Active Constraint Regions for Optimal Operation of Distillation Columns

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Active Constraint Regions for Optimal Operation of Distillation Columns

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ABSTRACT: When designing the control structure of distillation columns, with optimal operation in mind, it is important to know how the active set of constraints changes with disturbances. This issue has received little attention in the literature. This paper applies a procedure presented in an earlier paper, to find how the active constraints for distillation columns change with variations in energy cost and feed flow rate. The production of the most valuable product is maximized by keeping its purity on the minimum allowed, that is, by keeping the valuable product on spec. This is the "avoid product giveaway" rule, which means that the purity specification of the valuable product is always an active constraint. We find that this rule generally holds for distillation for the common case when the price of the on-spec product is independent of purity. The paper includes three case studies: a single distillation column with constant product prices, a single column where the price of the most valuable product is dependent on purity, and two distillation columns in series. In all three case studies, there is a bottleneck, corresponding to a feed flow rate above which the column(s) cannot operate without breaking constraints.

■ INTRODUCTION

The literature on control of distillation processes is vast; some examples include Luyben et al.[1] (general), Waller et al.[2] (sensitivity to disturbances), Skogestad and Morari[3] (selection of control structure), and Nagy et al.[4] (advanced control). The surveys by McAvoy[5] and Skogestad,[6] covering the 1980s, illustrate just how widely the area has been researched. However, few papers deal with optimal operation where a key issue is to identify the active constraints. The two main main exceptions are Maarleveld et al. from 1975[7] and Gordon from 1986[8] who both discuss the issue of active constraints for distillation columns. Apart from these two papers, this issue has received little attention. This is strange considering that optimal control of any process plant is completely dependent on which process constraints are active at the operating point.

Also, when designing ordinary feedback control schemes, knowing the active constraint regions is important. For example, when seeking a *self-optimizing control structure*,[9] one needs to know which variables are constrained and which are not. The active constraints are always selected as controlled variables to be used for feedback, in the case of output constraints, or simply set to be constant, in the case of input constraints. Also, a control structure which works fine for one set of active constraints may be infeasible for another. In some cases, it may be necessary to switch to another control structure, whereas in other cases one control structure may be optimal in one region and near-optimal in neighboring regions.

It is easy to understand how active constraints can influence on the choice of control structure; if a variable is optimally at its constraint value, it cannot be used to control another variable without accepting economic loss. This is because when we want to use the constrained variable for control, we cannot keep it at its optimal value at all times. How the optimal states of the model vary with disturbances is often formulated as a so-called *multiparametric programming* problem.[10−12] One seeks to find a solution which itself is parametrized by the disturbances. The problem can be linear, quadratic, or some other nonlinear type of problem; in chemical engineering applications like distillation, the latter is almost always the case.

In our previous paper,[13] we outlined a procedure for sketching the active constraint regions for chemical processes using few optimizations and applied it on a "toy example". In this paper, we seek to use this method on distillation columns. The paper is structured as follows:

First, we discuss optimal operation of distillation columns. Next, the case studies are described, and the main results are given. We then give a short summary of the method for identifying active constraint regions from ref 13, discuss the efficiency of the method, and go more in detail about optimal operation of distillation columns. We finally discuss the *avoid product giveaway* rule. The following case studies are included:

> Case study Ia. A single distillation column, constant product prices
> Case study Ib. A single distillation column where the distillate price is proportional to purity x_D
> Case study II. Two distillation columns in sequence, again with constant product prices

In all three case studies, we use a simple distillation model with 40 equilibrium stages with the feed entering at the middle stage. The model uses the following assumptions: constant relative volatilities, constant molar overflow, constant pressure over the entire column, equilibrium at every stage, and negligible vapor holdups. This is the "Column A" model used in ref 14, but the product purity specifications are more lax (95% for one product versus 99% for both products for column A). The relative volatility is 1.5 for cases Ia and Ib with a single column (as for "column A"), but for case II with two columns,

it is 1.33 for the A/B split in the first column, and 1.5 for the B/C split in the second column. In the discussion, we give some more insight into the behavior of objective and constraint functions for the optimization problem in case study Ia.

