



# Design and control of an olefin metathesis reactive distillation column

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## Abstract

This paper considers the design and control of a reactive distillation column in which one reactant is consumed and two products are formed ( $A \rightleftharpoons B + C$ ). The volatilities are  $\alpha_B > \alpha_A > \alpha_C$ , i.e. the reactant is intermediate boiling between the two products. The metathesis of 2-pentene is considered as the demonstrative example. The column has a single feed of the intermediate boiling reactant. The distillate contains mostly light component and the bottoms mostly heavy.

Three designs are considered: the base case (low-conversion/low-pressure), a low-conversion/high-pressure case and a high-conversion/high-pressure case. The base design is obtained from the literature, and the other two steady-state designs are optimized with respect to the total annual cost. All the designs are found to be openloop stable. Five control structures are studied for the base design. Then the best two structures are applied to the remaining two designs. This category of reactive distillation exhibits less challenging problems than other categories since it uses a single feed, which eliminates the need for the control structure to perfectly balance two fresh feeds.

Simulation results demonstrate that effective dynamic control is provided by a control structure that uses two temperatures to maintain the purities of both product streams. No internal composition measurement is required. This structure is found to be robust and stable and rejects loads and tracks setpoints very well. © 2002 Elsevier Science Ltd. All rights reserved.

*Keywords:* Reactive distillation; Process design and control; Olefin metathesis

## 1. Introduction

Reactive distillation is sometimes an excellent alternative to conventional flowsheets with separate reaction and separation sections. It is particularly effective for reversible reactions with low equilibrium constants. Of course the reactant and product volatilities must be such that products can be removed and reactants retained inside the column. The temperature levels for both reaction and vapor–liquid equilibrium must overlap. Significant reductions in both capital and energy costs are possible in some systems.

The literature in reactive distillation has grown rapidly in recent years and is summarized in the new book by Doherty and Malone (2001). Steady-state design is considered in many papers. Openloop dynamics and multiple steady states are discussed in several others. However, there have

been relatively few papers that discuss closedloop control of reactive distillation columns. These are reviewed in a recent publication (Al-Arfaj & Luyben, 2000).

We have studied control structures for two generic types of reactive distillation columns in previous papers (Al-Arfaj & Luyben, 2000; Luyben, 2000; Al-Arfaj & Luyben in press, submitted for publication).

1. A hypothetical ideal system and the methyl acetate system with two reactants and two products:  $A + B \rightleftharpoons C + D$ .
2. The ETBE system with two reactants and a single product:  $A + B \rightleftharpoons C$ .

In this paper, we extend these studies to consider another generic class of reactive distillation in which there is a single reactant and two products:  $A \rightleftharpoons B + C$ . The specific chemical system considered is olefin metathesis, whose steady-state design has been studied by Okasinski and Doherty (1998). The reactant is intermediate boiling between the light and heavy products. As our study illustrates, this reactive distillation system is much more easily controlled than any of the systems previously studied. The control structure is relieved

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of the need to provide a way to perfectly balance the two reactants fed to the column so that reaction stoichiometry is satisfied.

## 2. Process description

Olefin metathesis is used in the petrochemical industry. In this paper, we consider the metathesis of 2-pentene to form the two products 2-butene and 3-hexene.



This reaction is a good candidate for reactive distillation for several reasons:

1. The reaction takes place in the liquid phase at temperatures that are suitable for vapor–liquid equilibrium requirements of distillation.
2. All the components are hydrocarbons and similar in chemical structure. Deviations from ideality are negligible, and no azeotropes are formed.
3. The normal boiling points of the components allow an easy separation between the reactant 2-pentene (310 K) and the two products (light distillate product 2-butene at 277 K and heavy bottoms product 3-hexene at 340 K).

The reaction is kinetically controlled. Detailed physical properties and kinetic data are provided by Okasinski and Doherty (1998) and Chen, Huss, Malone, and Doherty (2000). We use this information as the basis of our steady-state and dynamic models. The model assumptions are:

1. Ideal gas and Raoult's law are valid.
2. Power-law kinetics.
3. No heat of reaction.
4. Constant molar overflow.
5. Constant molar holdup distribution.
6. Partial reboiler and a total condenser (no reaction takes place in either of them).

## 3. Steady-state designs

Three different design cases are considered in this paper. The first is that studied by Okasinski and Doherty (1998), which has a low conversion (93%) and operates at atmospheric pressure. The second has the same conversion but the pressure is increased so that cooling water can be used in the condenser. The third case is a high conversion design. Similar control structures are studied for all the three cases.

### 3.1. Base case (low-conversion/low-pressure)

Okasinski and Doherty (1998) proposed a design method for kinetically controlled reactive distillation. They applied their method to the metathesis of 2-pentene. They provide a review of this process as well as all the chemical and

physical data. Chen et al. (2000) use this case as an example in their work in which they show the steady-state effect of tray holdup and reflux ratio on conversion.

The base design, which is taken from Okasinski and Doherty (1998), is summarized in Table 1. The temperature and composition profiles are shown in Fig. 1. In this design, the product purities are 93.4 mol%. The column diameter is around 2 m, and the stage holdup is 6.1 kmol, which is equivalent to an acceptable tray height of 20 cm. Pressure is 1 atm. The column has 12 reactive trays and is fed in the middle.

### 3.2. Steady-state design 2 (low-conversion/high-pressure)

The base-case design at 1 atm pressure gives a reflux-drum temperature of 280 K. This is too low to permit the use of cooling water in the condenser, so refrigeration would have to be used. This means very expensive heat removal. If there is no high-temperature limitation, the operating pressure could be increased so that cooling water can be used. The exothermic reaction has a low heat of reaction (about  $-25$  kJ/kmol), so the equilibrium constant does not decrease significantly as temperature is increased. In this second design, the operating pressure is set at 5 atm, giving a reflux-drum temperature of 329 K (the bubblepoint temperature of the  $\text{C}_4^-$  distillate).

#### 3.2.1. Effect of design parameters

To obtain a better understanding of the process, the effects of various design parameters on the steady-state design are explored. The number of trays, the operating pressure, the holdup per tray and the reflux ratio are studied. In all cases the fresh feed is located in the middle of the column. This is justified by the fact that the average relative volatility between the reactant and each of the two products is about the same ( $\alpha = 2.5$  for  $\text{C}_4/\text{C}_5$  and 2.3 for  $\text{C}_5/\text{C}_6$ ). If the degree of separation difficulty were greater for one of the separations, the number of trays in the corresponding section would be relatively larger in that section than in the other.

Fig. 2A shows the effect of pressure on conversion for different numbers of trays. Reflux ratio is fixed at 4, and tray holdup is constant at 6.1 kmol. The conversion drops after reaching an optimum pressure when only a small number of trays are used. This is due to the adverse impact of pressure on the relative volatilities in this hydrocarbon system. However, this does not happen when there are a large number of trays because separation capacity is not limiting performance. These results indicate that high conversions cannot be achieved in a 12-tray column, even if it is operated at the optimum pressure of about 2.5 atm. Additional trays are required.

Fig. 2B shows that higher conversion can alternatively be obtained when larger tray liquid holdups are used. However, there are hydraulic constraints on how much liquid can be used on each tray because excessive liquid heights

Table 1  
Details design variables\*

		Low conversion/ low pressure (base)	Low conversion/ high pressure	High conversion/ high pressure
Pressure (atm)		1	5	5
Conversion (%)		93.4	93.4	99.0
Total trays		12	28	38
Feed tray		7	15	20
Reflux ratio		4	1.7	1.84
Vapor boilup (kmol/h)		250	135	142
Diameter (m)		2	1	1
Holdup (kmol/tray)		6.1	1.5	1.5
Distillate comp. (mole fraction)	$C_4^=$	0.9342	0.9335	0.9901
	$C_5^=$	0.0655	0.0657	0.0099
	$C_6^=$	0.0003	0.0008	0.0000
Bottoms comp. (mole fraction)	$C_4^=$	0.0002	0.0006	0.0000
	$C_5^=$	0.0657	0.0662	0.0099
	$C_6^=$	0.9341	0.9332	0.9901
Temperature (K)	$T_1$	333.5	395.7	400.8
	$T_{N_F}$	308.2	361.1	361.3
	$T_{N_T}$	281.1	332.5	328.3
TAC (MM \$/year)		1.4314	0.3074	0.3387

\*All cases are fed the same feed (100 kmol/h  $C_5^-$ ) and have the same product flowrates (distillate = 50 kmol/h and bottoms = 50 kmol/h). Tray numbering is from the bottom up.

produce large pressure drops. Conversion decreases with increasing pressure due to the limited separation capacity. Fig. 2C shows that as the reflux ratio increases the conversion increases and the separation becomes easier. This suggests that higher reflux ratios could be used to offset the small number of trays, but the energy cost would probably make this alternative uneconomical.

### 3.2.2. Optimum economic steady-state design

In order to come up with a more realistic design, an optimization with respect to the number of trays and reflux ratio is performed. The objective function is minimum total annual cost (TAC) of both energy and capital. The optimum column has more trays than the base case and requires less heat input. Running at a pressure of 5 atm means that vapor density is higher, so vapor velocities are lower. Both these result in a smaller diameter column of about 1 m at the optimum. Since the diameter is decreased by a factor of two from the base-case design, the holdup is reduced by a factor of four for the same tray liquid height. A reasonable liquid height of 20 cm is used in order to maintain a reasonable pressure drop through the tray. Therefore, the holdup is 1.5 kmol/tray. The feed tray is always in the middle of the column.

For a given total number of trays, the reflux ratio is varied to achieve the same conversion as the base case (93.4%). Fig. 3A shows that the optimum number of trays is about 28. The reflux ratio for this column is 1.7, and the operating pressure is 5 atm. Fig. 3B shows that the optimum number

of trays for a high-conversion case (99%) considered in the next section has increased to about 38 with, of course, a higher total annual cost. Figs. 3A and B show that the TAC is not very sensitive to the total number of trays. For example, changing the total number of trays in the optimum high-conversion design by  $\pm 2$  ( $\pm 5\%$ ) results in an increase in the TAC by only 0.13%.

More details of the low-conversion/high-pressure design are given in Table 1. Fig. 4 gives temperature and composition profiles throughout the column. Note the significant temperature changes in both the stripping and rectifying sections as the product purities change. This indicates that it may be possible to use temperatures to maintain product purities at both ends of the column. The control studies discussed later confirm this expectation.

### 3.3. Steady-state design 3 (high-conversion/high-pressure)

To explore the effect of conversion on both the steady-state design and the dynamic control, a 99% conversion case is studied. The optimization yields the design given in Table 1 (last column). The optimum column has 38 reactive trays, reflux ratio of 1.84, holdup of 1.5 kmol/tray and operating pressure of 5 atm. Temperature and composition profiles are shown in Fig. 5.

