Article

Operability for Processes with Recycles: Interaction between Design and Operation with Application to the Tennessee Eastman Challenge Process

Kwo-Liang Wu, and Cheng-Ching Yu

More About This Article

Additional resources and features associated with this article are available within the HTML version:

- Supporting Information
- Access to high resolution figures
- Links to articles and content related to this article
- Copyright permission to reproduce figures and/or text from this article

View the Full Text HTML

Operability for Processes with Recycles: Interaction between Design and Operation with Application to the Tennessee Eastman Challenge Process

Kwo-Liang Wu and Cheng-Ching Yu*

Department of Chemical Engineering, National Taiwan Institute of Technology, Taipei, Taiwan 106, R.O.C.

In this work, a complex process with recycles, Tennessee Eastman Challenge Process, is studied to explore the operability for plants with recycles. A simplified process model is constructed to provide physical insight into the Eastman plantwide process. The results show that the composition distribution in the reactor (or the recycle flow rate) plays an important role in determining the optimal operating condition, i.e., achieving maximum one-pass conversion. The trajectory of these optimal operating points at different production rates is employed to construct the optimal operating policy (OOP). From the OOP, input multiplicity is observed for the plant with recycles. Therefore, care has to be taken in operating plants with recycles. The optimal operating policy also indicates an inherent constraint on the production rate imposed by the process, e.g., process constraint. However, the design of the control system may lead to an even more limited operability. Moreover, the OOP can be used to evaluate the appropriateness of the designed equipment. The results show that the recycle compressor is not adequately designed and, subsequently, leads to an even smaller operating range. A design procedure is summarized to analyze the process, control, and equipment design aspects of recycle processes. The results indicate that complex recycle processes can be analyzed in a systematic way and, more importantly, all these analyses are based on a rather simple process model in a transparent manner.

1. Introduction

Since the introduction of the Tennessee Eastman Challenge Process (Downs and Vogel, 1993), several academic workers have discussed the control aspects of the process. The Tennessee Eastman (TE) process involves a two-phase reactor, a condenser/separator, a stripper, a gas recycle stream, four fresh feeds, and seven components which fit into the more complex side of the plantwide control problems, e.g., HDA process (Douglas, 1988), Luyben Challenge Process (Luyben and Luyben, 1995), and Vinyl Acetate Process (Luyben, 1996a).

Several authors use the tools in linear multivariable control to analyze the TE process. The multivariable interaction, integral controllability, and disturbance rejection aspects are analyzed by McAvoy and Ye (1994) using the relative gain array (RGA) and Niederlinski index, and multiloop SISO controllers are designed. The robustness of the multivariable TE process is explored by Banerjee and Arkun (1995), and a control configuration design procedure is also proposed. Nonlinear controllers are also developed and tested on the TE process (Ricker and Lee, 1995a; Kanadibhotla and Riggs, 1995). Kanadibhotla and Riggs (1995) point out that a nonlinear compensation, generic model control (GMC), should be used only when necessary. Ricker (1996) compares the nonlinear model predictive control (NMPC) to multiloop SISO controllers, and he concludes that there appears little, if any, advantage to use NMPC in this application. Comparisons are made for different control structures (Luyben, 1996b) using decentralized controllers. Efforts are also made to improve the composition control in the TE process. Nonlinear

modeling and state estimation aspects are studied by Ricker and Lee (1995b). McAvoy et al. (1996) use nonlinear inferential parallel cascade control to provide better disturbance rejection. Georgakis and co-workers provide a tiered framework for the design of plantwide regulatory control structures (Price et al., 1994; Lyman and Georgakis, 1995; Vinson et al., 1995). Qualitative insight of the recycle structure leads to the following procedure: design of the production rate control first, followed by the design of the inventory controls (Price et al., 1994). Most of the studies mentioned deal either with the linearized model or with the complete nonlinear process. Little physical insight is gained from these two levels of abstraction for the TE process.

Ricker (1993), among the first, tries to provide insight into the Tennessee Eastman process. A simplified version of a two-phase reactor is employed to mimic behaviors of the TE process (Ricker, 1993). An optimal operating condition for the TE reactor can be obtained by solving an optimization problem. Recently, optimal steady-state operation of the full-blown TE process is obtained via an optimization procedure, e.g., MINOS 5.1 routine (Ricker, 1995). Actually, the simplified process model offers useful physical insight into complex processes with recycles. For example, the simplified TE reactor of Ricker (1993) enables one to see the effects of process variables from material balances. Douglas (1988) uses a simplified process model to evaluate economic trade-off for process design and steady-state control, and successful applications to several complex plants with recycle are also illustrated. Similar approaches can be seen in Luyben's work (Luyben and Luyben, 1995; Luyben et al., 1996; Luyben, 1996a,b; Lyman and Luyben, 1996) where behaviors of complex recycle plants can be understood via simple models and control strategies are devised accordingly. The operability aspect of a simple recycle process is explored by

---

  * Author to whom all correspondence should be addressed. Telephone: +886-2-737-6620. Fax: +886-2-737-6644. E-mail: ccyu@ch.ntit.edu.tw.
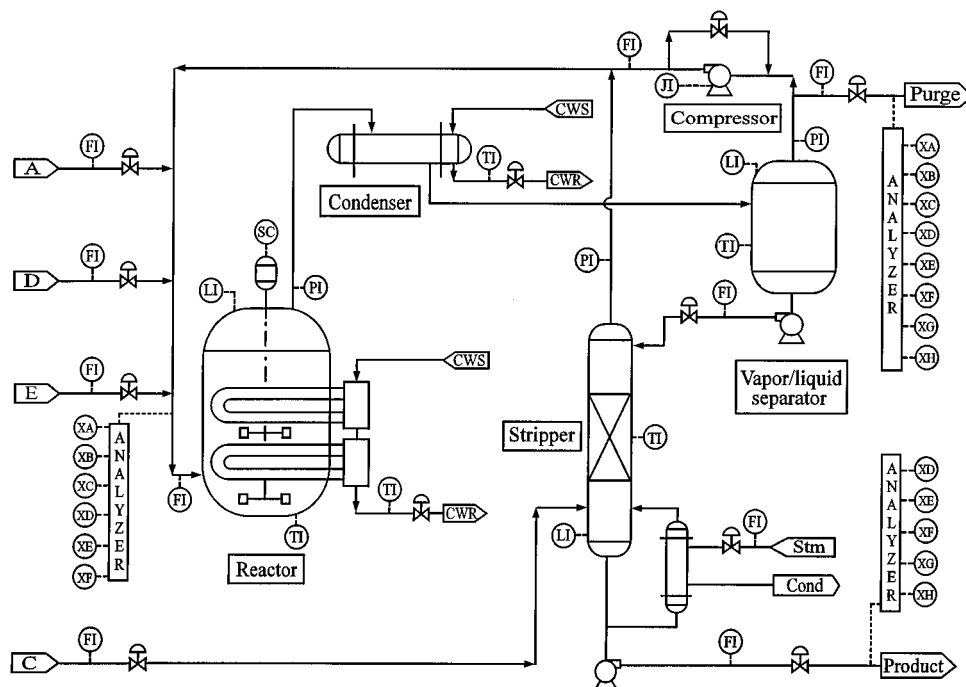
Figure 1. Tennessee Eastman Challenge Process.

Table 1. Material Balance Data (Base Case G/H = 50/50)

Process Stream Data

					stream name	
	A feed	D feed	E feed	C feed	strp ovhd	reactor feed
stream number	1	2	3	4	5	6
molar flow (kg·mol/h)	11.2	114.5	98.0	417.5	465.7	1890.8
temp (°C)	45.0	45.0	45.0	45.0	65.7	86.1
mole fraction						
A	0.999 90	0.000 00	0.000 00	0.485 00	0.432 63	0.321 88
B	0.000 10	0.000 10	0.000 00	0.005 00	0.004 44	0.088 93
C	0.000 00	0.000 00	0.000 00	0.510 00	0.452 64	0.263 83
D	0.000 00	0.999 90	0.000 00	0.000 00	0.001 16	0.068 82
E	0.000 00	0.000 00	0.999 90	0.000 00	0.072 56	0.187 76
F	0.000 00	0.000 00	0.000 10	0.000 00	0.008 85	0.016 57
G	0.000 00	0.000 00	0.000 00	0.000 00	0.019 64	0.035 61
H	0.000 10	0.000 00	0.000 00	0.000 00	0.008 08	0.016 59

			stream name		
	reactor product	recycle	purge	separator underflow	product
stream number	7	8	9	10	11
molar flow (kg·mol/h)	1476.0	1201.5	15.1	259.5	211.3
temp (°C)	120.4	102.9	80.1	80.1	65.7
mole fraction					
A	0.271 64	0.329 58	0.329 58	0.000 00	0.004 79
B	0.113 93	0.138 23	0.138 23	0.000 00	0.000 09
C	0.197 63	0.239 78	0.239 78	0.000 00	0.010 08
D	0.01 075	0.012 57	0.012 57	0.002 22	0.000 18
E	0.177 22	0.185 79	0.185 79	0.137 04	0.008 36
F	0.021 59	0.022 63	0.022 63	0.016 69	0.000 99
G	0.123 02	0.048 44	0.048 44	0.472 69	0.537 24
H	0.084 23	0.000 00	0.000 00	0.311 36	0.438 28

Unit Operation Data

	reactor	separator	stripper
temp (°C)	120.4	80.1	65.7
pressure (kPa gauge)	2705.0	2633.7	3102.2

Wu and Yu (1996), and the production rate changes are handled using a balanced control configuration. Again, simplified models offer useful information for the design of control systems.