■ OPTIMAL OPERATION OF DISTILLATION COLUMNS

Form of the Optimization Problem. The optimization problem we are dealing with is a nonlinear steady-state problem on the form

$$\min_{u} \quad J(x, u, d)$$

$$\text{subject to} \quad f(x, u, d) = 0$$

$$c(x, u, d) \leq 0 \tag{1}$$

where J is the economical objective, $f(x,u,d)$ is the process model equation, and $c(x,u,d)$ are the process constraints. x represents the internal variables (states) in the process model, u are the variables we may manipulate (specify), and d are the disturbances. $c \leq 0$ is generally a vector of constraints, and a particular constraint $c_i \leq 0$ is said to be *active* if at the optimal operating point we have equality, that is, $c_i = 0$. A constraint is inactive if $c_i < 0$.

In ref 13, we have elaborated more on various formulations, how the optimality conditions can be related to the suggested method for finding active constraint regions as a function of the disturbances d, and the significance of the Lagrange multipliers at the optimal solution. In short, the latter can be summarized as follows: *At the optimal solution, the magnitude of the Lagrange multiplier λ tells us how much we sacrifice in terms of the objective when we back off from an active constraint.*

To define the problem, we need to formulate a cost function which captures the plant economy we are interested in. This cost function is to be minimized with respect to the available degrees of freedom u (while satisfying given constraints c) for the expected range of process disturbances d.

Plant Economics and Objective Function. In distillation, the cost is related to the feed streams, as well as heating and cooling (and possibly other utilities, like pumping). The profit comes from selling the products. To optimize operation on a short time scale, say within a few hours, there is no need to include fixed costs such as capital cost, manpower, and maintenance. The operational objective (cost) to be minimized can be written as

$$J = \sum_{i=1}^{n_f} p_{f,i} F_{f,i} + \sum_{j=1}^{n_U} p_{U,j} F_{U,j} - \sum_{k=1}^{n_P} p_{P,k} F_{P,k} \tag{2}$$

Here, n_f, n_U, and n_P are the number of feed streams, utility streams, and product streams, respectively. Correspondingly, p stands for the price of each stream in ($/mol) and F stands for the feed flow rate (in mol/s).

For a single distillation column with one feed stream and two products, no side streams, and no heat integration, we may simplify the cost function to

$$J_1 = p_F F + p_L L + p_V V - p_D D - p_B B \tag{3}$$

where F, L, V, B, and D are the flow rates of feed, reflux, boilup, bottoms, and distillate, respectively; see Figure 1. The objective function can in most cases be simplified further. For a given feed, an energy balance for the column gives that the reboiler
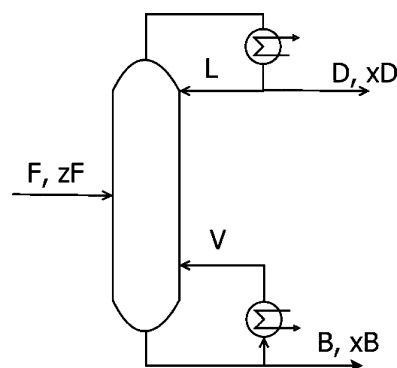
Figure 1. Conventional distillation column with one feed and two products.

heat duty and condenser heat duty, and also the internal liquid and vapor flows inside the column, are nearly proportional. It is therefore reasonable to combine the terms to give a new and simpler objective function

$$J_2 = p_F F + p_V V - p_D D - p_B B \tag{4}$$

where p_V in eq 4 is approximately equal to $p_L + p_V$ in eq 3. This is the form of objective function we will use in the case studies included in this paper. (Proof: The utility terms can be written as

$$p_L L + p_V V = p_L \Delta H_{vap,L} |Q_C| + p_V \Delta H_{vap,V} |Q_B| \tag{5}$$

where Q_C is negative. For a given feed (Fh_F), and approximately constant product composition (Dh_D and Bh_B approximately constant), we have $\Delta Q_C = -\Delta Q_B$, so change in reboiler duty and condenser duty are the same, and since a constant term does not matter for the optimization, it may be combined into a single term in eq 4 where p_V in eq 4 is equal to $(p_L \Delta H_{vap,L} + p_V \Delta H_{vap,V})/\Delta H_{vap,V}$ in eq 3 QED).

Provided that we satisfy the product specifications, the prices are usually constant (independent of process states). However, in some cases, they may depend on product quality, for example, if we pay or get paid only for the valuable component in a stream, in which case the price can be written

$$p' = px \tag{6}$$

where x is the mole fraction of the component we get paid for, and p is the price for the pure component. In this paper, we include one case study where we only get paid for the light component in the distillate, i.e., $p_D' = p_D x_D$ where x_D is the mole fraction of light (valuable) component.