Table 1 shows that the higher conversion case has more trays (38 versus 28) and a slightly higher reflux ratio (1.84

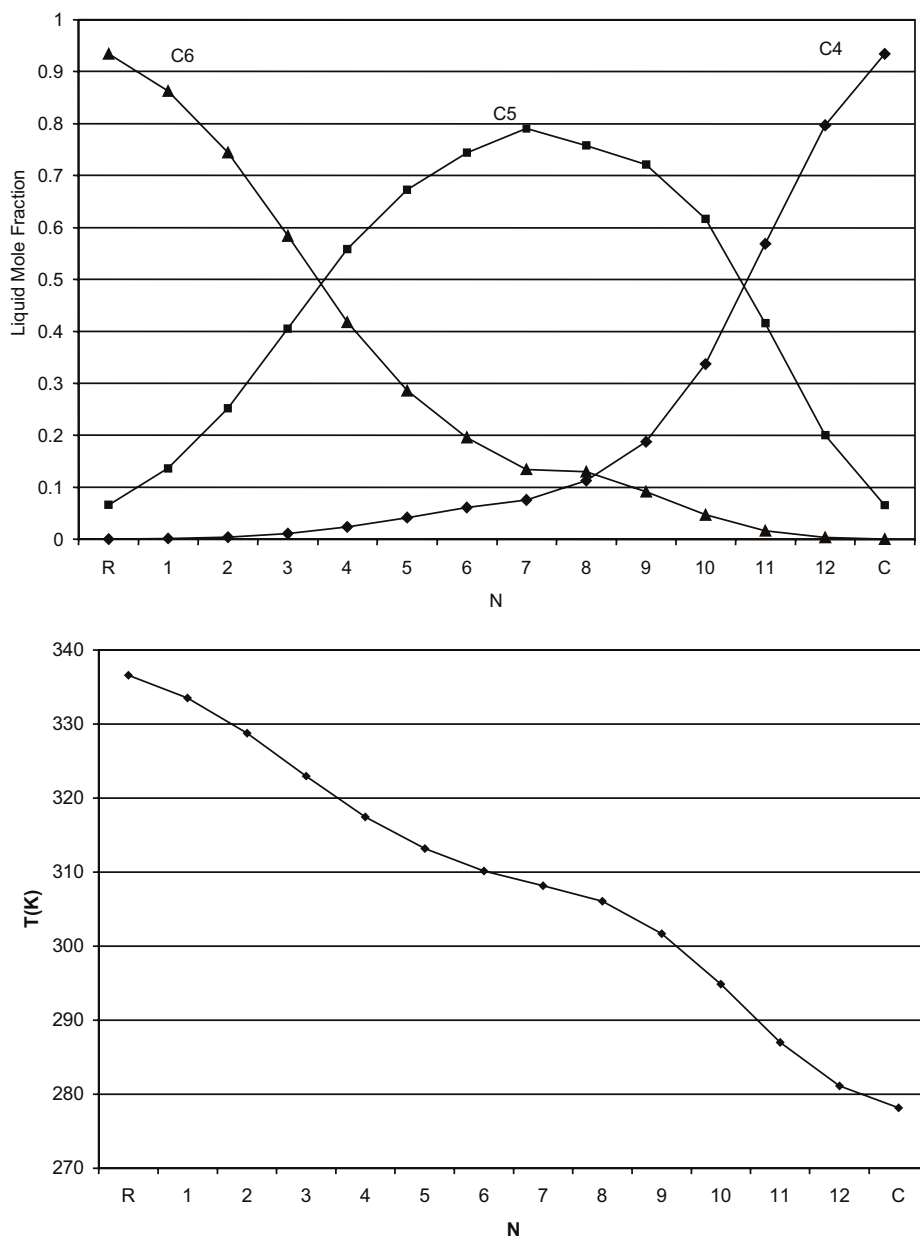


Fig. 1. Composition & temperature profile (design 1).

versus 1.7). It is interesting to compare the cost of these designs. The base design is the most expensive design since it uses high reflux (high energy) and refrigeration in the condenser, which is very expensive (\$20/GJ for 5°C). The other two designs are reasonably close to each other. Another comparison is to look at the cost per unit of product. Since all the designs use the same amount of fresh feed, the raw material cost is fixed. Therefore, to calculate the cost per unit of product we divide the TAC by the annual production rate. For example, for the base design, it is  $1,431,430 \text{ [$/y]} / (0.934 * 50 \text{ [kmol/h]}) / 8160 \text{ [h/y]} = 3.76 \text{ [$/kmol]}$ . The cost per unit of product for the low-conversion/high-pressure design is 0.81 [\$/kmol] and for the high-conversion/high-pressure de-

sign is 0.84 [\$/kmol]. From this, one can suggest that the low-conversion/high-pressure design is more economical; however, since the product purity is not high, a recovery system may be needed that would increase the overall cost. The high-conversion/high-pressure design should not require any further processing.

#### 4. Control studies

Four control structures are studied for the base-case design, as shown in Figs. 6 and 7. All the structures are SISO structures with PI controllers (P only on levels).

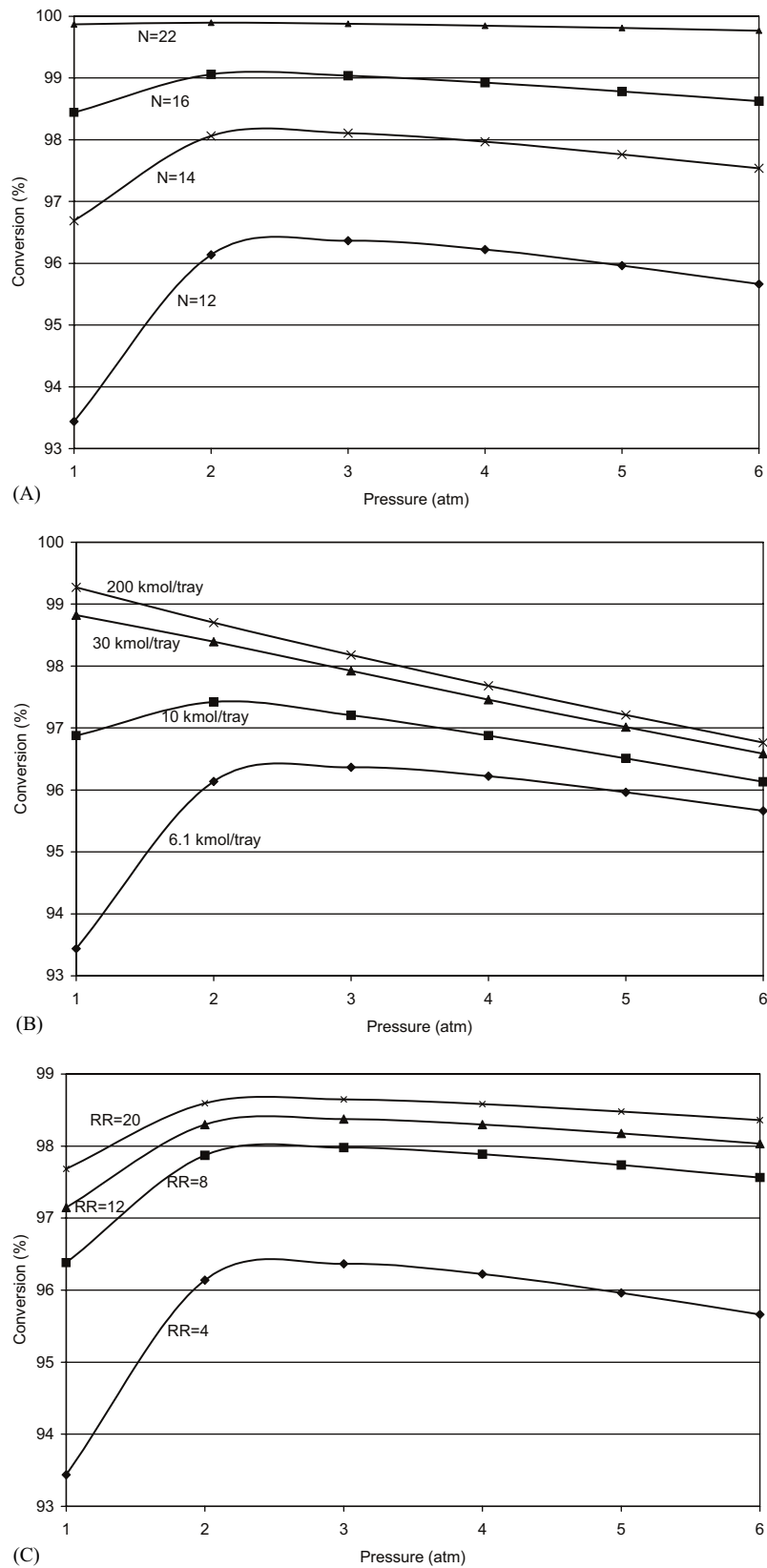


Fig. 2. (A) Effect of changing  $N_T$ , RR = 4, holdup = 6.1 kmol/tray,  $D = 50$  kmol/h; (B) effect of changing tray holdup, RR = 4,  $N_T = 12$  kmol/tray,  $D = 50$  kmol/h and (C) effect of changing reflux ratio,  $N_T = 12$ , holdup = 6.1 kmol/tray,  $D = 50$  kmol/h.

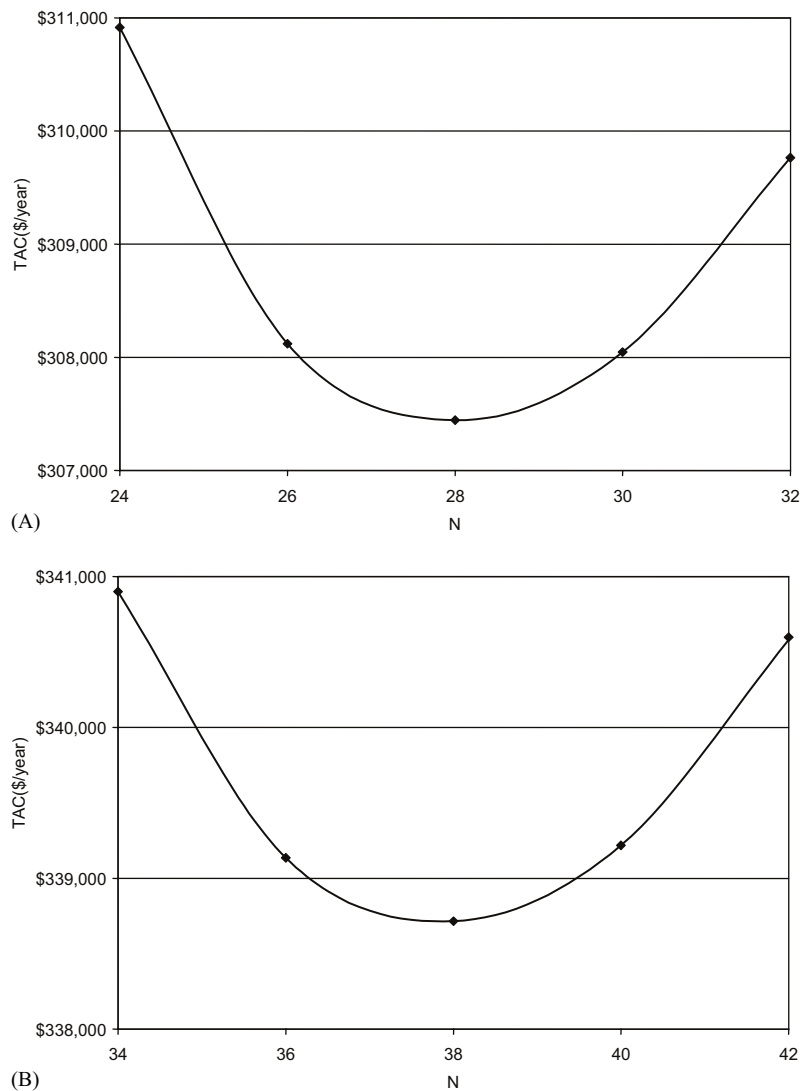


Fig. 3. (A) Optimum TAC (Design 2—93.4% conversion) and (B) optimum TAC (Design 3—99.0% conversion).

