18). Both increasing and decreasing temperature result in a decrease in throughput since the normal inlet temperature is at the peak of the curve.

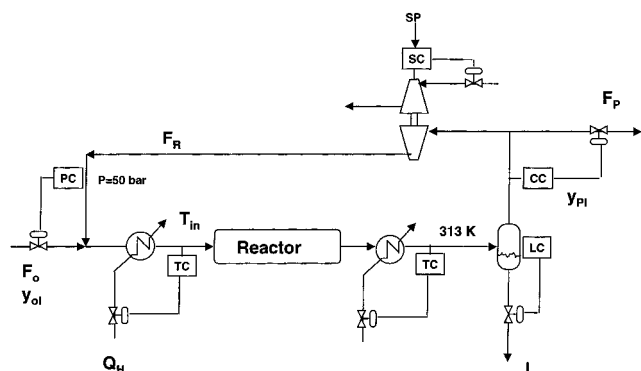


Figure 16. CS4: inert composition controlled by the purge gas; pressure controlled by fresh feed; recycle fixed.

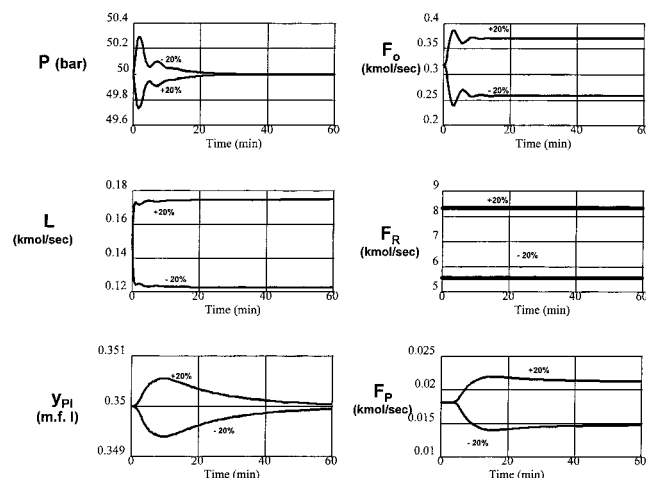


Figure 17. CS4 (with inerts): +20% and -20% recycle.

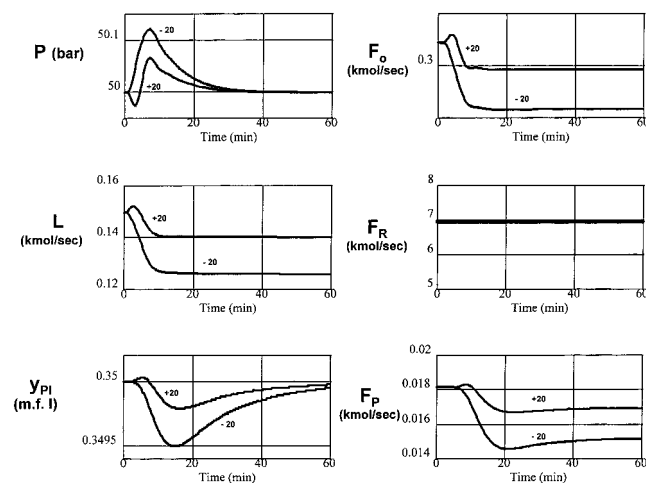


Figure 18. CS4 (with inerts): +20 and -20 K inlet temperature.

The effect of changing the feed inert concentration is also interesting. As Figure 19 shows, an increase in the feed inert concentration produces a decrease in the production of C, but the flowrate of the fresh feed actually increases! This occurs because the purge flowrate increases and much more A and B are lost.

5.2.4. Control Structure for Nonstoichiometric Feeds. As mentioned earlier, in a real application the fresh feed will not contain exactly stoichiometric amounts of the two reactants. Any slight misbalance will lead to a gradual buildup in the reactant that is in excess. When inert components are in the feed and a purge gas exists, any imbalance in the fresh feed will be compen-

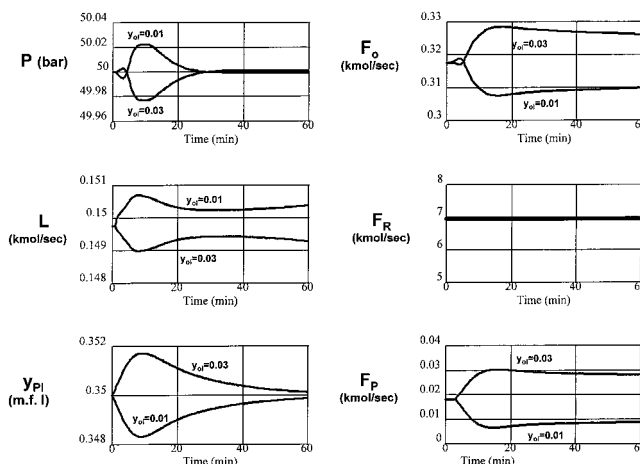


Figure 19. CS4 (with inerts): feed inerts changed from 0.02 to 0.03 and from 0.02 to 0.01.