Degrees of Freedom. As explained in ref 15, when we assume a given feed and given pressure, a distillation column has got two steady-state degrees of freedom. Dynamically, there are four remaining manipulated variables, but there are two levels that need to be controlled dynamically, but which have no steady-state effect. The two degrees of freedom can, for example, be selected as two flow rates,

$$u = [L \; V] \tag{7}$$

but any pair of two independent specifications can be used. For example, we may control (i.e., specify) the concentration of the key impurity in each product stream or two tray temperatures. For optimization, we should choose as degrees of freedom the variables that make the problem easiest to solve numerically.

Constraints. The constraints ($c(x,u,d) \leq 0$) will typically consist of product purity requirements and restrictions on

operating conditions. Typically, there will be maximum and minimum limits on internal flows (in a column, defined by the weeping and flooding points[16]). In addition, there may be restrictions on column pressure, since column pressure has a big influence on condensation and evaporation temperatures. The product purity constraints are usually expressed in terms of the mole fraction of the main component in each stream:

- Distillate specification: $x_D \geq x_{D,min}$ where x_D is the mole fraction of light key component in D
- Bottoms specification: $x_B \geq x_{B,min}$ where x_B is the mole fraction of heavy key component in B

For a two-component mixture, this is unproblematic, but for a multicomponent mixture, this constraint formulation may cause problems, as there may be more than one composition that satisfies the constraint (except for the lightest and heaviest components). Thus, one may get a more robust problem specification by instead giving the following specifications:[17]

- Distillate specification: $x_D \leq x_{D,max}$ where x_D is the mole fraction of heavy key impurity in D
- Bottoms specification: $x_B \leq x_{B,max}$ where x_B is the mole fraction of light key impurity in B

Regarding the capacity constraints, there may be many just in one column: maximum available reboiler heating or condenser cooling, flooding and weeping points, and possibly maximum flows of product streams due to a potential downstream bottleneck. In this paper, we simplify and assume that it is sufficient to specify a maximum vapor boilup V

$$V \leq V_{\max} \tag{8}$$

In the case study with two columns, we use different values of V_{max} for the two columns.

Disturbances. The disturbances (d) are the variables that influence on either the process or the economical objective but which we cannot influence. For a distillation column, or a chemical process unit in general, the feed conditions (flow rate, temperature, pressure, and composition) are important disturbances. The constraint values (including purity specifications and flow limits) are also generally important disturbances. Finally, prices of feeds, products, and energy will be subject to change and should also be considered disturbances. In the case studies included here, we consider only the feed flow rate (F) and energy cost (p_V) as disturbances.

The main reason for not including other disturbances, for example, in feed composition, feed enthalpy, or product purity specifications, is that it is difficult to show the constraints regions graphically when there are more than two disturbances. We believe that the two selected disturbances in feed rate and relative energy price are very relevant in most applications. In practice, one should focus on the disturbances that are expected to be important for future operation, as a complete map of all disturbances will be very time-consuming.

■ CASE STUDIES

Case Study Ia: One Distillation Column, Constant Product Price. For the first case study, we consider a single distillation column with 41 stages and feed entering at stage 21, separating a feed mixture of equal fractions of A and B, with

relative volatility $\alpha_{AB} = 1.5$. The optimization problem may be formulated as follows:

$$\min_{u} \quad J(u, d) = p_F F + p_V V - p_B B - p_D D$$

$$\text{subject to:} \quad x_B \geq x_{B,\min}$$

$$x_D \geq x_{D,\min}$$

$$V \leq V_{\max} \tag{9}$$

where $u = [L \ \ V]$ and $d = [F \ \ p_V]$. The model equations (component mass balances for each stage and equilibrium calculations) are solved explicitly and are thus not shown in eq 9. The constraints on x_B and x_D refer to the mole fractions of the main component in each stream (component B in the bottoms stream (B) and component A in the distillate stream (D), respectively). The p values refer to the prices of each respective stream. The prices and other data used in this case study are shown in Table 1.

Table 1. Data Used for Case Studies Ia and Ib

variable	value
α_{AB}	1.5
z_F	0.5
F	variable (1−1.6 mol/s)
p_F	1$/mol
p_B	1$/mol
p_D	2$/mol
p_V	variable (0.01−0.02 $/mol)
$x_{B,min}$	0.9900
$x_{D,min}$	0.9500
V_{max}	4.008 mol/s

The case study uses the feed rate F and the energy price p_V as disturbances, and our goal is to establish the regions where the constraints on x_B, x_D, and V are active, while using as few optimizations as possible. The feed rate F varies from 1.1 to 1.6 mol/s. The prices for the feed (F) and bottoms product (B) are both set at a reference price of 1 $/mol, whereas the valuable distillate product (D) is 2 $/mol.