The controllers are tuned using the Tyreus–Luyben tuning method (Tyreus & Luyben, 1992). The relay-feedback method (Yu, 1999) is used to obtain the ultimate gain and ultimate period. Valves are designed to be half open at steady state. Two measurement lags of 30 s each are used in all composition or temperature loops. Similar labeling of control structures is used in this paper so that comparison with previous work can be easily made. Only the two most promising control structures are tested for the other two designs.

This system uses ideal chemical and physical models. It is expected that this process will be openloop stable and exhibits less nonlinearity compared to other systems that feature complex relationships (e.g. methyl acetate reactive distillation). To examine the dynamic stability of the openloop system, a large 100% pulse in feed rate is made. This huge pulse signal is used in order

to excite any nonlinearity that might be present in the system. The reflux flow and the vapor boilup are held constant. The reflux drum level is controlled by the distillate flowrate, and the base level is controlled by the bottoms flowrate. Simulation results for all three design cases show that the system returns to its nominal conditions. These results indicate that the system is openloop stable.

The control structures studied use either dual composition control, single-end composition control, dual temperature control or single-end temperature control.

#### 4.1. CSI (dual composition)

This is the basic structure where the purities of the two products are both controlled by direct composition con-

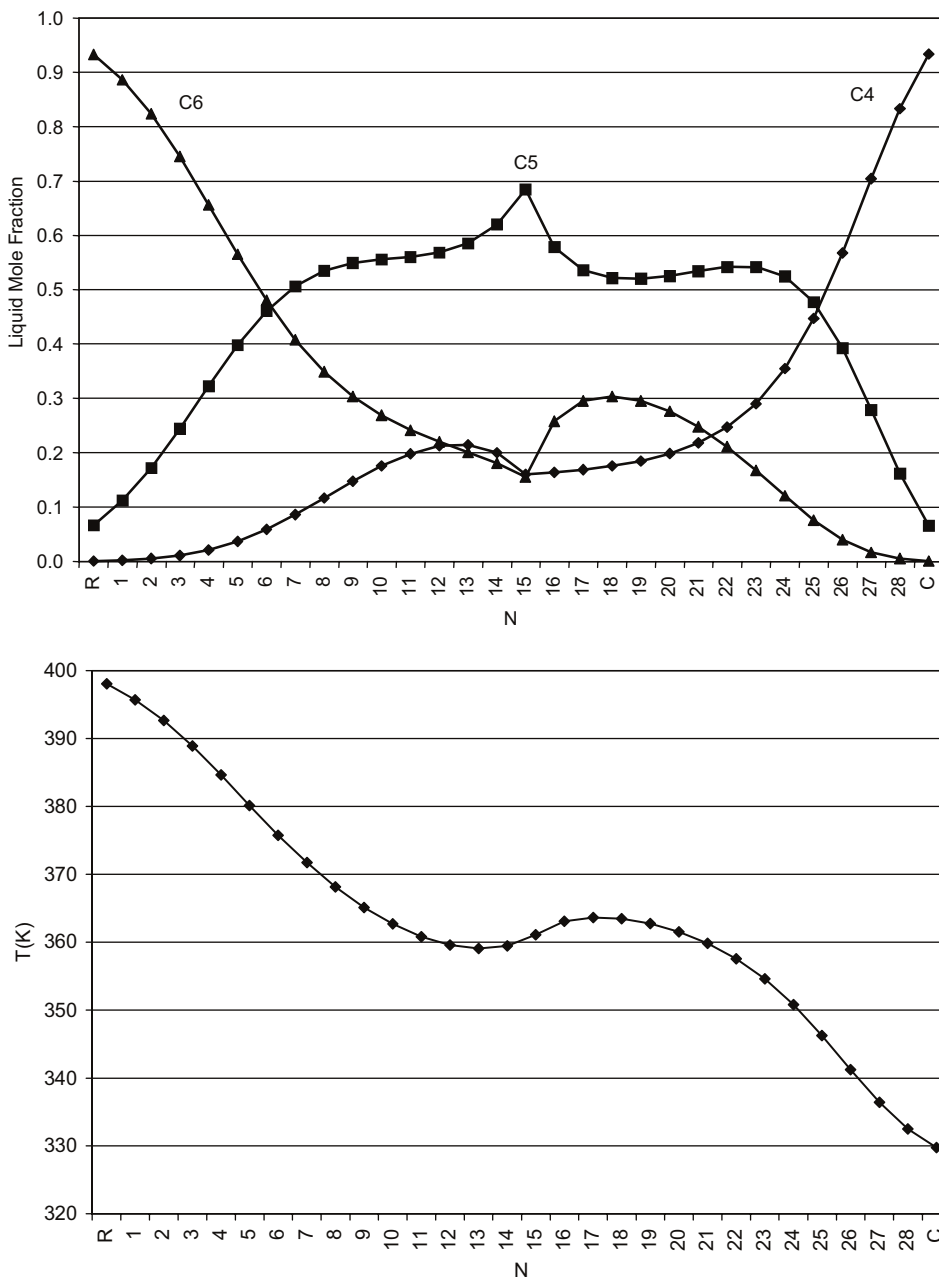


Fig. 4. Composition and temperature profile (Design 2).

trollers. The reflux flowrate is manipulated to control the butene purity in the distillate. The hexene purity in the bottoms is controlled by manipulating the vapor boilup (Fig. 6A). A slight variation of this structure was also studied in which the C5 impurities in the products were controlled instead of the product purities. There was little difference in performance.

4.2. CS1-RR (single-end composition)

The purpose of this structure is to study the effect of eliminating one of the composition control loops. The reflux

ratio is fixed. The hexene purity of the bottoms is controlled by manipulating the vapor boilup (Fig. 6B).

4.3. CS5-RR (single-end temperature)

In this structure, there are no composition analyzers. A temperature in the stripping (or rectifying) section is controlled by the reboiler heat input. The reflux ratio is kept constant (Fig. 7A). The location of the temperature control tray is selected using the singular value decomposition method (Moore, 1992, Chapter 8).

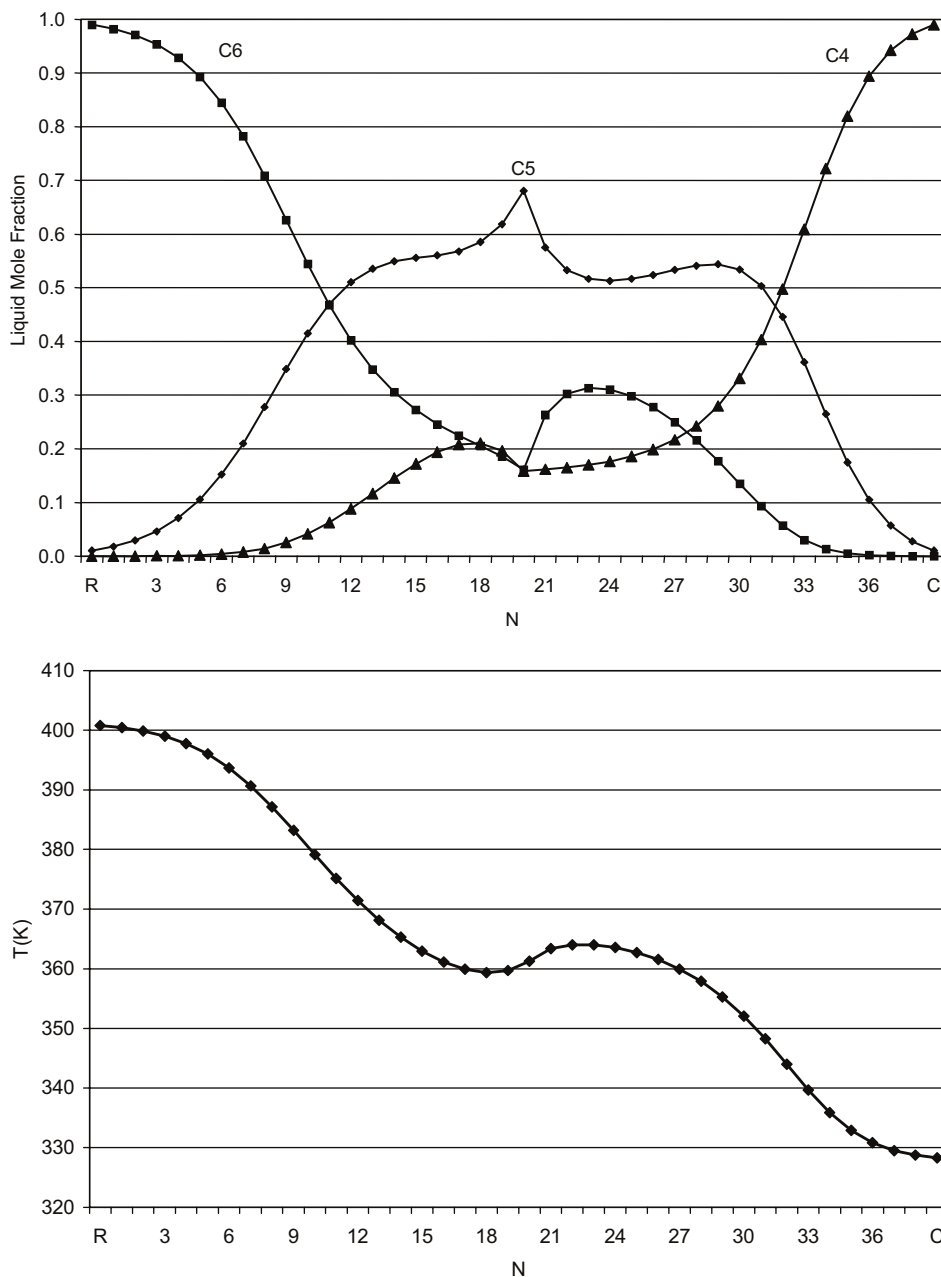


Fig. 5. Composition and temperature profile (Design 3).