The objective of this work is to study the operability of the Tennessee Eastman Challenge Process. The remainder of this paper is organized as follows. A simplified TE process is constructed in section 2 and process characteristics are also explored. Section 3 addresses the issues of optimal operations at the nominal production rate, and the effects of the composition distribution and physical variables are explored at different production rates. In section 4, constraints resulting from process, control, and equipment are
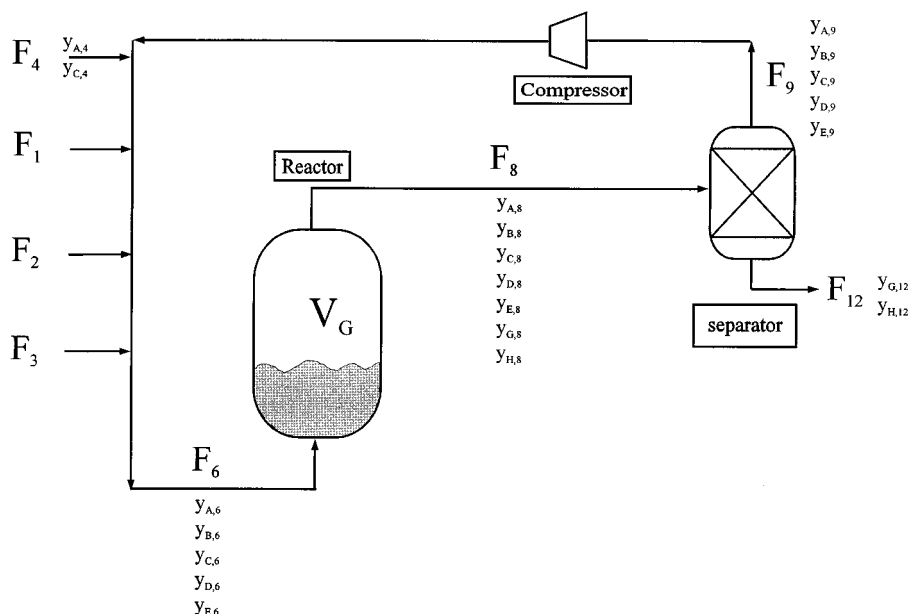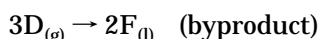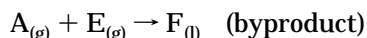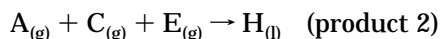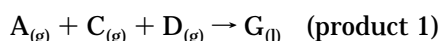
Figure 2. Simplified Tennessee Eastman process.

studied as the production rate is varied. Section 5 investigates the effect of the reactor temperature and pressure on the optimal operating policy. A design procedure is summarized in section 6 for the design and optimal operating policy for plants with recycles. Conclusions are drawn from this study in section 7.

2. Process Description

The TE problem is based on a Tennessee Eastman process (Downs and Vogel, 1993). The process involves five unit operations: a two-phase reactor, a condenser, a separator, a stripper, and a recycle compressor (Figure 1). The reactor is a CSTR in which the exothermic and irreversible reactions occur.

$$A_{(g)} + C_{(g)} + D_{(g)} \rightarrow G_{(l)} \quad \text{(product 1)}$$

$$A_{(g)} + C_{(g)} + E_{(g)} \rightarrow H_{(l)} \quad \text{(product 2)}$$

$$A_{(g)} + E_{(g)} \rightarrow F_{(l)} \quad \text{(byproduct)}$$

$$3D_{(g)} \rightarrow 2F_{(l)} \quad \text{(byproduct)}$$

These are gas-phase reactions with liquid products. We are interested in the operability, i.e., production rate changes of the products G and H, of the TE process. Also notice that an inert component B makes up 0.5% of feed stream 4, and it must be purged from the system. Table 1 gives the steady-state material balance data for the base case (G/H = 50/50).

2.1. Simplified Process Model. In order to understand the effects of process variables, e.g., reactor temperature (T), pressure (P), vapor holdup (V_G), on the operation of the TE process, several assumptions are made. One principle employed is that the composition distribution in the reactor and the ratio of recycle flow to the product flow should be as close to the nominal condition as possible. In the reactor, only two major reactions are considered, and as a result of smaller rate constants, the byproduct and the reactions associated with the byproduct are eliminated. Since products G and H are the heavier components, in the separation section, only one process unit is employed to separate the products from the light components, i.e., A, B, C,

D, and E. Therefore, the feed stream C (F_4 in Figure 2) is introduced directly to the reactor, and the stripper is eliminated. In order to match the composition distribution in the reactor, the presence of the inert component B in the reactor is assumed but the purge stream and the trace amount of B in the feed are not taken into consideration. In summary, the following assumptions are made to obtain a simplified process model: (1) byproduct F and byproduct reactions are not considered, (2) the separator and stripper are lumped together as a single separation unit, and (3) the purge stream is eliminated and a trace amount of B in the feed is not considered. It should be emphasized that in this work we are more interested in the inherent process characteristics, and the simplified model presented here is capable of providing a qualitative process description of the TE process.

Therefore, we arrive at the following equations describing the material balances:

$$F_1 + F_4 y_{A,4} = r_1 + r_2 \tag{1}$$

$$F_4 y_{C,4} = r_1 + r_2 \tag{2}$$

$$F_2 = r_1 \tag{3}$$

$$F_3 = r_2 \tag{4}$$

$$F_{12} y_{G,12} = r_1 \tag{5}$$

$$F_{12} y_{H,12} = r_2 \tag{6}$$

where r_1 and r_2 are the rates of reactions forming G and H, respectively. The reaction rates of the gas-phase reactions are (Downs and Vogel, 1993)

$$r_1 = k_1(T) \, P^{2.5279} V_G y_{A,8}^{1.1544} y_{C,8}^{0.3735} y_{D,8} \tag{7}$$

$$r_2 = k_2(T) \, P^{2.5279} V_G y_{A,8}^{1.1544} y_{C,8}^{0.3735} y_{E,8} \tag{8}$$

where k_1 and k_2 are the rate constants which follow the Arrhenius expression, P is the reactor pressure, V_G denotes the vapor volume of the reactor, and $y_{i,8}$ is the vapor phase mole fraction of component i (Figure 2).

From the overall balances (eqs 1−6), it immediately becomes clear that the reaction rates play a deciding role for the production rate changes and product distribution. Since the reaction rates are determined by the temperature (T), pressure (P), vapor volume (V_G), and composition distributions of the reactants (e.g., $y_{A,8}$, $y_{C,8}$, $y_{D,8}$, and $y_{E,8}$), the importance of the two-phase reactor in operating the TE process then becomes clear.

As far as the process operation is concerned, several parameters can be defined to characterize the operating condition.

(1) Production rate is described by a dimensionless parameter, throughput factor:

$$\eta_{TP} = \frac{\text{production rate}}{\text{nominal production rate}} = \frac{\mathcal{R}}{\bar{\mathcal{R}}} \qquad (9)$$

where $\mathcal{R}$ is the production rate ($\mathcal{R} = r_1 + r_2$) and $\bar{\mathcal{R}}$ denotes the nominal production rate (i.e., $\bar{\mathcal{R}} = \bar{r}_1 + \bar{r}_2$ and the overbar stands for the nominal value).

(2) The product distribution is defined as

$$\kappa = \frac{\text{production rate of H}}{\text{production rate of G}} = \frac{y_{H,12}}{y_{G,12}} \qquad (10)$$

and the product distribution varies from 0.1 (G/H = 90/10) to 0.9 (G/H = 10/90) as described by Downs and Vogel (1993).

(3) The recycle ratio denotes the ratio of the recycle flow rate (e.g., F_9) to the production rate (e.g., F_{12}):

$$RR = \frac{\text{recycle flow rate}}{\text{product flow rate}} = \frac{F_9}{F_{12}} \qquad (11)$$

Following these definitions, the production rate can be expressed as

$$\eta_{TP}\bar{\mathcal{R}} = \mathcal{R} = r_1 + r_2 \qquad (12)$$

From eqs 5 and 6 and the definition of κ, we have

$$\eta_{TP}\bar{\mathcal{R}} = (1 + \kappa)r_1 \qquad (13)$$

Substituting eq 7 into eq 13, one obtains

$$\eta_{TP}\bar{\mathcal{R}} = (1 + \kappa)k_1 P^{2.5279} V_G y_{A,8}{}^{1.1544} y_{C,8}{}^{0.3735} y_{D,8} \quad (14)$$

where $y_{i,8}$ represents the mole fraction of component i in the vapor phase of the reactor.