In our case study, the relative energy price p_V/p_F varies between 0.01 and 0.02, and as we argue in the following, this is a reasonable relative price range. For hydrocarbon feed mixtures, the energy can be generated by burning some of the feed, and since the heat of combustion is about 100 times larger than the heat of vaporization for hydrocarbons, we expect for hydrocarbons that p_V/p_F should be about 0.01 (or less, if cheaper energy sources are available). However, in general, for other feed mixtures, the relative energy price can vary greatly, from 0 and up to a value similar to the feed and products (about 1). Also note that energy prices can vary greatly from one day to the next, depending on external conditions and prices. For cryogenic applications, where cooling rather than heating is costly, the relative energy price p_V/p_F may exceed 0.02 or more, even for hydrocarbon mixtures. The reason is that cryogenic cooling requires electricity as the energy source.

Let us now generate the active constraint regions as a function of the two selected disturbances (F, p_V). To start, we use our knowledge about the nature of the process model and the optimization problem to state the following:

1. With $N_C = 3$ inequality constraints, there may be at most $2^{N_C} = 8$ sets of active constraints, possibly including

infeasible regions where there are more active constraints than degrees of freedom. Here, we have two degrees of freedom (u), so the region with three active constraints will be infeasible. As we will conclude later, there are only three regions in this case.

2. The constraint on x_D will be active for all values of (F, p_V). This is because separating this stream to a higher purity will require that we reduce the flow rate D or increase the internal streams L and V. Since we do not get paid for the increased purity, this is not profitable. In other words, we should seek to *avoid product give-away*.[8,18] This rule is discussed in more detail later.

3. At low energy cost p_V, the constraint on x_B will be inactive, meaning that we should overpurify the bottom product. This is because we get a better price for the distillate, and by overpurifying the bottom product, we move component A from bottoms to distillate. This is profitable when energy is cheap.

4. As p_V increases, the optimal value of x_B decreases, and at $p_V = p_{V,1}$, it reaches $x_{B,min}$. Since the column stage efficiency is assumed constant at 100% (rather than dependent on flow), the value of $p_{V,1}$ is independent of F.

5. Bottleneck: There exists a maximum feed rate F_{max}, above which we cannot achieve feasible operation, i.e., satisfy all three constraints. This can be seen from a simple degree of freedom consideration: Assume we keep both purities at their constraint values by adjusting L and V. As we increase F, all other flows, including L and V will increase proportionally. Eventually, we reach $V = V_{max}$, where a further increase in F will force us to break one of the purity constraints.

6. From the above, we can conclude that we will have three feasible regions: (I), x_D active; (II), x_D and V_{max} active; and (III), x_D and x_B active. Regions I and III will be separated by a straight line (as explained above). The same goes for the border between the infeasible Region (IV) and the others. See Figure 2.
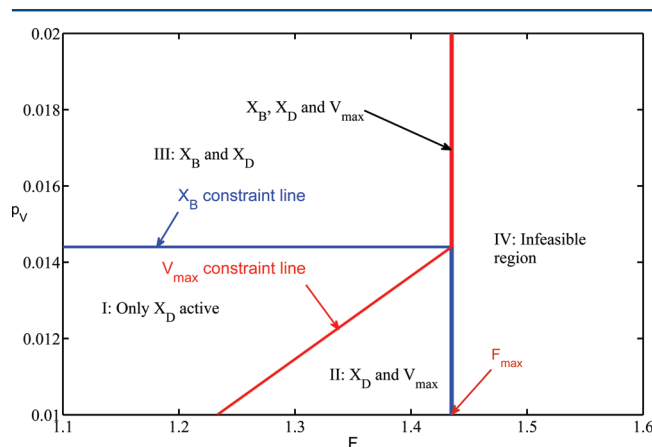
Figure 2. Active constraint regions for single column with fixed prices (case Ia).

7. The border between Regions I and II intersects with the border between Regions I and III exactly at F_{max}. The border between Regions I and II is the only one for which we cannot say *a priori* whether it will be straight or not.

Using the method described in our previous paper,[13] we obtain the following numerical values, which are sufficient for sketching the active constraint regions (shown in Figure 2):

- The maximum feed rate is $F_{max} = 1.435$ mol/s.
- For all $F < F_{max}$, $x_B = x_{B,min}$ is active when $p_V > 0.0144$ \$/mol.
- At $p_V = 0.01$, $V = V_{max}$ is active for $F > 1.233$ mol/s.