#### 4.4. CS7 (dual temperature)

The CS7 structure was shown to work well in the methyl acetate system (Al-Arfaj & Luyben submitted for publication). In that system, two temperatures are used to manipulate the two fresh feed streams. There are two products produced in the chemical reaction of the methyl acetate process. The CS7 structure was shown not to work in the ETBE system (Al-Arfaj & Luyben in press) because only one product is produced.

The olefin metathesis process produces two products, so dual temperature control should work. A temperature in the rectifying section is controlled by manipulating reflux flowrate. A temperature in the stripping section is controlled by manipulating vapor boilup. See Fig. 7B.

Singular value decomposition (SVD) (Moore, 1992, Chapter 8) is used to select the most sensitive trays to be controlled. The SVD method suggests that either choice of pairing could be used ( $R-x_D$  and  $V-x_B$  or the reverse). However, SVD uses only steady-state information.



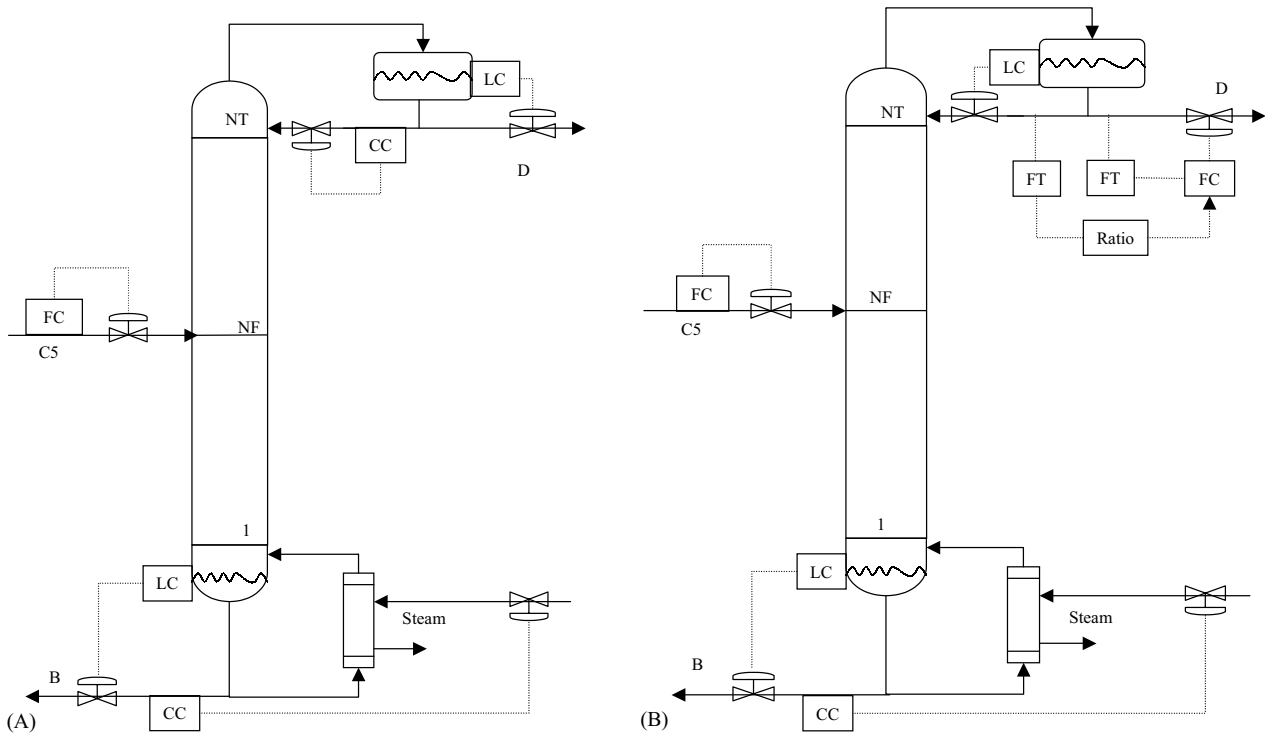


Fig. 6. (A) Control structure 1 and (B) control structure 1-RR.

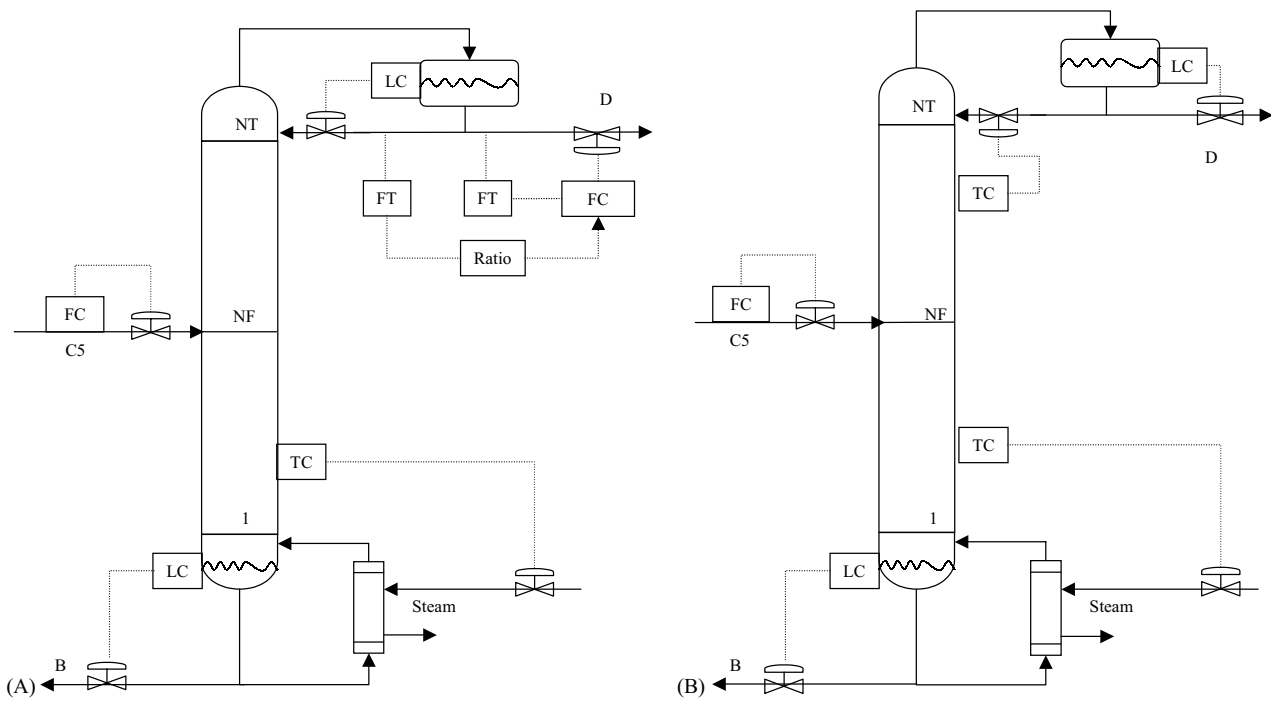


Fig. 7. (A) Control structure 5-RR and (B) control structure 7.

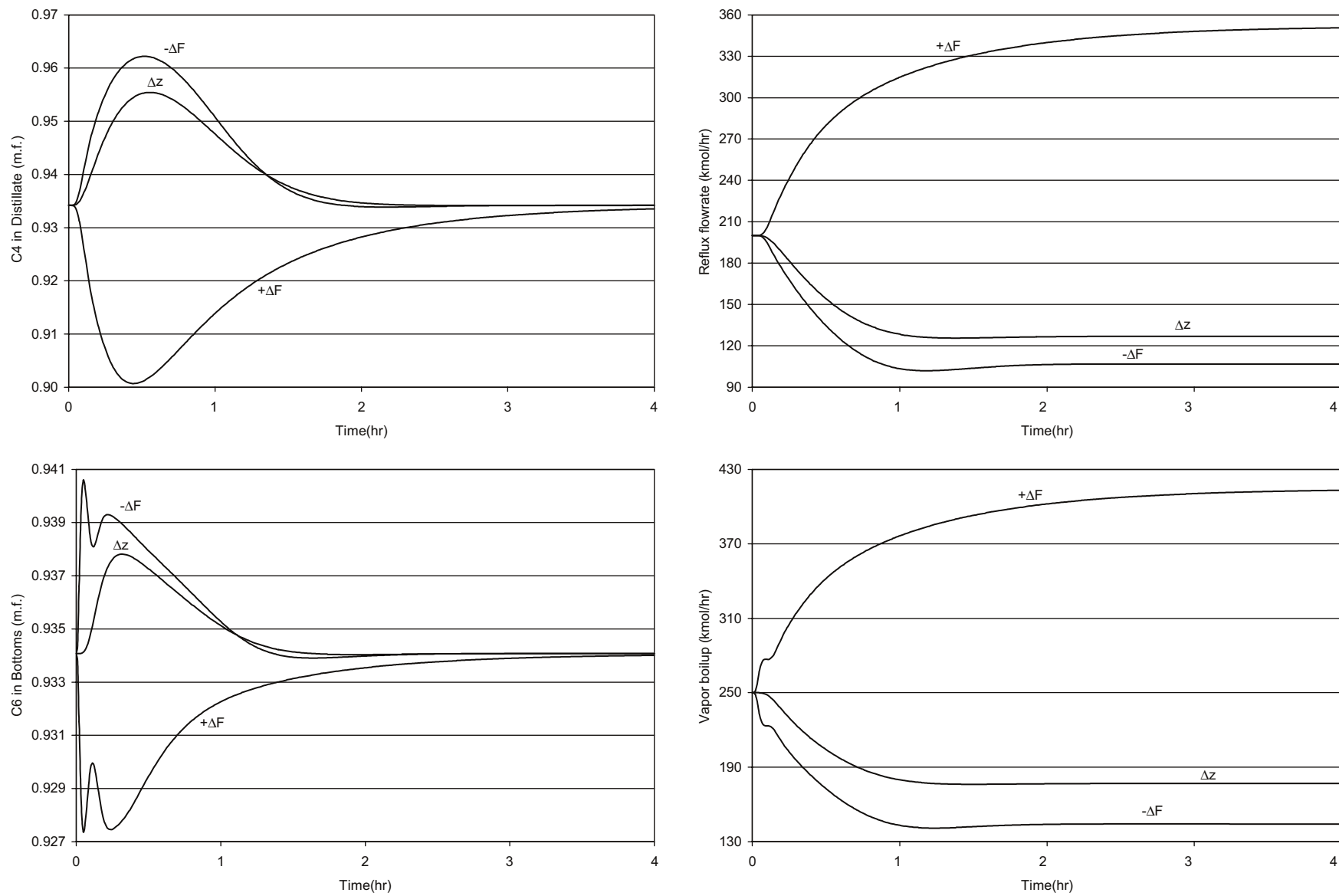


Fig. 8. Response of CS1 (Design 1).