Similar to the approach of Douglas (1988), substitutions will be made such that the production rate can be expressed in terms of variables relevant to the process operation. Material balances around the separator in Figure 2 gives

$$F_8 = F_9 + F_{12} \qquad (15)$$

$$F_8(y_{G,8} + y_{H,8}) = F_{12} \qquad (16)$$

From the definitions of RR and κ, the vapor-phase composition of G at the reactor outlet ($y_{G,8}$) then becomes

$$y_{G,8} = \frac{1}{1 + \kappa} \frac{1}{1 + RR} \qquad (17)$$

and $y_{H,8}$ can also be obtained immediately following eq 17:

$$y_{H,8} = \frac{\kappa}{1 + \kappa} \frac{1}{1 + RR} \qquad (18)$$

Equations 17 and 18 give explicit expressions for the mole fractions of G and H in the reactor outlet. Since this is a two-phase reactor with noncondensable components A, B, and C, the liquid-phase composition profile in the reactor gives

$$x_{D,8} + x_{E,8} + x_{G,8} + x_{H,8} = 1 \qquad (19)$$

where $x_{i,8}$ is the mole fraction of component i in the liquid phase of the reactor. The liquid-phase composition can be eliminated from the vapor−liquid equilibrium (VLE) relationship which follows Raoult's law (Downs and Vogel, 1993):

$$\frac{P}{P_D^y(T)}y_{D,8} + \frac{P}{P_E^y(T)}y_{E,8} + \frac{P}{P_G^y(T)}y_{G,8} + \frac{P}{P_H^y(T)}y_{H,8} = 1 \qquad (20)$$

where $P_i^y(T)$ is the vapor pressure of component i and the Antoine equations for the vapor pressure are given by Downs and Vogel (1993). Comparing eqs 7 and 8 and using the definition of κ, we have

$$\frac{y_{E,8}}{y_{D,8}} = \kappa\frac{k_1(T)}{k_2(T)} \qquad (21)$$

Substituting eqs 17, 18, and 21 into eq 20, we obtain

$$y_{D,8} = \frac{\dfrac{1}{P} - \left(\dfrac{1}{P_G^y(T)} + \dfrac{\kappa}{P_H^y(T)}\right)\dfrac{1}{(1 + \kappa)(1 + RR)}}{\left(\dfrac{1}{P_D^y(T)} + \dfrac{\kappa k_1}{P_E^y(T)\, k_2}\right)} \qquad (22)$$

Equation 22 shows that $y_{D,8}$ (or $y_{E,8}$) can be expressed explicitly as a function of κ, RR, T, and P. Material balances around the separator give

$$\frac{RR}{1 + RR} = y_{A,8} + y_{B,8} + y_{C,8} + y_{D,8} + y_{E,8} \qquad (23)$$

After substituting eqs 21 and 22 into eq 23, $y_{C,8}$ can be represented as

$$y_{C,8} = \frac{RR}{1 + RR} - y_{A,8} - y_{B,8} - \\ \left(\frac{k_2 + \kappa k_1}{k_2}\right)\left[\frac{\dfrac{1}{P} - \left(\dfrac{1}{P_G^y(T)} + \dfrac{\kappa}{P_H^y(T)}\right)\dfrac{1}{(1 + \kappa)(1 + RR)}}{\left(\dfrac{1}{P_D^y(T)} + \dfrac{\kappa k_1}{P_E^y(T)\, k_2}\right)}\right] \qquad (24)$$

Since $y_{B,8}$ is assumed constant in the simplified model (i.e., $y_{B,8} = \bar{y}_{B,8}$), the additional variable in eq 24 is the vapor-phase composition of A in the reactor. Therefore, the production rate for the TE process (eq 14) now can be expressed in terms of relevant process variables:
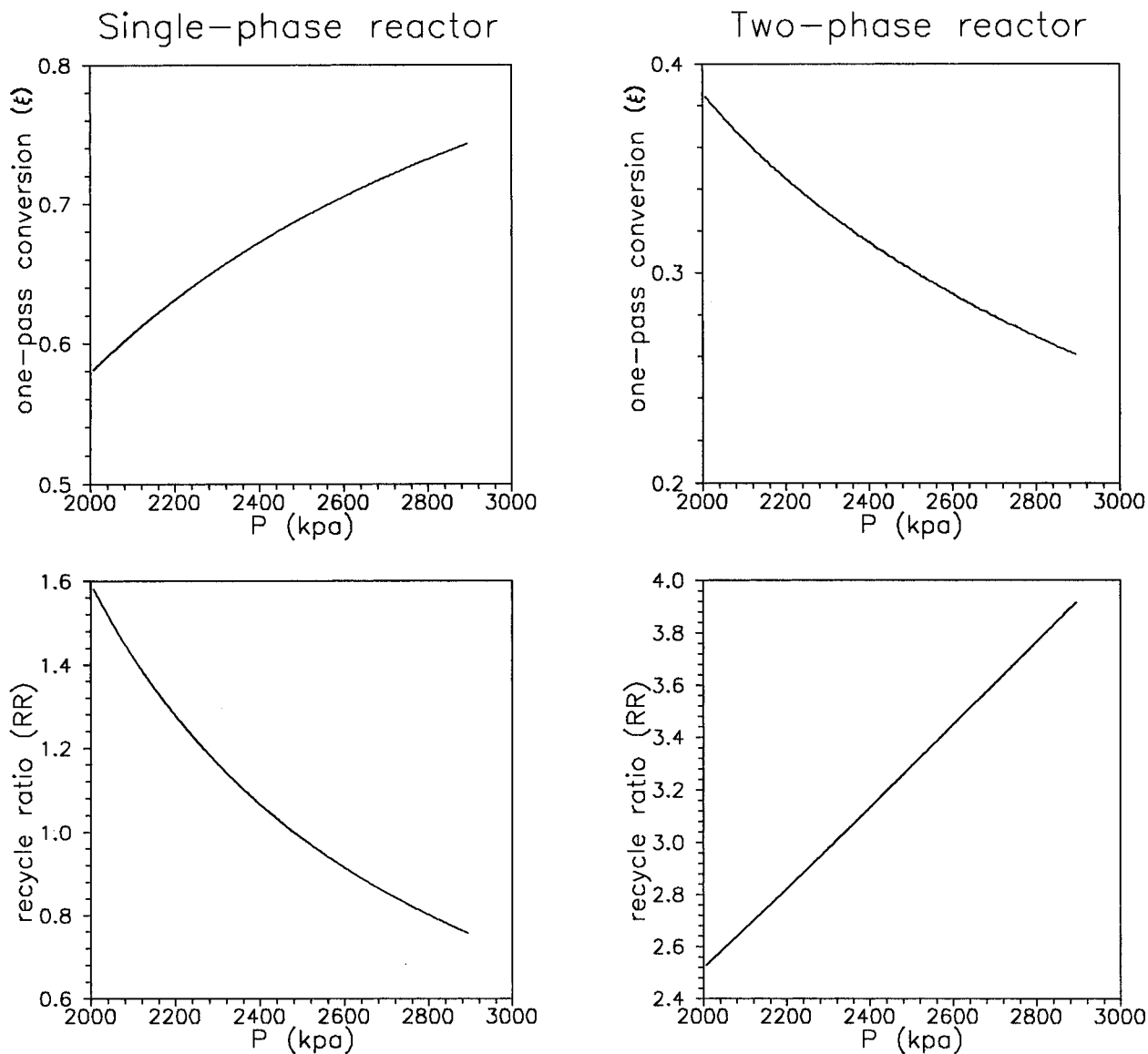
Single-phase reactor

Two-phase reactor

Figure 3. Effects of pressure on the behavior of single-phase and two-phase reactors in the recycle process.

$$\eta_{TP} = \frac{1 + \kappa}{\mathcal{R}} k_1 P^{2.5279} V_G y_{A,8}^{1.1544} \left\{ \frac{RR}{1 + RR} - \right.$$

$$y_{A,8} - \bar{y}_{B,8} - \left(\frac{k_2 + \kappa k_1}{k_2}\right) \times$$

$$\left. \left[\frac{\frac{1}{P} - \left(\frac{1}{P_G^y(T)} + \frac{\kappa}{P_H^y(T)}\right)\frac{1}{(1 + \kappa)(1 + RR)}}{\left(\frac{1}{P_D^y(T)} + \frac{\kappa k_1}{P_E^y(T)\ k_2}\right)} \right]^{0.3735} \right\} \times$$

$$\left[\frac{\frac{1}{P} - \left(\frac{1}{P_G^y(T)} + \frac{\kappa}{P_H^y(T)}\right)\frac{1}{(1 + \kappa)(1 + RR)}}{\left(\frac{1}{P_D^y(T)} + \frac{\kappa k_1}{P_E^y(T)\ k_2}\right)} \right] \quad (25)$$

In this expression, $y_{A,8}$ is the only composition variable ($y_{B,8}$ is assumed constant). Equation 25 shows the

relationship among four process variables T, P, V_G, and $y_{A,8}$ and one operating parameter RR, which gives the basis for the understanding of the steady-state behavior of the TE process. Notice that we have 5 degrees of freedom for the case of variable production rate.

2.2. Unique Process Characteristic. The two-phase reactor of the TE process behaves differently from a typical single-phase reactor. It is a common understanding that the conversion increases as we increase the reactor holdup. Unfortunately, this is not the case for the TE reactor. Comparisons are made between a single-phase reactor and the TE two-phase reactor.

Assume a single-phase reactor with the same temperature, pressure, and reaction kinetics except that the product is formed in the vapor phase, i.e., $G_{(g)}$ and $H_{(g)}$. It is also assumed that the volume of this single-phase reactor V_G is the same as the vapor volume of the TE reactor. The process flowsheet is the same as Figure 2. Notice that, without the VLE constraint, the case of the single-phase reactor has one more (6) degrees of freedom (i.e., one less equation).