Notice that the line between Regions I and II is shown as being straight. This is because it is based on only two data points; in reality, it is slightly curved. The constraint lines in Figure 2 are as follows:

- Red: V_{max} becomes active.
- Blue: x_B becomes active.

The vertical parts of these two constraint lines indicate $F = F_{max}$. Table 2 lists optimal data at selected points in the disturbance space (all flows are in [mol/s]).

Table 2. Single Column (Case Ia): Values of Key Variables at Selected Disturbances (F,p_V)[a]

region(s)	I	II	III
F [mol/s]	1.2	1.4	1.3
p_V [\$/mol]	0.01	0.01	0.015
L [mol/s]	2.827	3.276	2.949
V [mol/s]	3.454	**4.008**	3.627
D [mol/s]	0.627	0.731	0.678
B [mol/s]	0.573	0.669	0.622
x_D	**0.9500**	**0.9500**	**0.9500**
x_B	0.9920	0.9920	**0.9900**
J [\$/s]	−0.536	−0.625	−0.566

[a]Numbers in bold indicate active constraints.

In Figure 3, we show how the Lagrange multipliers λ for the active constraints behave at $p_V = 0.01$. As expected, the λ
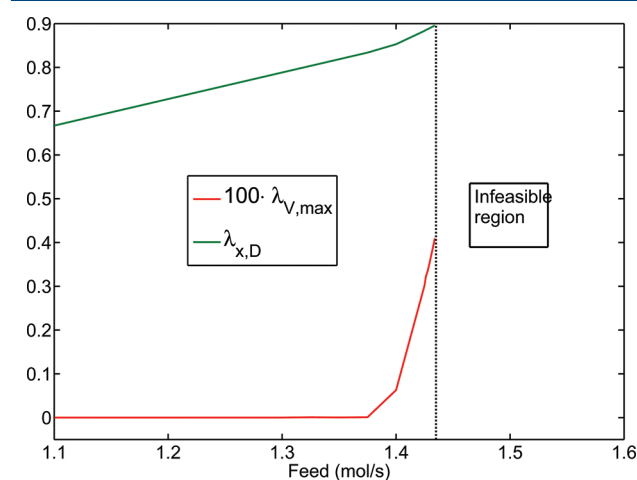
Figure 3. Single column (case Ia): Lagrange multipliers for active constraints at $p_V = 0.01$.

corresponding to x_D is always positive. The λ corresponding to V_{max} becomes nonzero at $F = 1.233$ mol/s. Notice that it increases very slowly up to $F = 1.37$; this means that, up to $F = 1.37$, this constraint does not influence strongly on the plant objective.

Case Study Ib: One Distillation Column, Variable Product Price. In the second study, the constraints are the

same, but the objective function is altered to make the price of the distillate stream proportional to its purity, that is, one gets paid for the valuable component only:

$$J(u, d) = p_F F + p_V V - p_B B - p_D' D \qquad (10)$$

where

$$p_D' = p_D x_D \qquad (11)$$

p_D and the other prices are the same as in case study Ia (Table 1). The consequence of the varying price p_D' is that we now may have additional regions where the constraint on x_D is *inactive*. The reason for this is: When energy is cheap enough, we may overpurify the distillate D without giving away anything (as we get paid for the extra component A in the distillate D). Since component B in the distillate is now worthless, it is profitable to send it to the bottom instead. As the energy cost p_V increases, the purity constraints become active. Just as for the first case, we can deduce some things about the active constraint regions *a priori*, before carrying out any optimizations:

1. As above, theoretically, there may be at most 7 regions (since the region with three active constraints is infeasible). As we will show, only five of these regions are present.
2. The lines separating the different regions where only purity constraints are active, will be horizontal. In the following, the p_V values corresponding to these lines will be referred to as $p_{V,1}$ and $p_{V,2}$.
3. We will have a region where no constraints are active and one where $V < V_{max}$ is the only active constraint.
4. We will have a region where *one* purity constraint is active and $V = V_{max}$. The border between this region and the previous one will be vertical. This can be explained as follows: When V is fixed at V_{max}, p_V has no influence on the optimal solution; the F value for which the next constraint becomes active is independent of p_V, and thus, the line is vertical. This F value will be referred to as F_1.
5. As for the previous case, we have a value of F for which all constraints are active, and for any higher F value, we cannot satisfy all constraints. This value will be referred to as F_{max}.
6. The two regions with two active constraints will meet in a point on the line $F = F_{max}$, just like Regions II and III in Figure 2.
7. As $p_V \rightarrow 0$, the value of F at which $V_{optimal} = V_{max}$ will also approach 0. This is because, when the energy utility is free and there is a benefit from extra purity, we want to maximize both purity and flow rate of the distillate stream. This means we can use the point $F = 0$, $p_V = 0$ when constructing the diagram.