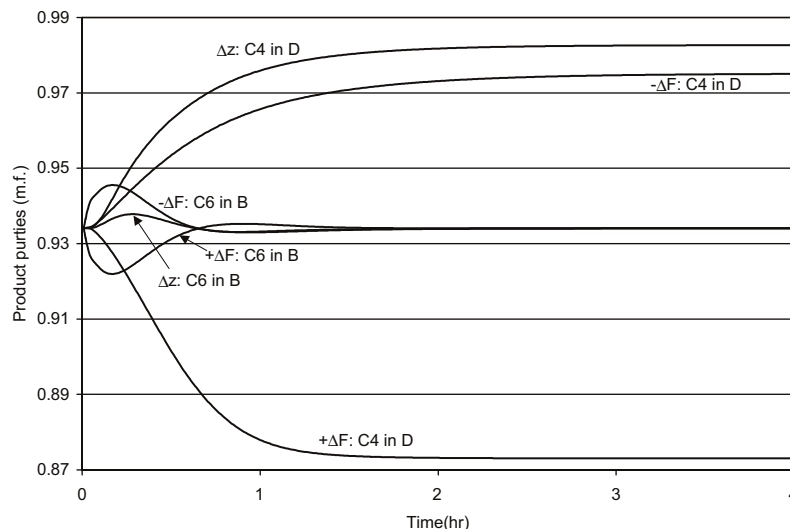


Fig. 9. Response of CS1-RR (Design 1).

As expected, dynamic simulations showed that the first pairing shown in Fig. 12 works well. The alternative is unstable.

#### 4.5. Disturbances

A number of disturbances were studied, but results from only three are reported. These disturbances are probably larger than typical in a plant environment, but they provide a good indication of the robustness of each structure and show how well load disturbances are rejected. The setpoint tracking changes are not reported here due to the limited space, but results are similar.

1. A step change of positive 25% in the fresh feed flowrate ( $+\Delta F$ ).
2. A step change of negative 25% in the fresh feed flowrate ( $-\Delta F$ ).
3. Change in the feed composition ( $\Delta z$ ) from  $C_5^- = 1.0$  to  $C_4^- = 0.15$ ,  $C_5^- = 0.70$ , and  $C_6^- = 0.15$ .

#### 4.6. Simulation results for design 1

##### 4.6.1. CS1 (dual composition)

The control of the two end purities is very effective. Fig. 8 shows that this structure is able to reject the feed rate disturbances and the feed composition disturbance.

##### 4.6.2. CS1-RR (single-end composition)

Fig. 9 shows that bottoms purity is well controlled. However, distillate purity moves significantly away from the steady-state level.

It is interesting to compare the effectiveness of single-end control in reactive distillation with that seen in conventional distillation. For a feed flowrate change in a con-

ventional column, holding the purity in one end and keeping reflux ratio constant results in the purity of the other end staying very close to the desired level. This occurs because all the flowrates vary directly with the feed flowrate.

However, in reactive distillation this situation no longer applies. As shown in Fig. 9, the distillate purity is not maintained when a feed rate is made even if the bottoms composition is controlled and the reflux ratio is held constant. Changing the feed rate in a reactive column changes the load on the reactive zone. This results in a change in the fractionation required, which means the reflux ratio has to change to maintain the same conversion and product purities. Thus, simply holding a constant reflux ratio is ineffective in reactive distillation with single-end control.

##### 4.6.3. CS5-RR (single-end temperature)

The steady-state gains between tray temperatures and vapor boilup, with reflux ratio fixed, show that there are two equally sensitive trays: Trays 3 and 11. When Tray 3 temperature is controlled by the vapor boilup, distillate purity moves significantly away from the desired level (see Fig. 10A). When Tray 11 temperature is controlled, bottoms purity is not maintained as shown in Fig. 10B.

##### 4.6.4. CS7 (dual temperature)

Singular value decomposition analysis for the system with two manipulated variables (vapor boilup and reflux) suggests that the temperature on Tray 10 should be controlled by the reflux flowrate and the temperature on Tray 5 by reboiler heat input. Fig. 11 shows the responses of the system to the three disturbances. The system is very robust, and the purities of both products are maintained

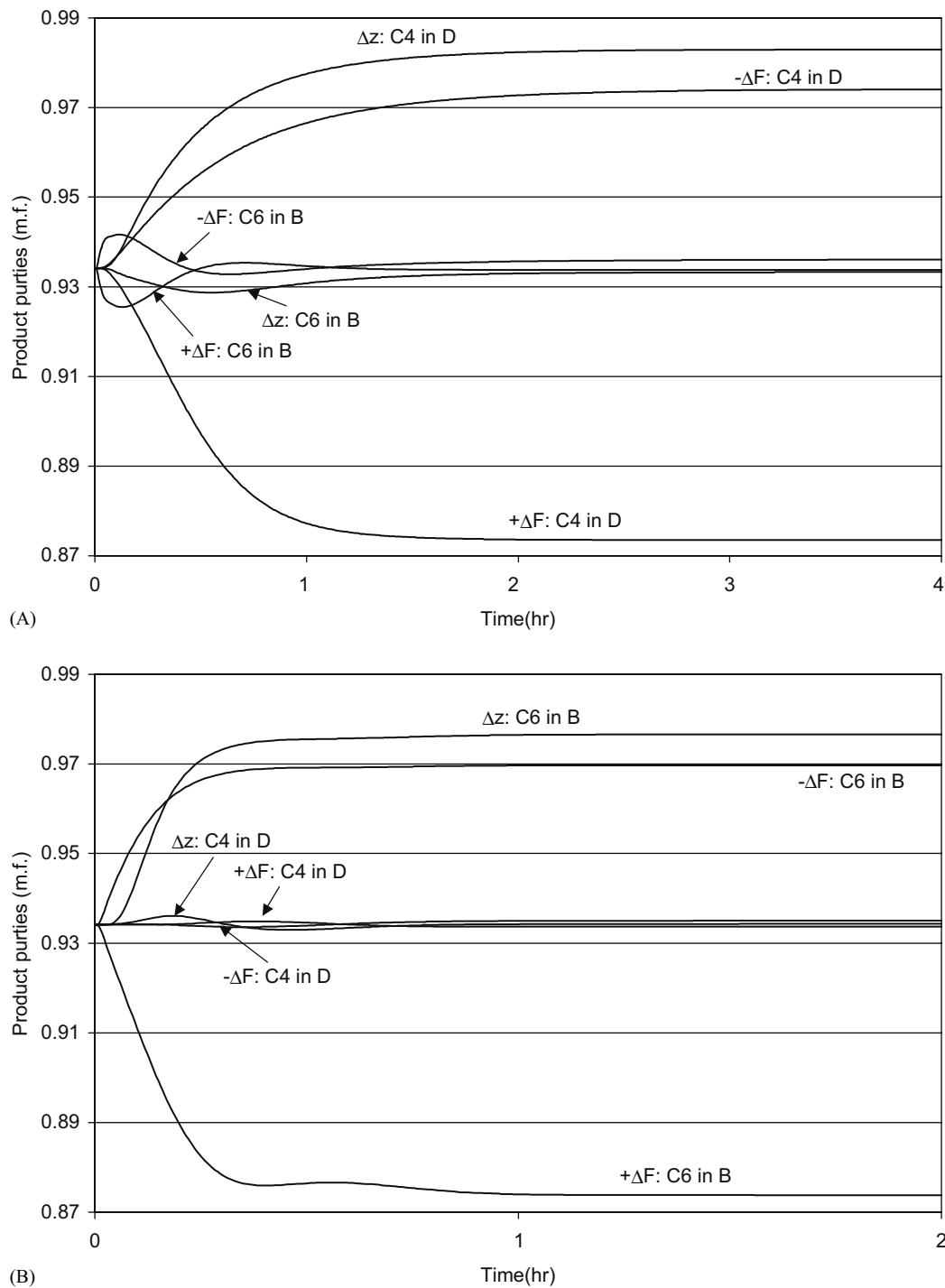


Fig. 10. (A) Response of CS5-RR (T3 by  $V_s$ )-Design 1 and (B) response of CS5-RR (T11 by  $V_s$ )-Design 1.

reasonably close to their setpoints. The feed composition disturbance produces the greatest change in product purities.

Changing the pairing to control Tray 10 with reboiler heat input and Tray 5 with reflux flowrate does not work (the column shuts itself down). As expected, the poor dynamics of this pairing produce an unstable system. This is confirmed

by calculating the Niederlinski Index for this pairing, which is negative.

#### 4.6.5. Comparison of dual composition versus dual temperature control

Fig. 12 gives a direct comparison between the use of composition and temperature control. The performance of