Consider the case when we increase the reactor pressure. Intuitively, for the single-phase reactor, as the molar holdup in the reactor increases (as a result

of pressure increase), we can have a better one-pass conversion. That also implies a smaller recycle flow. These are exactly the results shown in Figure 3 for the single-phase reactor. However, the opposite behavior is observed for the case of the two-phase TE reactor. Since the reactor pressure affects the vapor holdup as well as the composition profile in the reactor (e.g., eq 20), the VLE constraint in the two-phase reactor may result in wrong-way behavior. Figure 3 clearly indicates that the TE reactor shows a unique process characteristic. That is, a better one-pass conversion can be obtained as we lower the reactor pressure. This unusual characteristic is quite desirable from an operation point of view, since a higher pressure means higher equipment investment and higher operating cost (e.g., compressor horsepower). Furthermore, a smaller recycle flow rate is needed for the same production rate (Figure 3). This interesting result can be derived from the simplified process model (eq 25) by looking at the relationships between the reactor pressure and the optimized (giving the smallest RR) composition of A.

3. Process Analyses—Nominal Production Rate

The on-going analyses show that the complex TE process can be understood physically. Before getting into the issue of operability, the nominal steady state is investigated. By nominal condition we mean that the production rate is at 100% (i.e., $\eta_{TP} = 1$) with a specified product distribution (i.e., a given κ). At the nominal production rate, $\eta_{TP} = 1$, with the assumption that the composition of the inert is constant, we have four independent variables left (e.g., as can be seen from eq 25). They can be chosen as one composition variable, e.g., chosen from $y_{A,8}$, $y_{C,8}$, $y_{D,8}$, ..., plus three physical variables, e.g., T, P, V_G. In order to explore the effect of different types of variables on the process operation, the contribution to the production rate $\mathcal{R}$ can be classified into the effect of composition variables ($\mathcal{R}_y$) and the effect of physical variables ($\mathcal{R}_p$). That is

$$\mathcal{R} = [(1 + \kappa)k_1(T) P^{2.5279} V_G][y_A^{1.1544} y_C^{0.3735} y_D]$$
$$= \mathcal{R}_p \mathcal{R}_y \tag{26}$$

$\mathcal{R}_y$ represents the contribution of the composition distribution to the production rate, and $\mathcal{R}_p$ shows the effects of temperature (T), pressure (P), and reactor volume (V_G) on the production. With the definitions of $\mathcal{R}_p$ and $\mathcal{R}_y$, a step-by-step approach is taken in steady-state analyses.

3.1. Composition Distribution. From the reaction rate expressions (eqs 7 and 8), it is clear that the distribution of the reactants can affect the production rate. At the nominal condition, however, it may change the flow distribution in this recycle structure. In the TE process, if the other 3 degrees of freedom are chosen as physical variables, i.e., T, P, and V_G, we have only one independent variable for the composition. This composition can be of any component except B. In this work, $y_{A,8}$ is selected to represent the composition distribution (e.g., eq 25). Mathematically, we would like to investigate the relationship between $y_{A,8}$ and the internal flow rate for a given $\mathcal{R}_y$. Figure 4 shows the relationship between the recycle ratio and the composition distribution, $y_{A,8}$. Several observations can be made immediately from Figure 4. First, there is a limited range of $y_{A,8}$ (or RR) to meet the specifications on the production rate and product quality. Second, the nomi-
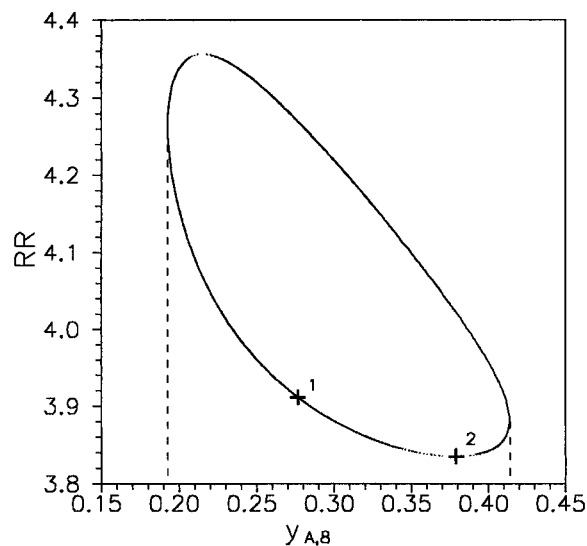

Figure 4. Relationship between $y_{A,8}$ and RR at the nominal production rate with constant T, P, and V_G.

nal steady state given in the original literature (point 1 in Figure 4) is not at its optimal point (point 2 in Figure 4). By optimal we mean the operating point gives the lowest recycle ratio (RR) at the given T, P, and V_G. It should be emphasized that all computations and implications made here are based on a simplified model, and, despite the effort made to mimic the original system, they are only an approximation. The third observation is that input multiplicities can be found for the TE process. It is not new to find the existence of the input multiplicity in chemical reactors (Aris and Amundson, 1958; Perlmutter, 1965) or in recycle processes (Tyreus and Luyben, 1993; Luyben et al., 1996). The input multiplicity observed here is associated with the reaction kinetics (e.g., eq 26). In other words, different composition distributions may lead to the same value of $\mathcal{R}_y$ and, subsequently, result in the same production rate. It is important to point out that different steady states may lead to different operating costs. Notice that the operating cost can be inferred from the recycle ratio since the recycle flow rate is associated with compressor work and heat input to the system. In this case one would like to operate at the steady state which gives a smaller recycle ratio.

3.2. Effects of Physical Variables. In addition to the composition distribution, the physical terms (or $\mathcal{R}_p$) may also affect the internal flows. Here, a constant production rate is assumed, and the effects of reactor temperature (T), reactor pressure (P), and vapor volume (V_G) are explored. As we increase the temperature, the recycle flow rate (or the recycle ratio RR) decreases as shown in Figure 5a. This coincides with one's intuition. However, for an increase in the reactor pressure, the recycle ratio also increases (Figure 5b). As mentioned earlier, this is a unique characteristic of the TE process, since one would expect a decrease in RR as the molar holdup in the reactor (e.g., reactor level for a liquid-phase reaction) increases. However, as we increase the vapor volume (i.e., decrease the liquid level) in the reactor, the recycle ratio also decreases accordingly as shown in Figure 5c. This is, again, within one's expectation. Therefore, care should be taken when one changes the reactor pressure and the behaviors of temperature and vapor volume are similar to that of the typical single-phase reactor.

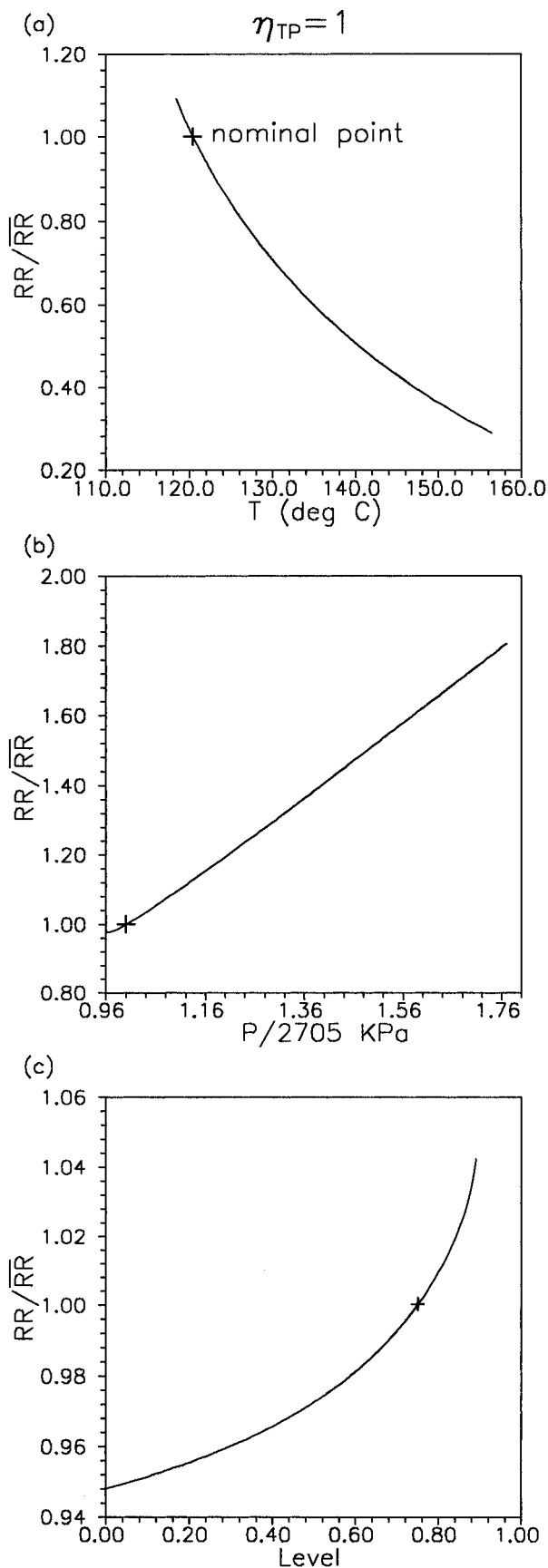Actually, the relationship between RR and P can be

Figure 5. Effects of reactor temperature (top), pressure (middle), and liquid level (bottoms) on the recycle ratio at the nominal production rate with constant reactor composition.

$$RR = \frac{\left(\dfrac{1}{P_G^y(T)} + \dfrac{\kappa}{P_H^y(T)}\right)}{\left[\dfrac{1}{P} - \left(\dfrac{1}{P_D^y(T)} + \dfrac{k_1\kappa}{k_2 P_E^y(T)}\right)\bar{y}_{D,8}\right](1 + \kappa)} \quad (27)$$

Equation 27 clearly indicates the unusual behavior between P and RR.