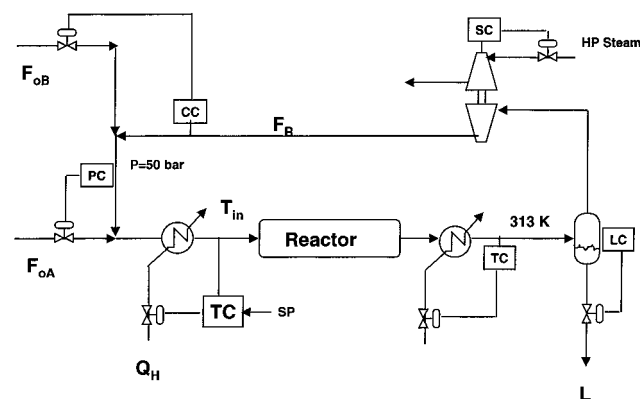


Figure 20. Nonstoichiometric feeds.

sated for by losses of the excess component in the purge gas. Thus, the existence of the purge gas provides some self-regulation to the system.

However, when inert components are not in the feed and there is no purge gas, the imbalance must be handled by having a control structure that detects the buildup or depletion of at least one of the reactants and adjusts fresh feed stream accordingly. Figure 20 illustrates one such scheme. See Tyreus and Luyben⁶ and Luyben et al.⁵ for other control structures that accommodate this situation.

6. Conclusion

In this paper we have studied the steady-state economic design of processes with exothermic reversible reactions, which feature important trade-offs among the reactor size, recycle flowrate, and reactor inlet temperature. The economic impact of inert components in the fresh feed stream is shown to be very strong.

We also have explored several alternative control structures and have developed basic regulatory control structures for both the inert-free and the inert cases. Effective control is obtained in the face of quite large disturbances.

Nomenclature

- A = reactant component
- B = reactant component
- C = product component
- c_{pj} = heat capacity of component j ($\text{kJ kmol}^{-1} \text{K}^{-1}$)

CS1, CS2, CS3, CS4 = control structures
 D_R = reactor diameter (m)
 F = flowrate in the reactor (kmol/s)
 F_C = production rate (kmol of C/s)
 F_j = flowrate of component j (kmol/s)
 F_o = fresh feed flowrate (kmol/s)
 F_{oA} = fresh feed flowrate of reactant A (kmol/s)
 F_{oB} = fresh feed flowrate of reactant B (kmol/s)
 F_P = purge flowrate (kmol/s)
 F_R = recycle flowrate (kmol/s)
 I = inert component
 k_F = specific reaction rate of the forward reaction (kmol/
 $s^{-1} \text{ bar}^{-2} (\text{kg of catalyst})^{-1}$)
 k_B = specific reaction rate of the reverse reaction (kmol/
 $s^{-1} \text{ bar}^{-1} (\text{kg of catalyst})^{-1}$)
 K_c = controller gain
 K_u = ultimate gain
 L = liquid flowrate leaving the separator drum (kmol/s)
 L_R = reactor length (m)
 M_D = holdup in the separator drum (kmol)
 MW_j = molecular weight of component j (kg/kmol)
 NR = number of reactor lumps
 P = total pressure (bar)
 P_j = partial pressure of component j (bar)
 P_1, P_2 = suction and discharge pressures of the compressor
 (bar)
 P_u = ultimate period (radians/s)
 q = catalyst activity parameter
 Q_H = heat transfer rate in the preheater (kJ/s)
 R = perfect gas law constant ($\text{bar m}^3 \text{ kmol}^{-1} \text{ K}^{-1}$)
 \dot{R}_C = rate of production of C (kmol of C/s)
 T = temperature in the reactor (K)
 T_1 = compressor suction temperature (K)
 T_{in} = reactor inlet temperature (K)
 V_{gas} = volume of gas in the reactor (m^3)
 V_{drum} = volume of gas in separator drum (m^3)
 W = compressor work ($\text{kJ s}^{-1} \text{ kmol}^{-1}$)

W_{cat} = mass of the catalyst (kg)
 y_{nj} = composition of component j in lump n (mole fraction
 of component j)
 y_{oj} = composition of the fresh feed (mole fraction of
 component j)
 y_{pj} = composition of the purge gas (mole fraction of
 component j)
 τ_m = measurement of the lag time constant (s)
 τ_I = controller reset time (s)
 ω_u = ultimate frequency (radians/s)
 λ = heat of reaction (kJ/kmol of C produced)
 ρ_{av} = average molar density (kmol/m^3)
 γ = ratio of heat capacities

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