This means if we take the assumption that the regions with $V = V_{max}$ are also separated by straight lines from the regions where $V < V_{max}$, we are left with the task of finding just a few values for F (F_1 and F_{max}) and p_V ($p_{V,1}$ and $p_{V,2}$). The actual values are shown in Table 3, and the resulting active constraint regions are shown in Figure 4. Table 4 gives optimal data at selected points in the disturbance space (one point inside each region). In Figure 4, we have three constraint lines:

- The *red* constraint line indicates where x_B becomes active.
- The *blue* constraint line indicates where V_{max} becomes active.

Table 3. Single Column (Case Ib): Values for F and p_V Needed to Draw Figure 4

variable	value
F_1	1.23
F_{max}	1.44
$p_{V,1}$	0.014
$p_{V,2}$	0.106
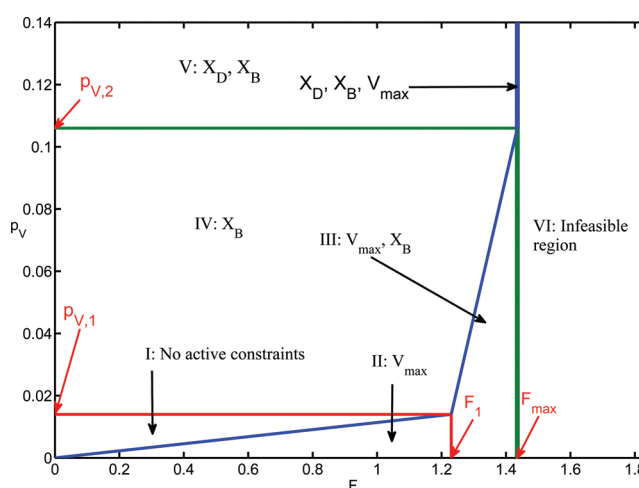
Figure 4. Single column (case Ib): Active constraint regions with purity-dependent distillate price ($p_D' = p_D x_D$).

Table 4. Single Column (Case Ib): Values of Key Variables at Selected $(F, p_V)^a$

region(s)	I	II	III	IV	V
F [mol/s]	0.7	1.2	1.3	1.0	1.0
p_V [\$/mol]	0.013	0.010	0.010	0.05	0.12
L [mol/s]	2.048	3.408	3.355	2.443	2.268
V [mol/s]	2.423	**4.008**	**4.008**	2.951	2.790
D [mol/s]	0.3749	0.600	0.653	0.508	0.521
B [mol/s]	0.3751	0.600	0.647	0.492	0.479
x_D	0.9905	0.9918	0.9853	0.9755	**0.9500**
x_B	0.9901	0.9914	**0.9900**	**0.9900**	**0.9900**
J [\$/s]	−0.3411	−0.5558	−0.6004	−0.3605	−0.1969

[a]Numbers in bold indicate active constraints.

- The *green* constraint line indicates where x_D becomes active.
- At $F = F_{max}$, the blue and green constraint lines are vertical, and they intersect in the point $(F_{max}, p_{V,2})$.

It is also worth noticing that the value of p_V for which x_D becomes active (p_{V_2}) is quite high. This is because the number of stages in the column (column A[14]) was designed to obtain 99% purity in both ends. This makes overpurifying cheap in terms of energy.

Case Study II: Two Distillation Columns in Sequence. In the third and final case study, we consider two distillation columns in series, both with the same number of stages (41) and feed entering at stage 21. The feed now contains three components, A, B, and C, where A is the most volatile, C is the least volatile, and B is the most valuable product. The two columns with stream names are shown in Figure 5. The relevant parameters are summarized in Table 5. For simplicity, we will refer to x_{A,D_1}, x_{B,D_2}, and x_{C,B_2} simply as x_A, x_B, and x_C, respectively.
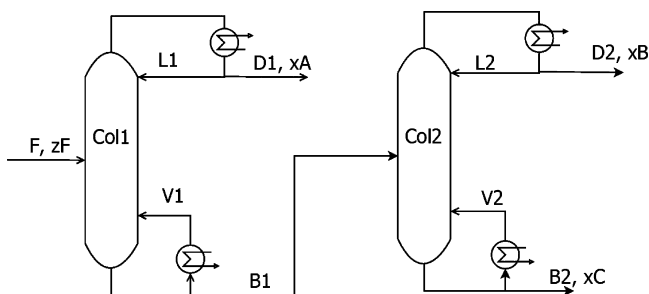
Figure 5. Two distillation columns in sequence.