4. Process Analyses—Production Rate Changes

Let us examine the case when the production rate is subject to change (i.e., $\eta_{TP} \neq 1$). This leads to another independent variable, and, quite often, the throughput manipulator is chosen as an external flow rate (Luyben, 1996b). Thus, in this study, we simply use η_{TP} (eq 25) to represent the throughput manipulator, and this leads us again to four more independent variables (e.g., same number as the nominal condition). The three physical variables, T, P, and V_G, are assumed constant for the moment, and the relationships between the composition distribution and recycle ratio will be studied for the case of variable production rate. This assumption will be relaxed in the next section.

4.1. Process Constraint-Inherent Limitation. From the material balances point of view, we cannot observe any production rate limitation for the separator, but the chemical reactor, on the other hand, does show a limit on the overall conversion. Therefore, it is within expectation that we will see a production rate limit for the reactor/separator process. This is exactly what happens to the TE process.

For the simplified TE process, if the fourth degree of freedom (e.g., the control degrees of freedom as termed by Luyben) is chosen as the recycle ratio RR (the first three are T, P, and V_G), we can explore the production rate limitation as well as the composition distribution. If the flow ratio RR is controlled at 95% of the original value, we can calculate $y_{A,8}$ for a given η_{TP} and vice versa (e.g., eq 25). The results (the rightmost curve in Figure 6) indicate that, indeed, there is an upper bound for the throughput factor η_{TP} and, again, multiple steady states are observed. It is interesting to note that, for the case of $RR/\overline{RR} = 0.95$, the maximum production rate is only 75% of the designed value (Figure 6). This result comes from the unfortunate choice of the set point for the recycle ratio. Therefore, one way to raise the throughput factor is to change the set point of RR. Figure 6 shows that the upper bound of η_{TP} increases initially as we increase the set point of the recycle ratio. However, a further increase in the recycle ratio (e.g., $RR/\overline{RR} > 1.04$) results in a decrease in the maximum production rate. Again, this is a result of the input multiplicity, as we mentioned earlier.

In order to achieve the highest possible production rate, one can fix the set point for the recycle ratio at 1.04 (Figure 6). However, this operating policy is not desirable since the recycle cost is a part of the operating cost (Douglas, 1988) and the plant can be operated with a smaller recycle cost (i.e., a smaller RR) as the production rate changes. Thus, one way to maintain the lowest recycle cost at various production rates is to follow the maximum trajectory of the $RR/\overline{RR}$ contour. This optimal trajectory is indicated by the solid circles in Figure 6, and the open circles in Figure 6 indicate the infeasible region of the trajectory. This trajectory is termed the optimal operating policy (OOP) hereafter. Figure 7a shows the optimal operating policy when the
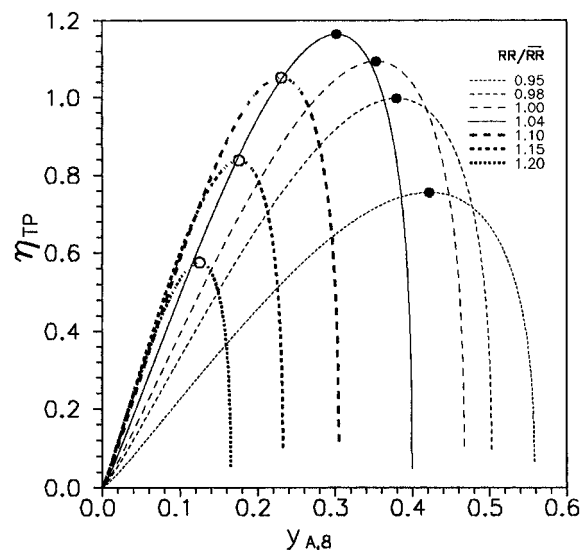
derived analytically. For $\eta_{TP} = 1$ and a given composition distribution, eq 25 can be rearranged to give

Figure 6. Relationship between throughput factor (η_{TP}) and reactor composition ($y_{A,8}$) for constant recycle ratio (RR) operation.

recycle ratio is chosen as the controlled variable. Similar results (Figure 7b) can be obtained if one prefers to control the composition $y_{A,8}$. The OOP's (e.g., Figure 7a,b) serve as the basis for the set point selection, control structure design, and equipment design, as will be shown later.

Figure 7a indicates that, while keeping T, P, and V_G constant, the maximum production rate that can be achieved is 116% of the design value (i.e., $\eta_{TP,max} = 1.16$). This is an inherent limitation of the process: process constraint. However, one should not devise a control structure, give a set point, or design a piece of equipment which fail to meet this upper bound. Therefore, for the production rate change, one should move along the left-hand side of the hill by adjusting the set point of RR (Figure 7a). It is also observed that the original steady state (the cross in Figure 7a) is a little off from the optimal RR. Similar operating policy can be constructed (Figure 7b) if the control of $y_{A,8}$ is preferred.

4.2. Control Structure Constraint. As pointed out earlier, the OOP can be achieved by adjusting the set point of the controlled variable as the production rate changes. In plant operation, however, the set points are hardly changed. Here, let us explore how this practice will affect the already limited operability.

In the simplified TE process, we have five control degrees of freedom. Three of them are the physical variables, T, P, and V_G and one is the throughput manipulator (η_{TP}). This leaves us with one more variable to be controlled. This additional variable can be a composition (e.g., $y_{A,8}$, $y_{C,8}$, $y_{D,8}$, or $y_{G,8}$), a flow rate (e.g., F_9), or a flow ratio (e.g., RR). For the compositions, it seems we have four candidate controlled variables, i.e., $y_{A,8}$, $y_{C,8}$, $y_{D,8}$, or $y_{G,8}$. However, eqs 17 and 22 indicate that controlling $y_{D,8}$, or $y_{G,8}$ is equivalent to controlling RR. Therefore, we have four distinctly different control structures, i.e., using $y_{A,8}$, $y_{C,8}$, RR, and F_9 as the additional controlled variable (Table 3). Here, we use the nominal steady-state value (Table 2) as the set point for the controlled variable. The results (Figure 8) show that the selected control structures impose an even more severe constraint on the operability. For example, if F_9 or $y_{C,8}$ is controlled at its set point, the TE process can only provide a little more than 1% increase in the production rate. However, if $y_{A,8}$ is controlled, the
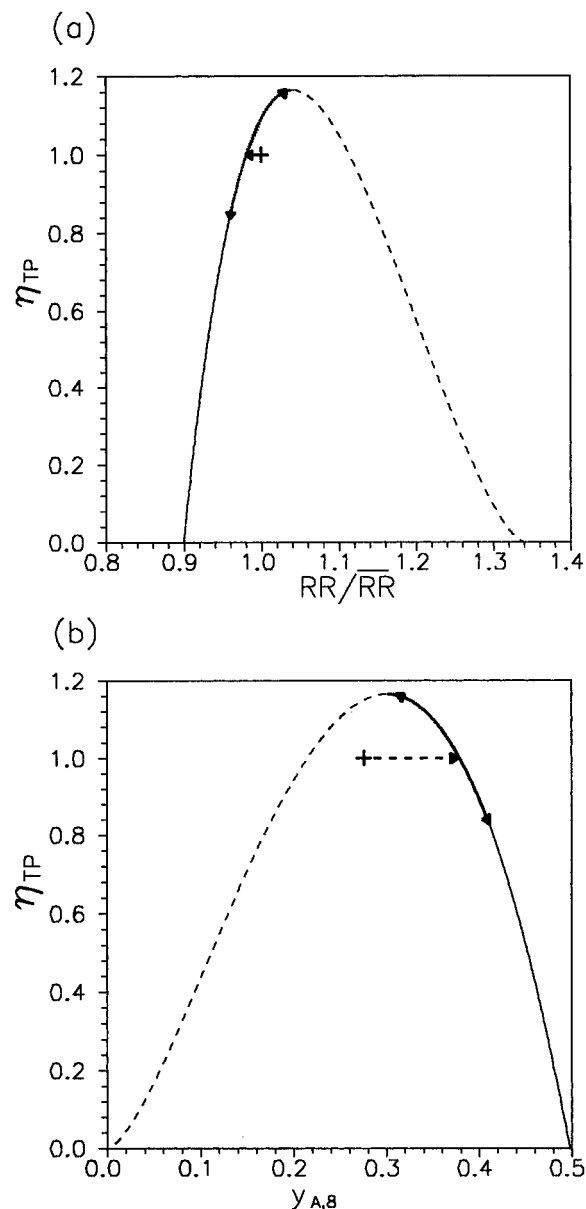

Figure 7. Trajectories of optimal operating policy (OOP) showing relationships between (a) η_{TP} and RR and (b) η_{TP} and $y_{A,8}$.

limitation on the throughput increase is fairly close to the inherent process constraint. In principle, this is the control strategy employed by Ricker (1996) and Luyben (1996b), where both authors are successful in choosing the best composition variable (y_A) to be controlled. Also notice that different manipulated inputs and types of controllers are used by Ricker and Luyben. Table 3 gives the upper and lower bounds for these four control structures. Therefore, extreme care has to be taken in designing and operating the control system for recycle processes. Moreover, the conventional constant set-point practice has to be modified in order to achieve a better operability.