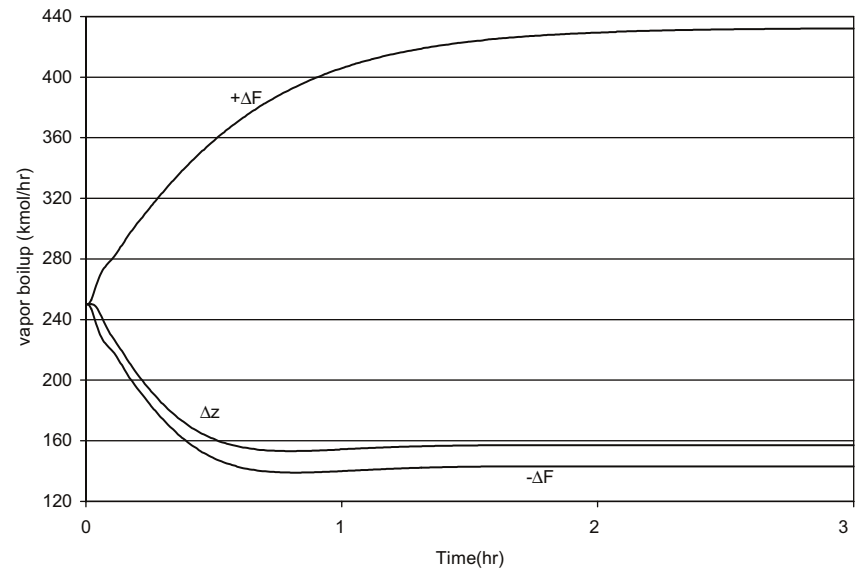
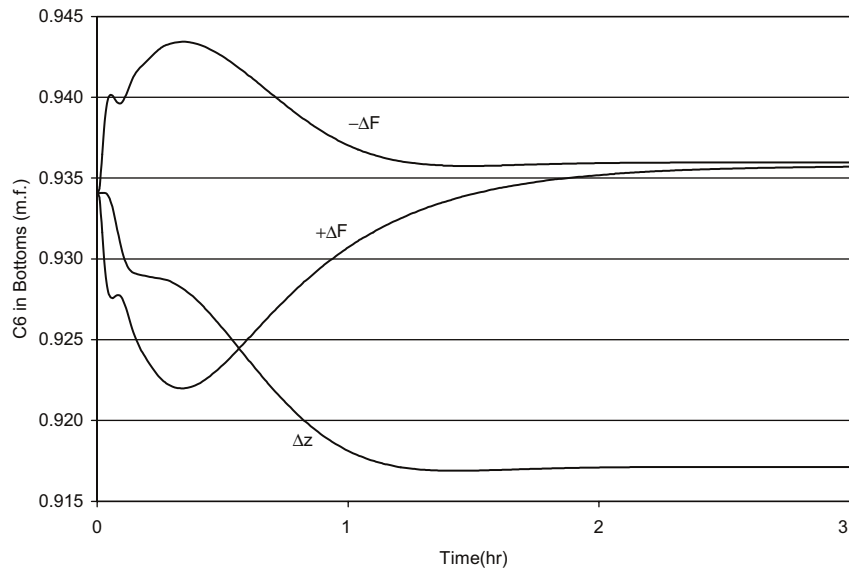
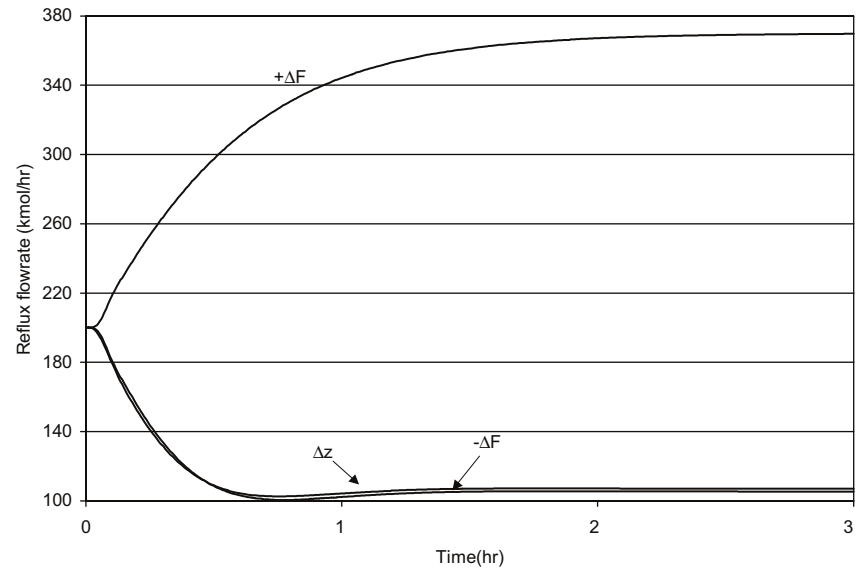
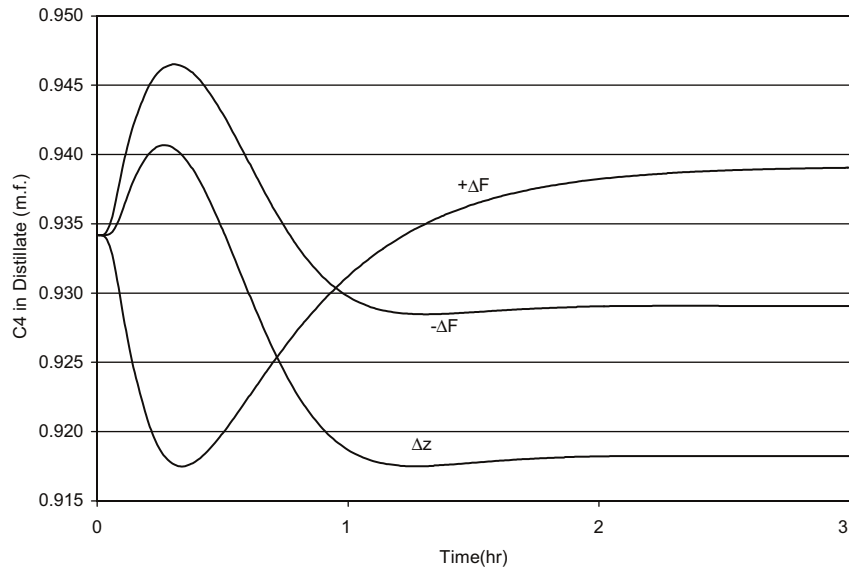


Fig. 11. Response of CS7-direct (Design 1).

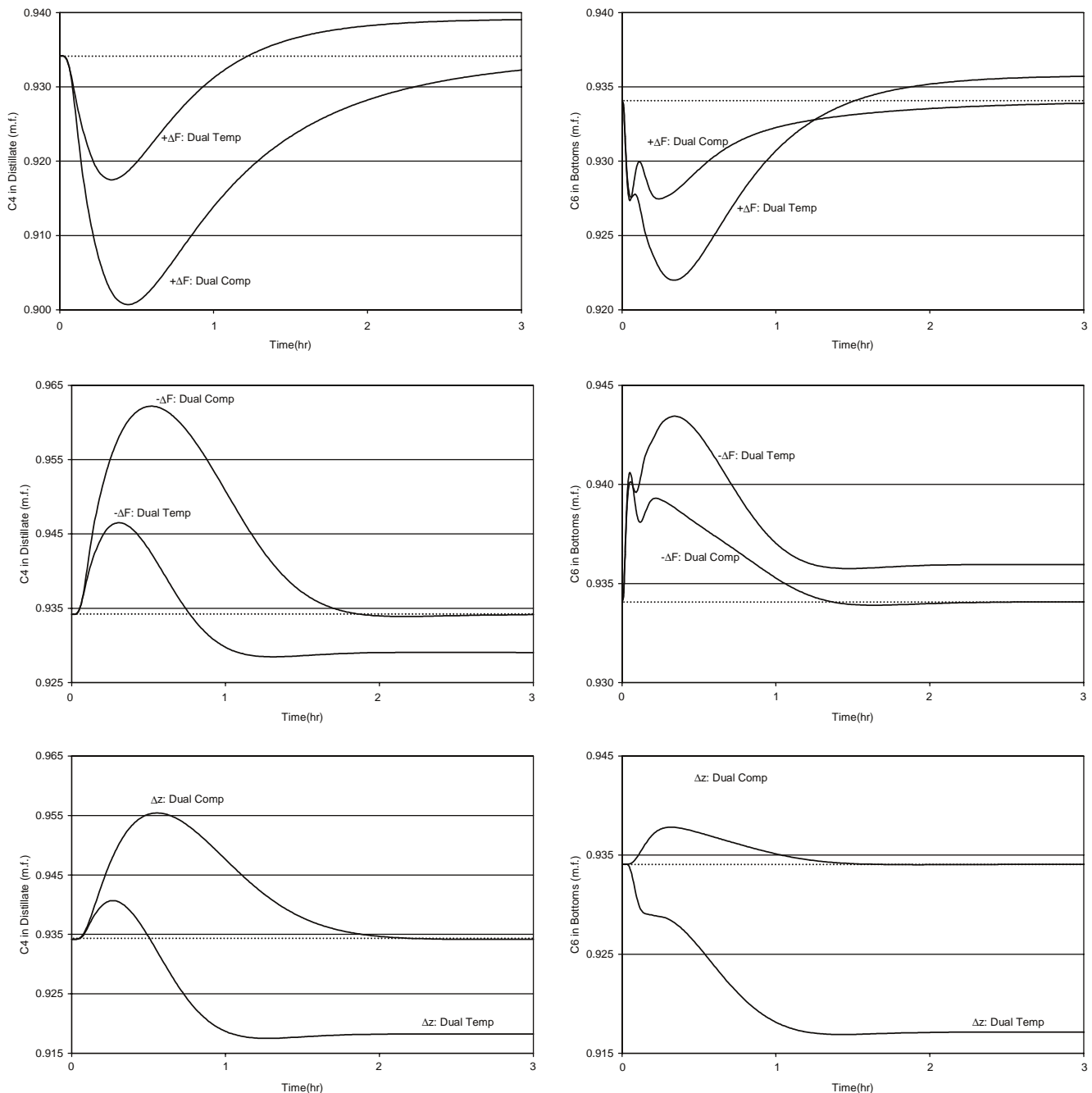


Fig. 12. Dual composition control vs. dual temperature control.

the dual temperature control (CS7) is dynamically better than that of the dual composition control for the feed flow disturbances. This is because the temperature sensors on the intermediate trays see the disturbances more quickly than the composition analyzers at the very ends of the column. However, the steady-state performance of the dual temperature structure is not as good as the dual composition. The product purities are not driven back to their desired levels. This is particularly true for the feed composition disturbance, which produces a drop in purity of about 2%.

These results suggest that a composition/temperature cascade control structure would provide the most effective control, both dynamically and at steady state.

#### 4.7. Simulation results for designs 2 and 3

Simulation results using the dual composition control (CS1) and the dual temperature control (CS7) are presented. The same disturbances are used.