Table 5. Data for Two Columns (Case II)

variable	value
α_{AB}	1.333
α_{BC}	1.5
zF	[0.4 0.2 0.4]
$V_{1,max}$	4.008 mol/s
$V_{2,max}$	2.405 mol/s
$x_{A,min}$	0.9500
$x_{B,min}$	0.9500
$x_{C,min}$	0.9500
p_F	1$/mol
p_A	1$/mol
p_B	2$/mol
p_C	1$/mol
p_V	variable

The objective function is again formulated as a function of (u,d)

$$J(u, d) = p_F F + p_V (V_1 + V_2) - p_A D_1 - p_B D_2 - p_C B_2 \tag{12}$$

with $u = [L_1 V_1 L_2 V_2]$ (this choice of u is not unique, and in the optimization, we actually use the four product compositions as degrees of freedom) and $d = [F\ p_V]$.

The constraints are defined as follows

$$x_A \geq x_{A,min}$$

$$x_B \geq x_{B,min}$$

$$x_C \geq x_{C,min}$$

$$V_1 \leq V_{1,max}$$

$$V_2 \leq V_{2,max} \tag{13}$$

Just as for the case of one column, we can deduce some things about the active constraints regions before carrying out optimization:

- There can be at most $2^5 = 32$ regions, of which 31 will be feasible (we have four independent inputs, so we cannot satisfy all five constraints). We will show that the actual number of regions in this case study is 8.
- There exists a value F_{max} above which we cannot satisfy all constraints.
- D_2 is the most valuable product stream, so the constraint on x_B will remain active for all disturbances d. This reduces the maximum number of regions to 16 (of which 15 are feasible).
- As for *both* one-column cases, the lines separating the regions with only purity constraints active will be

horizontal. The p_V values corresponding to these lines will be referred to as $p_{V,1}$ (the lower value) and $p_{V,2}$.

- Above $p_{V,2}$, there will be a region where all three purity constraints are active while both capacity constraints are inactive. The F value for which the first capacity constraint becomes active will be independent of p_V, meaning the region in question will be bordered to the right by a *vertical* line at an F we shall refer to as F_1.
- For low p_V, we will have a region where the constraint on x_B will be active along with the constraints on V_1 and V_2. At some value of F, one of the remaining purity constraints will become active. This F value is independent of p_V, because the term $p_V(V_1 + V_2)$ in the objective function is constant in this region, meaning this region will also be bordered by a vertical line to the right. This F value is referred to as F_2.
- At $p_{V,1}$, one of the capacity constraints (V_1 or V_2) will become active at some $F < F_{max}$. We refer to this value as F_3.

To sketch the active constraint regions, we need to find $p_{V,1}$, $p_{V,2}$, F_1, F_2, and F_{max}. These are summarized in Table 6. For all

Table 6. Two Columns (Case II): Values for F and p_V Needed to Draw Figure 4

variable	value
F_1	1.442
F_2	1.469
F_3	1.458
F_{max}	1.489
$p_{V,1}$	0.0382
$p_{V,2}$	0.1441

the region borders that are neither vertical nor horizontal, we also need an additional data point in addition to their intersection with one of the already established lines. By applying the same method as above, we come up with the regions shown in Figure 6. The constraint lines are given different colors:

- Along the *red* line, x_A becomes active.
- Along the *orange* line, V_1 becomes active.
- Along the *blue* line, V_2 becomes active.
- Along the *green* line, x_C becomes active.
- The black line indicates F_{max}, which is reached when x_C once again becomes active.

In the following discussion, we will refer to the various regions in Figure 6 as indicated in Table 7. Note that Region IX, with five active constraints, corresponds to the vertical black line in Figure 6.

When we examine Figure 6, we notice two things that may at first seem surprising.