4.3. Equipment Constraint. In the original TE process (Figure 1), there are several pieces of equipment that may run into constraints as the production rate increases. An obvious piece of equipment is the gas recycle compressor, and the other two possibilities are the cooling bundle in the reactor and the reboiler of the stripper. Since the stripper is not modeled explicitly, possible constraint in the reboiler design is not discussed
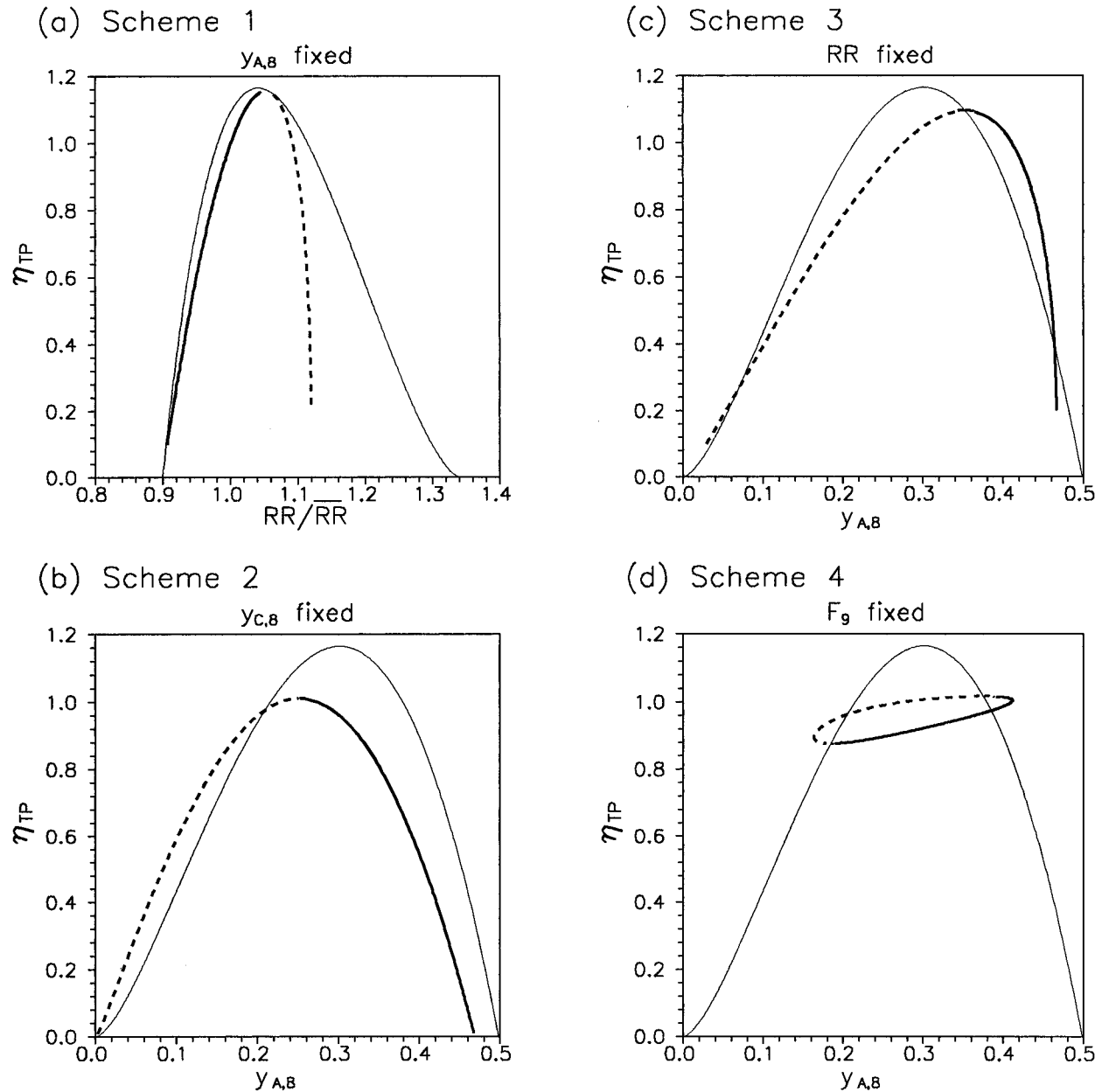
Figure 8. Comparisons between OOP and contour of selected control structures: (a) $y_{A,8}$ fixed, (b) $y_{C,8}$ fixed, (c) RR fixed, and (d) F_9 fixed.

here. The heat removal rate in the reactor depends on the liquid level and the agitator speed. If the liquid level in the reactor is above 50%, we cannot observe any constraint violation for the production rate of interest. In other words, the heat removal capacity of the cooling bundle is well above the heat generation as long as the liquid level is kept above 50%. However, the heat removal rate decreases as the reactor level drops below 50%. Figure 9 shows the heat generation and heat removal curves at the nominal production rate as the reactor level varies. Furthermore, if the level drops below 10%, the reactions will run away.

The equipment which is most likely to run into constraint is the recycle compressor. In order to model the flow capacity of the compressor, pressure drops are assumed (Downs and Vogel, 1993). Following Downs and Vogel, the pressure drop is proportional to the square of the corresponding flow rate and a vapor space is assumed at the outlet of the compressor (Figure 10). Therefore, the pressures in the separator (P_s) and the vapor space (P_v) can be calculated once the reactor

pressure (P) and flow rates are known. Define the pressure ratio

$$P_r = P_v/P_s \tag{28}$$

The maximum flow capacity of the compressor (F_{cp}) can be expressed as (assuming the spill-back valve is closed)

$$F_{cp} = F_{max} + \frac{F_{max}}{1.197}(1 - P_r^3) \tag{29}$$

where F_{max} is a constant with the value of 4957 kg·mol/h (Downs and Vogel, 1993). Equation 29 gives the characteristic of the recycle compressor. Therefore, provided with the flow rates and reactor pressure, F_{cp} can be computed immediately. It should be noticed that the compressor characteristic is not a flat curve, since the pressure ratio increases as we increase the throughput (Figure 11). On the other hand, the desired recycle flow (F_9) at different production rates can be represented
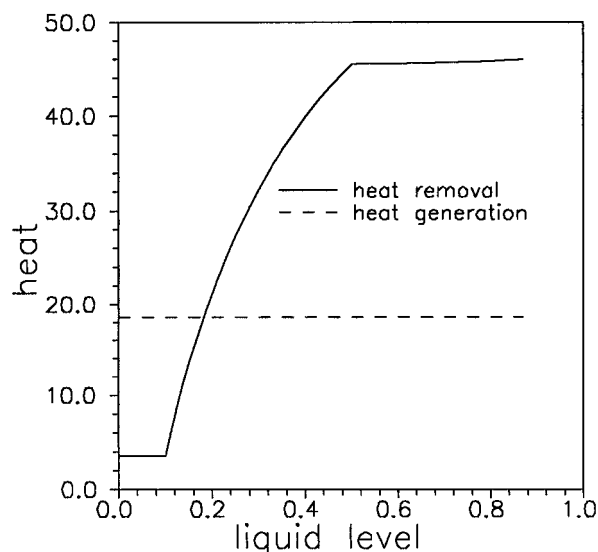
Figure 9. Effect of reactor liquid level on heat removal at the nominal production rate.

Table 2. Steady-State Values of the Simplified Model

	stream name			
	A feed	D feed	E feed	A/C feed
stream number	F_1	F_2	F_3	F_4
molar flow (kg·mol/h)	16.69	169.11	137.98	597.48
mole fraction				
A	1.0000	0.0000	0.0000	0.4860
B	0.0000	0.0000	0.0000	0.0000
C	0.0000	0.0000	0.0000	0.5140
D	0.0000	1.0000	0.0000	0.0000
E	0.0000	0.0000	1.0000	0.0000
G	0.0000	0.0000	0.0000	0.0000
H	0.0000	0.0000	0.0000	0.0000

	stream name			
	reactor feed	reactor product	recycle	product
stream number	F_6	F_8	F_9	F_{12}
molar flow (kg·mol/h)	2122.267	1508.089	1201.0	307.09
mole fraction				
A	0.340 85	0.276 03	0.346 62	0.000 00
B	0.085 08	0.119 73	0.150 34	0.000 00
C	0.280 21	0.190 71	0.239 47	0.000 00
D	0.088 22	0.012 00	0.015 07	0.000 00
E	0.205 64	0.197 90	0.248 50	0.000 00
G	0.000 00	0.112 14	0.000 00	0.550 70
H	0.000 00	0.091 49	0.000 00	0.449 30

Table 3. Maximum and Minimum Throughput Factor for Four Different Control Structures

structure	controlled variable	$\eta_{TP,max}$	$\eta_{TP,max}$
1	$T, P, V_G, \eta_{TP}, y_{A,8}$	1.160	0.000
2	$T, P, V_G, \eta_{TP}, y_{C,8}$	1.011	0.000
3	T, P, V_G, η_{TP}, RR	1.094	0.000
4	$T, P, V_G, \eta_{TP}, F_9$	1.017	0.876

as (eqs 9 and 10)

$$F_9 = \eta_{TP}RR\bar{F}_{12} \qquad (30)$$

Obviously, the design of the equipment can constrain the operability of the TE process. Figure 11 shows the maximum achievable throughput ratio imposed by the compressor. It is also true that one can move the constraint upward (in Figure 11) by installing a compressor with a larger horsepower.

Here, we would like to see whether the installed TE recycle compressor will further limit the production rate.