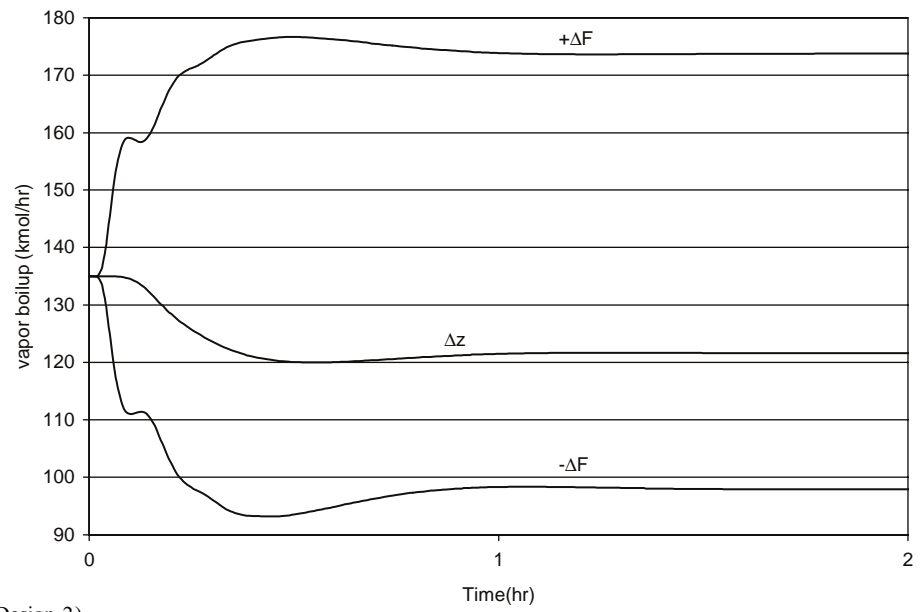
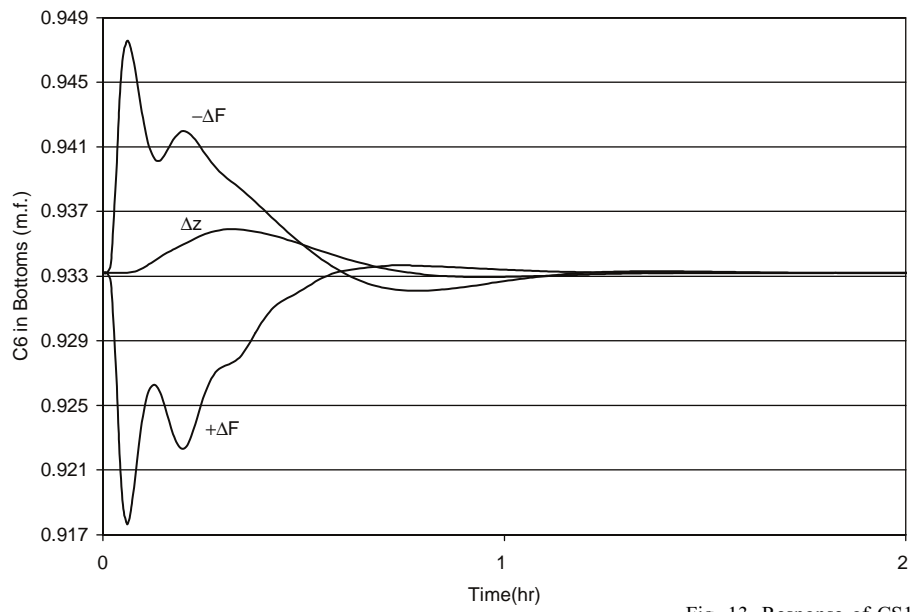
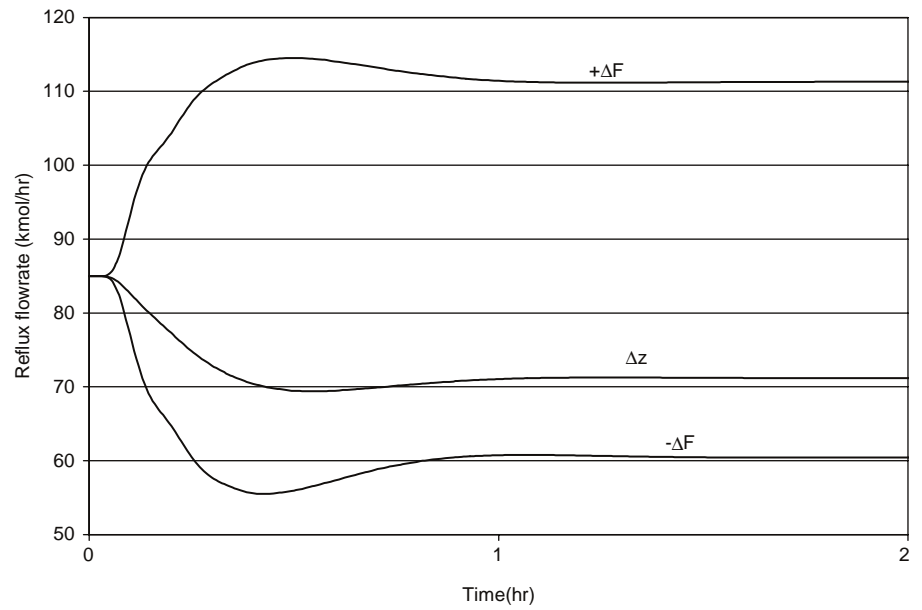
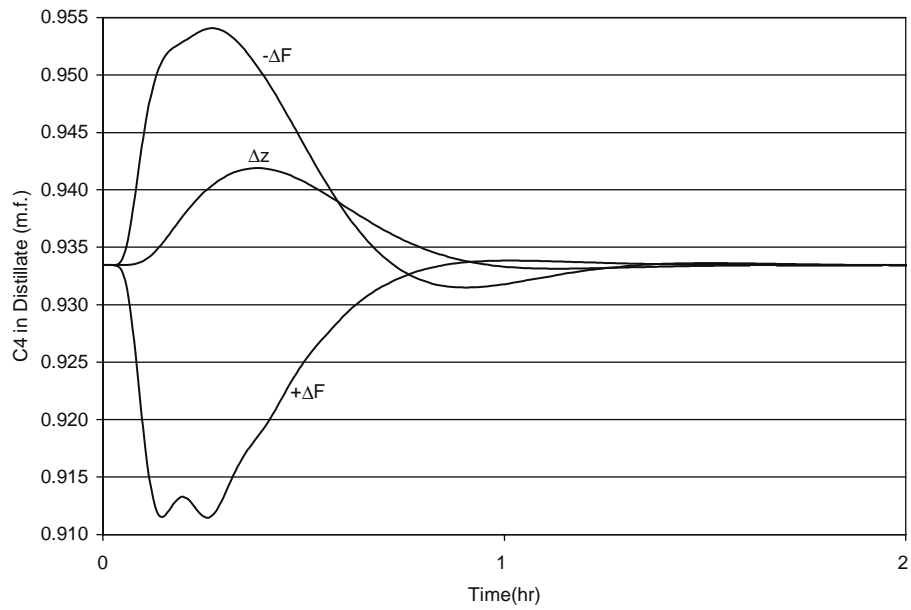


Fig. 13. Response of CS1 (Design 2).

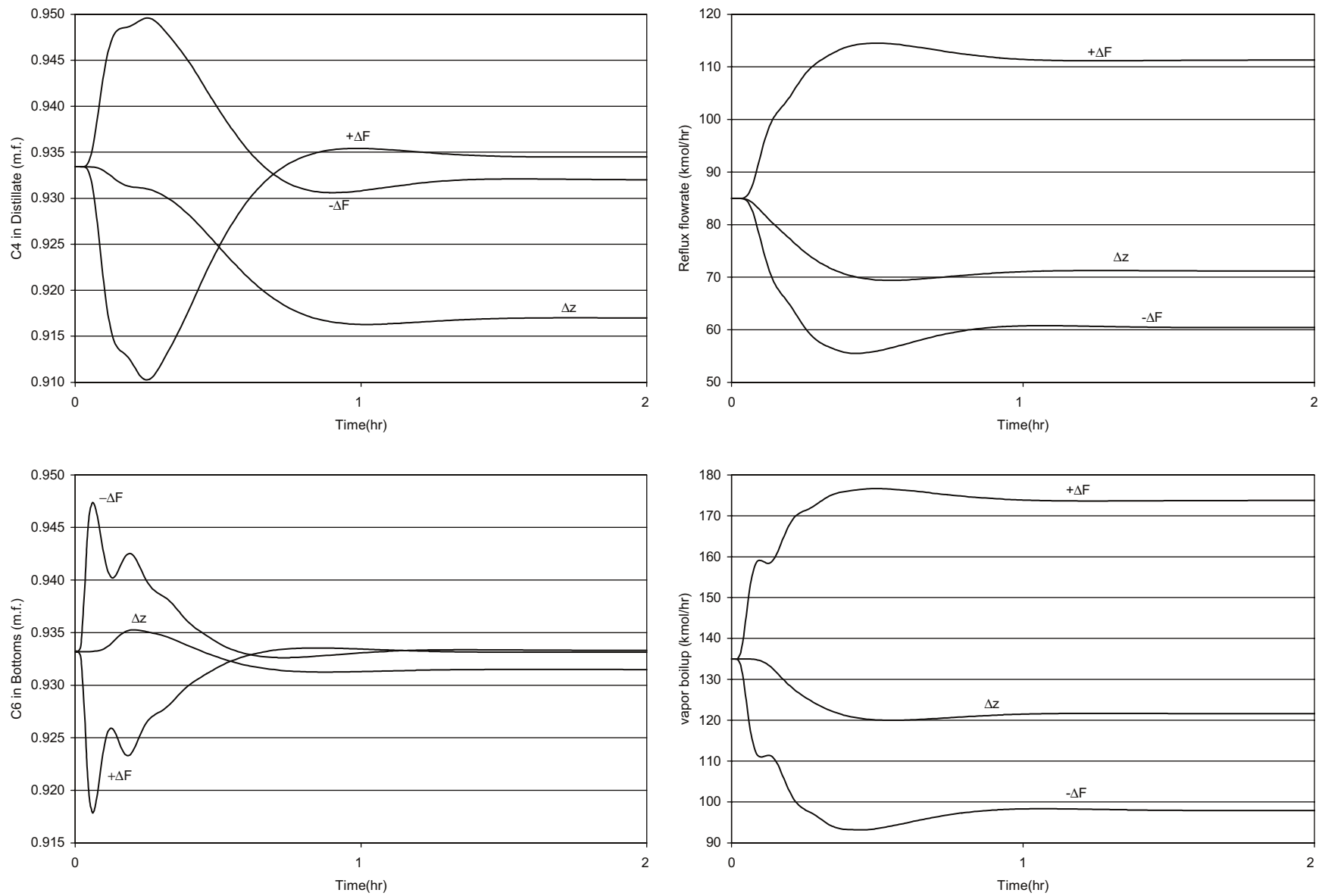


Fig. 14. Response of CS7 (Design 2).