Consider the optimal operating policy (Figure 7). Once the reactor pressure and corresponding flow rate are known, we can solve for F_{cp} and then the maximum achievable RR. Figure 12 shows that the compressor constraint (the solid line) prevents us from achieving the maximum production rate set by the process. Unless the recycle flow can be decrease by other means, the recycle gas compressor imposed an undesirable constraint on process operability. Figure 12 reveals that the issue between equipment design and process operation has to be dealt with. Despite the fact that the design problem is not discussed in this work, the proposed OOP offers a guideline for sizing process equipments.

5. Achieving Maximum Production Rate

The on-going analyses deal with cases where the composition distribution in the reactor is rearranged to achieve the maximum throughput factor. In this section, the more obvious variables, e.g., T, P, and V_G, for the optimization are studied.

5.1. Changing Reactor Temperature. In theory, the reactor temperature is one of the most effective variables in handling the production rate changes (eq 25). It should be emphasized, however, that there is a limited range where the reactor temperature can be varied, and in this case the high-temperature limit is 150 °C, as pointed out by Downs and Vogel (1993). Figure 12 shows how the optimal operating policy (OOP) changes with the reactor temperature. At the nominal reactor temperature only a 16% increase in the production rate is possible, but for a 10 °C increase in the temperature, the maximum achievable production rate change is +79% (e.g., point 5 in Figure 12).

In theory, one can operate the plant along the loci of OOP's (the dashed line consisting of points 5, 4, and 3 in Figure 12) as different production rates are set. Again, the capacity of the recycle compressor limits the operability of the TE process, as shown in Figure 12. Instead of achieving a 79% increase in the production rate, the throughput increase is limited to 43% (Figure 12) in spite of a rather small recycle ratio (e.g., RR/$\overline{RR}$ = 0.77). Furthermore, if we choose to reset the reactor temperature as the production rate changes, the operating policy now becomes moving along the line imposed by the equipment constraint.

5.2. Changing Reactor Pressure. Unlike the reactor temperature, the reactor pressure has a less profound effect on the production rate changes. Moreover, as mentioned earlier, for a given production rate, the recycle ratio (RR) decreases as we lower the reactor pressure. This has an important implication, since we can easily encounter the recycle flow rate limitation in the TE process.

Figure 13 shows OOP's when the reactor pressure is changed. Similar to the observation made earlier, Figure 13 reveals that a lower reactor pressure results in a smaller RR. However, we can achieve a higher maximum production rate (loci of solid circles in Figure 13) by raising the reactor pressure. Comparison between Figures 12 and 13 shows that the reactor pressure behaves differently from the reactor temperature. The effects of temperature indicate that a higher production rate comes with a smaller RR by increasing the reactor temperature, and, on the other hand, the effects of pressure reveal that a higher throughput factor goes along with a higher RR as the reactor pressure is increased. Therefore, it is not desirable to
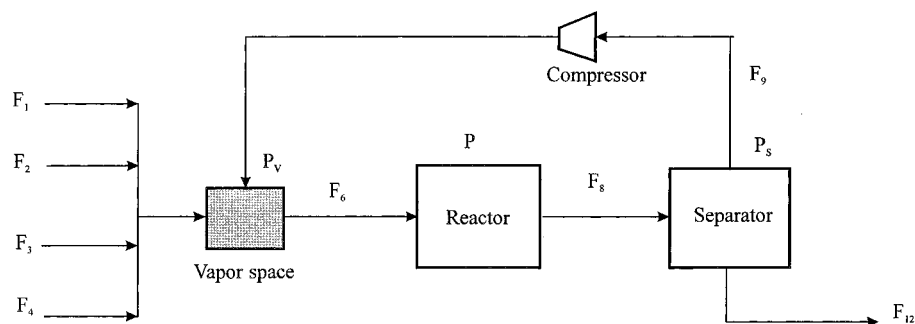
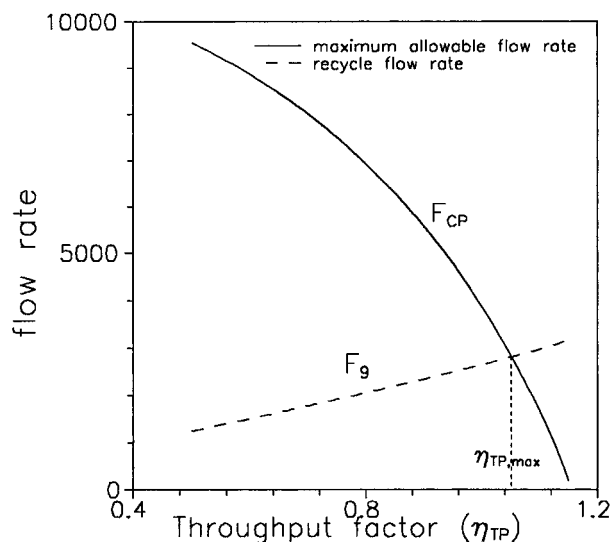**Figure 10.** Pressure for a simplified Tennessee Eastman process.

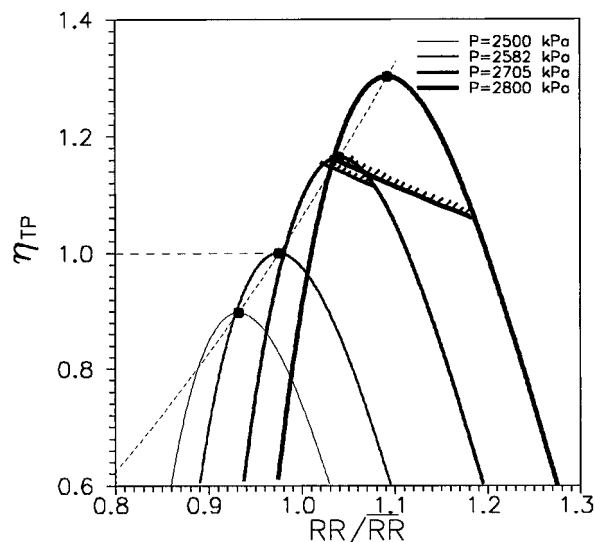**Figure 11.** Characteristic of recycle compressor corresponding recycle flow as the throughput factor was changed.
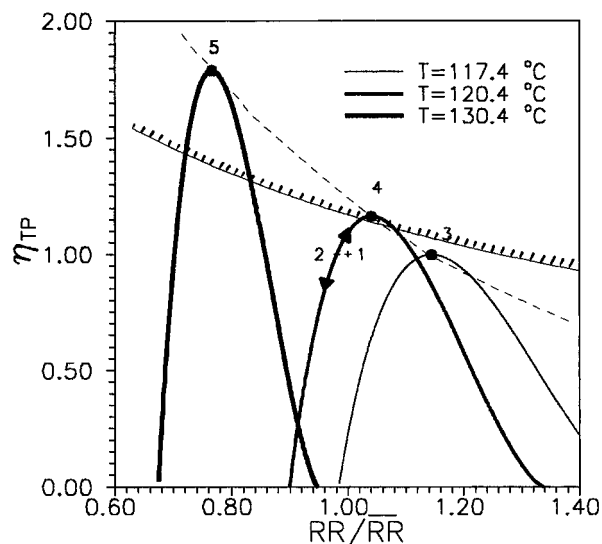

Figure 12. Optimal operating policy at different reactor temperatures and constraints imposed by the recycle compressor.

raise the production rate by increasing the reactor pressure since we may run into equipment constraint. Figure 13 shows that, as a result of limited compressor capacity, little production rate increase can be obtained as we increase the reactor pressure.

The interaction between equipment design and process operation is nicely illustrated in this case. Inadequately designed equipment (e.g., equipment constraint) leads to a completely different implication from the process point of view (e.g., process constraint).

Figure 13. Optimal operating policy at different reactor pressures.

5.3. Variable Reactor Temperature and Pressure. Here, we are interested in the maximum achievable product rate by changing both the reactor temperature and the reactor pressure. The temperature range of interest is between 100 and 150 °C, and the pressure range studied is between 2000 and 3000 kPa.

The loci of OOP's (e.g., solid circles in Figures 12 and 13) are generated as we change T and P, and the corresponding η_{TP} and $RR/\overline{RR}$ are shown in the contour plots (Figure 14a,b). Next, the equipment constraint is also shown in Figure 14. The results show that, in theory, the production rate can be raised to almost 200% of the nominal value, and this corresponds to the highest reactor temperature ($T = 150$ °C) and a moderate reactor pressure ($P = 2489$ kPa). Furthermore, as indicated earlier, an increase in the pressure in not favorable for the production rate increase at the high pressure range (Figure 14).

Notice that, in this work, the effect of the reactor level is not studied in detail, but the effect of V_G is quite obvious and can be understood from eq 25. One can increase the production rate by simply increasing the vapor volume V_G (or decreasing the reactor liquid level).

6. Summary

From the study of this TE process, a systematic procedure can be devised to analyze the operability of recycle processes. First, the rigorous process model can be simplified to a manageable level by eliminating or combining process units, and, generally, the model can be constructed from the reaction kinetics and material
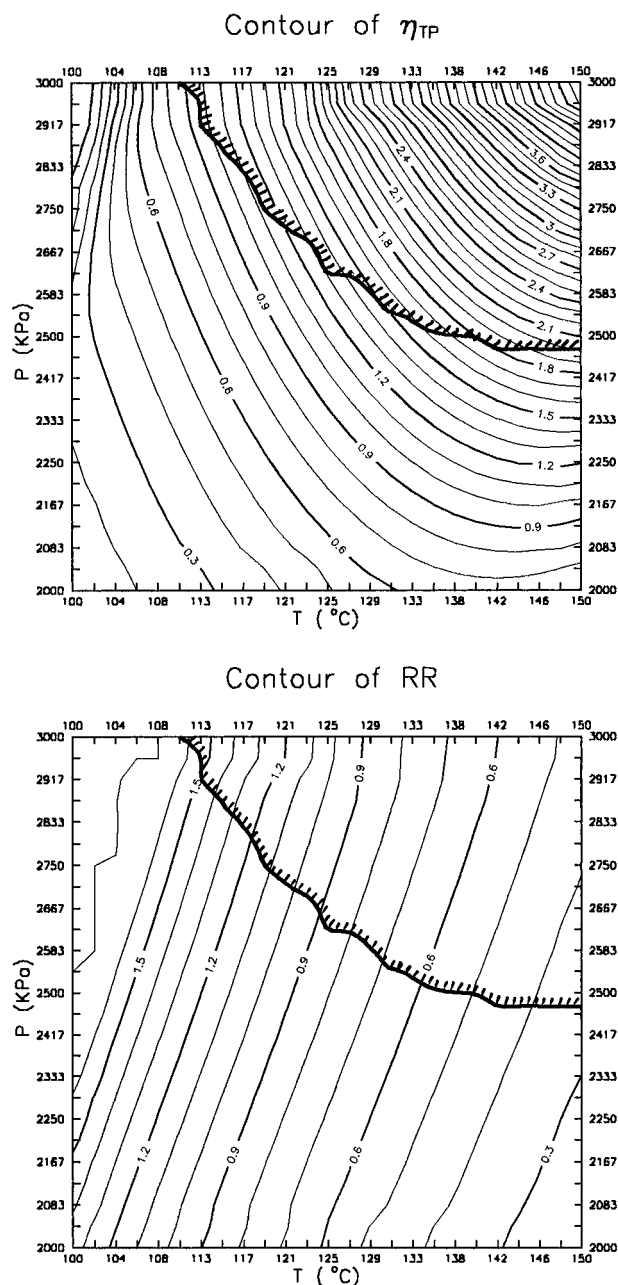
Contour of η_{TP}

Contour of RR

Figure 14. Effect of temperature and pressure on loci of the optimal operating policy.

balances. The elimination and/or combination of a process unit can be carried out according to the function of the units. For example, if possible, we can combine adjacent separators into a single separation unit. Second, the trajectory of optimal operating policy (OOP) can be established (Figures 6 and 7) by optimizing the composition distribution in the reactor as the production rate changes, and, subsequently, the inherent process constraint is explored (Figure 7). Third, possible equipment constraint is explored (Figures 10 and 11), and the constraint imposed by the equipment is plotted on the OOP (Figure 12). These three steps provide insight into the operation of recycle processes with regard to the inherent process constraint and adequacy of the designed process equipment. In order to achieve the maximum production rate, the set points of key controlled variables (e.g., $y_{A,8}$, RR in Figure 7) are adjusted (e.g., reset by throughput manipulator) as the product rate varies. However, if the constant set-point practice is preferred, the control structure has to be evaluated

such that it will not impose any further constraints on production rate changes (Figure 8).

Moreover, the effects of reactor temperature, pressure, and level on the operability can also be investigated in a similar manner, and the appropriateness of the reactor temperature and pressure can be evaluated (Figures 12 and 13) to achieve a better productivity. That is, we simply repeat the three-step procedure at different operating conditions. Therefore, the procedure can be summarized as follows:

S1. Construct a simplified process model by retaining only the essential process units (e.g., Figure 2).

S2. Construct an optimal operating policy (Figure 7), while the physical variables are held constant.

S3. Explore possible equipment constraint and show them on the OOP (Figure 12).

S4. Evaluate control structure constraint if the fixed set-point practice is preferred (Figure 8).

Note that steps S1–S4 are repeated as we explore the effects the reactor temperature and/or pressure.

7. Conclusion

In this work, process characteristics of recycle plants are studied. Using a simplified process model, the steady-state behavior of the TE process is studied. At the nominal condition, multiple steady states are observed and a unique pressure effect can also be seen for this two-phase reactor. Furthermore, the operability of the recycle process can be evaluated. The results show that there exists an inherent process constraint on the production rate. However, the design of the control system and the design of process equipment may further limit the operability of recycle processes. A four-step procedure is proposed to evaluate the appropriateness of the control structure as well as process equipments. Thus, optimal operating policy can be established to achieve a better productivity. More importantly, the complex recycle plant can be analyzed and understood via a rather simple process model which can be easily understood by most chemical engineers.

Acknowledgment

Nomenclature

F_{cp} = flow capacity of the compressor
F_i = molar flow rate of stream i
$y_{k,i}$ = vapor mole fraction of component k in the ith stream
$x_{k,i}$ = liquid mole fraction of component k in the ith stream
r_i = reaction rate for ith reaction
k_i = rate constant for ith reaction
V_G = vapor volume in the reactor
$\mathcal{R}$ = overall reaction rate
$\mathcal{R}_p$ = contribution to the overall reaction rate from physical variables
$\mathcal{R}_y$ = contribution from the reactant distribution
RR = recycle ratio (F_9/F_{12})
P_k^v = vapor pressure of component k
T = reactor temperature
P = reactor pressure
P_v = pressure in the vapor space
P_s = separator pressure
P_r = pressure ratio

Greek Symbols

η_{TP} = throughput factor
κ = product distribution factor
ξ = conversion

Superscripts

$^{-}$ = nominal value

Literature Cited

Aris, R.; Amundson, N. R. An Analysis of Chemical Reaction Stability and Control. *Chem. Eng. Sci.* **1958**, *7*, 121−131.

Banerjee, A.; Arkun, Y. Control Configuration Design Applied to the Tennessee Eastman Plant-wide Control Problem. *Comput. Chem. Eng.* **1995**, *19*, 453−480.

Douglas, J. M. *Conceptual Design of Chemical Processes*; McGraw-Hill: New York, 1988.

Downs, J. J.; Vogel, E. F. A Plant-wide Industrial Process Control Problem. *Comput. Chem. Eng.* **1993**, *17*, 245−255.

Kanadibhotla, R. S.; Riggs, J. B. Nonlinear Model Based Control of a Recycle Reactor Process. *Comput. Chem. Eng.* **1995**, *19*, 933−948.

Luyben, W. L. Design and Control Degrees of Freedom. *Ind. Eng. Chem. Res.* **1996a**, *35*, 2204−2214.

Luyben, W. L. Simple Regulatory Control of the Eastman Process. *Ind. Eng. Chem. Res.* **1996b**, *35*, 3280−3289.

Luyben, M. L.; Luyben, W. L. Design and Control of a Complex Process Involving Two Reaction Steps, Three Distillation Columns, and Two Recycle Streams. *Ind. Eng. Chem. Res.* **1995**, *34*, 3885−3898.

Luyben, M. L.; Tyreus, B. D.; Luyben, W. L. Analysis of Control Structures for Reaction/Separation/Recycle Processes with Second-Order Reaction. *Ind. Eng. Chem. Res.* **1996**, *35*, 758−771.

Lyman, P. R.; Georgakis, C. Plant-wide Control of the Tennessee Eastman Problem. *Comput. Chem. Eng.* **1995**, *19*, 321−331.

Lyman, P. R.; Luyben, W. L. Production Rate Changes in a Ternary Two-Recycle Process. *Ind. Eng. Chem. Res.* **1996**, *35*, 2198−2203.

McAvoy, T. J.; Ye, N. Base Control for The Tennessee Eastman Problem. *Comput. Chem. Eng.* **1994**, *8*, 383−413.

McAvoy, T. J.; Ye, N.; Gang, C. Nonlinear Inferential Parallel Cascade Control. *Ind. Eng. Chem. Res.* **1996**, *35*, 130−137.

Perlmutter, D. D. *Stability of Chemical Reactors*; Wiley: New York, 1965.

Price, R. M.; Lyman, P. R.; Georgakis, C. Throughput Manipulation in Plantwide Control Structure. *Ind. Eng. Chem. Res.* **1994**, *33*, 1197−1207.

Ricker, N. L. Model Predictive Control of a Continuous, Nonlinear Two-Phase Reactor. *J. Process Control* **1993**, *3*, 109−123.

Ricker, N. L. Optimal Steady-State Operation of the Tennessee Eastman Challenge Process. *Comput. Chem. Eng.* **1995**, *19*, 949−959.

Ricker, N. L. Decentralized Control of the Tennessee Eastman Challenge Process. *J. Process Control* **1996**, *6*, 205−221.

Ricker, N. L.; Lee, J. H. Nonlinear Model Predictive Control of the Tennessee Eastman Challenge Process. *Comput. Chem. Eng.* **1995a**, *19*, 961−981.

Ricker, N. L.; Lee, J. H. Nonlinear Modeling and State Estimation for the Tennessee Eastman Challenge Process. *Comput. Chem. Eng.* **1995b**, *19*, 983−1005.

Tyreus, B. D.; Luyben, W. L. Dynamics and Control of Recycle Systems. 4. Ternary Systems with One or Two Recycle Streams. *Ind. Eng. Chem. Res.* **1993**, *32*, 1154−1162.

Vinson, D. R.; Georgakis, C.; Fossy, J. Studies in Plant-wide Controllability Using the Tennessee Eastman Challenge Problem. Proceedings of the ACC, Seattle, 1995; pp 250−254.

Wu, K. L.; Yu, C. C. Reactor/Separator Processes with Recycle 1. Candidate Control Structure for Operability. *Comput. Chem. Eng.* **1996**, *20*, 1291−1316.