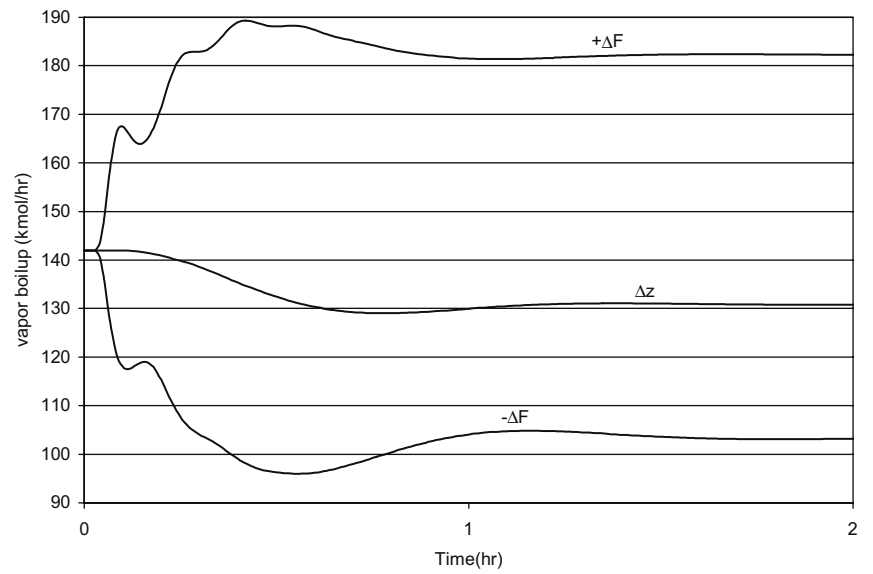
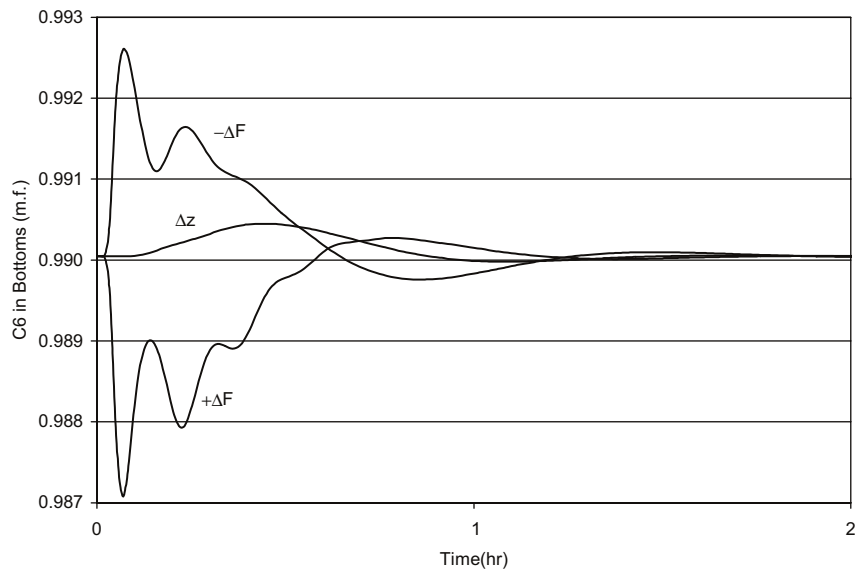
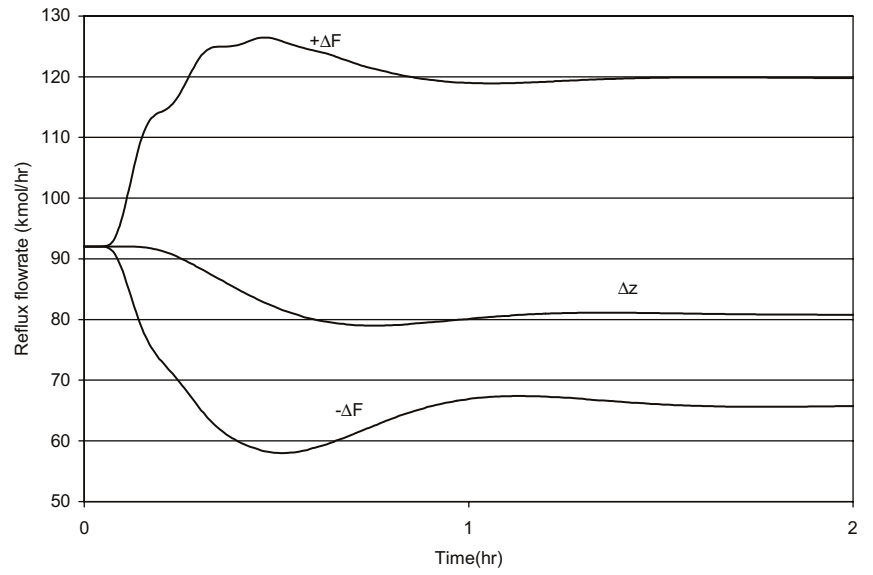
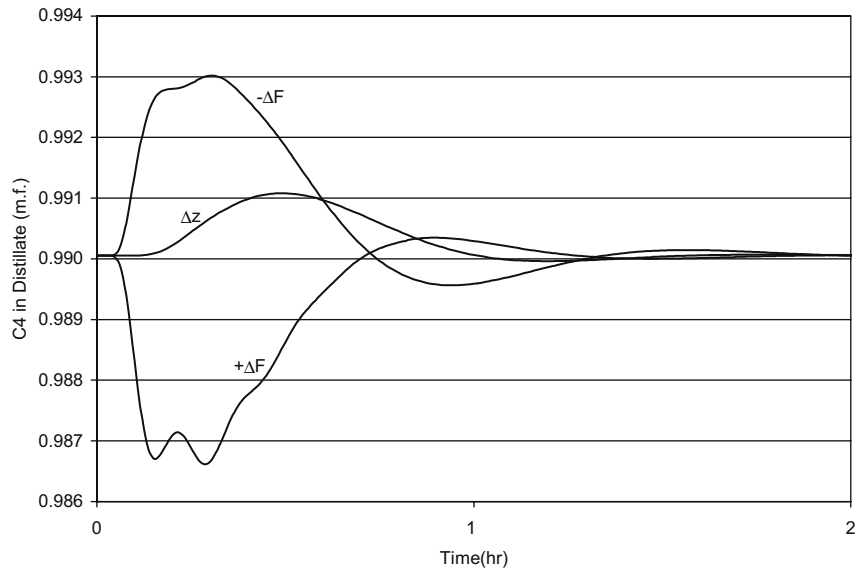


Fig. 15. Response of CS1 (Design 3).

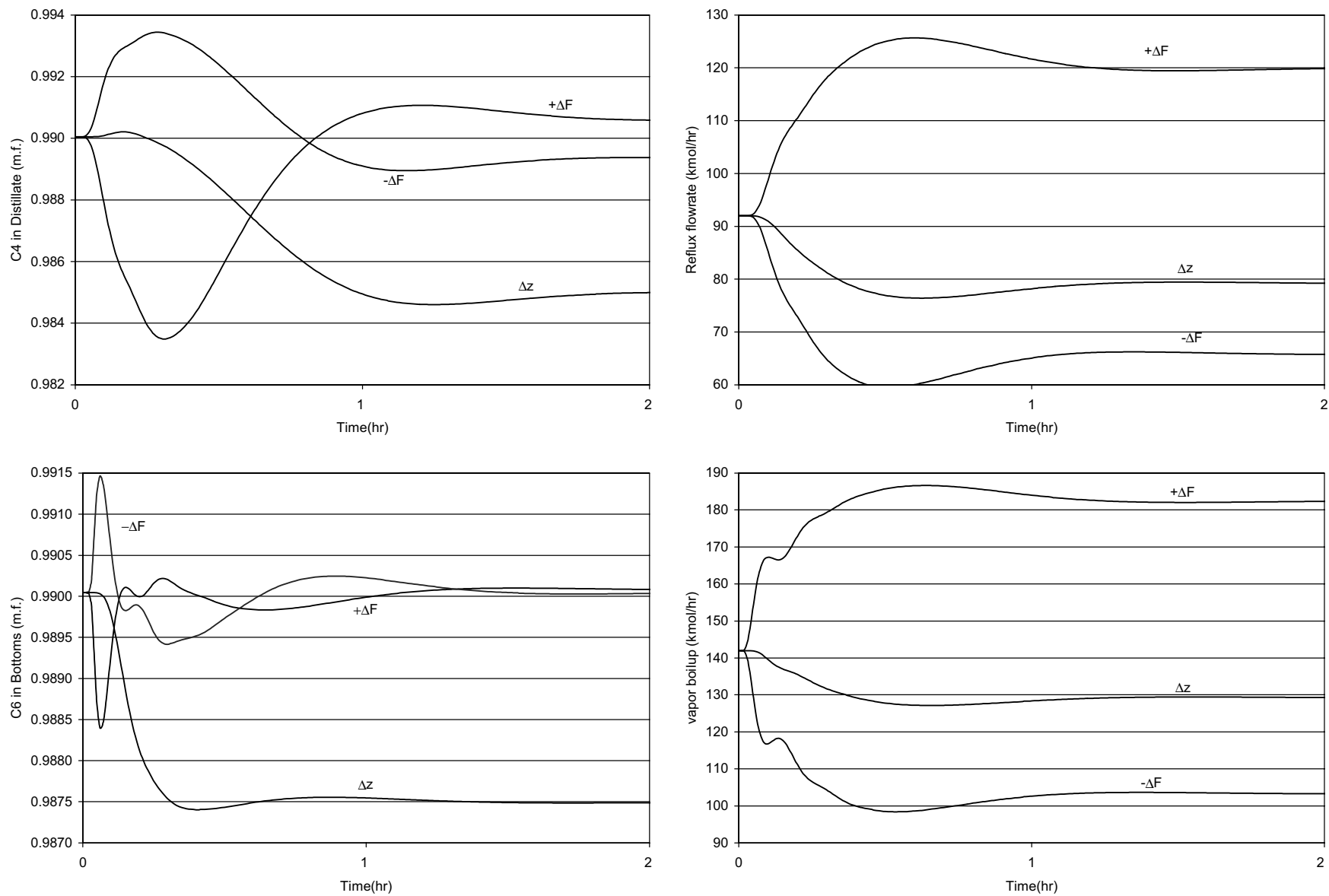


Fig. 16. Response of CS7 (Design 3).

#### 4.7.1. Design 2 (high pressure)

Fig. 13 shows that the CS1 dual composition control structure gives effective control. A comparison of Figs. 8 and 13 shows that the deviations in product purities are less in the high-pressure design. This is because the column has more trays than in the base case.

Fig. 14 gives results for the CS7 dual temperature control structure. The SVD method suggested controlling Tray 2 temperature with vapor boilup and Tray 24 temperature with the reflux flow. Fig. 14 shows that this is an effective structure, but product purity drops about 2% for the feed composition disturbance.

#### 4.7.2. Design 3 (high conversion)

Figs. 15 and 16 show the responses for the three disturbances for two control structures (dual composition and dual temperature). Trays 10 and 31 are used for temperature control. Both structures provide effective control, despite having high product purities. The deviations in product purities are less than that in the base case. Interaction between the loops did not present any problems.

### 5. Control comparison with other systems

The class of reactive distillation studied in this paper is significantly different from those studied in previous papers (ideal, methyl acetate and ETBE) because there is only a single feed. In the other systems, there are two reactant feed streams that must be perfectly balanced if the column is operated “neat” (no excess of one of the reactants). In the ideal and methyl acetate systems, two internal column temperatures could be used as an alternative to having an internal composition analyzer because two products are produced.

In the ETBE system an analyzer is required because there is only one reaction product, although the presence of inerts in the feed require two product streams from the column.

In the metathesis system, there is only one reactant feed stream. Therefore there is no need to have an internal composition analyzer to balance the stoichiometry. The metathesis system is also less nonlinear than the other two systems because of the almost ideal VLE.

### 6. Conclusion

Three different designs for the pentene metathesis are studied, starting with a base case design from the literature. The use of a cooling water in the condenser requires higher

operating pressures than that used in base case. Economics favor columns with more trays than the base case. As the desired conversion increases, more trays are required in the column.

Several control structures are examined for the three designs. Both dual composition and dual temperature control structures provide effective control. Differences in the performance for some control structures in conventional distillation columns and reactive distillation columns are pointed out.

### Notation

$B$	bottoms flowrate, kmol/h
$D$	distillate flowrate, kmol/h
$N$	total number of trays
$N_F$	feed tray
$N_T$	total number of trays
$R$	reflux flowrate, kmol/h
RR	reflux ratio
TAC	total annual cost
$V_S$	vapor boilup, kmol/h

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