- The line separating Regions II and V (part of the *orange* constraint line) has a negative slope. Thus, we have that the optimal value of V_1 *increases* with increasing p_V, which seems counterintuitive. However, this is compensated by a decrease in V_2; the sum $V_1 + V_2$ is actually decreasing, which is what we would expect.
- The next interesting feature about Figure 6 is that the border between Regions V and VII (part of the *green* constraint line) is *not* horizontal. Across this border, the constraint on x_C switches between active and inactive. The reason for this one not being horizontal is the
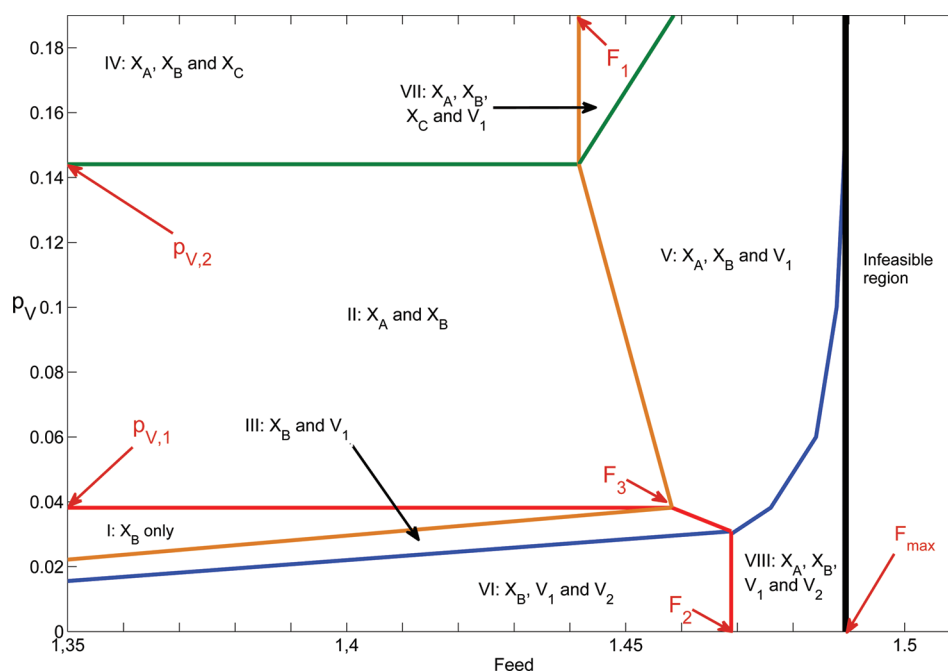
Figure 6. Two columns (case II): Active constraint regions.

Table 7. Two Columns (Case II): Active Constraints in Each Region

region number	constrained variable(s)
I	x_B
II	x_A, x_B
III	x_B, V_1
IV	x_A, x_B, x_C
V	x_A, x_B, V_1
VI	x_B, V_1, V_2
VII	x_A, x_B, x_C, V_1
VIII	x_A, x_B, V_1, V_2
IX	x_A, x_B, x_C, V_1, V_2

following: When starting with only the three purity constraints active, an increase in F leads to a proportional increase in all streams, until the first capacity constraint becomes active (in this case, this means V_1). Now, since V_1 is not allowed to increase further, either any extra A fed to the system must go to stream D_1, meaning the constraint on x_A is no longer active, or more A goes through to the second column where it enters the distillate stream D_2. Thus, we need to put more C into stream B_2, thus making the constraint on x_C inactive. Thus, one of two purity constraints *must* become inactive at this point. Of course, it will become active again once we reach F_{max}.

- The black line indicating F_{max} could be seen as part of the constraint line for x_C. However, the two are not connected; this is why we choose to show them in different colors.

It is also worth noting that the objective function J becomes positive above a fairly low value for p_V (approximately $p_V = 0.05$) for all $F \in \langle 0, F_{max} \rangle$. This means operation in this region is not economically profitable; thus, we would only operate in this region if we have to. Finally, Table 8 shows flow rates and compositions at the optimal solution at selected points in the disturbance space (one point in each of the eight regions).

■ DISCUSSION

Method for Finding Active Constraint Regions. In ref 13, we outlined a method for finding active constraint regions (illustrated with a two-dimensional example). It is based on that, when a constraint c_i changes from active to inactive, the sum of this constraint value and its corresponding Lagrange multiplier λ_i is zero. If we define this sum as s_i, we have that, when constraint i changes between active and inactive,

$$s_i(d) = c_{i,opt}(d) + \lambda_{i,opt}(d) = 0 \tag{14}$$

where $c_{i,opt}(d)$ is the optimal value of c_i given the disturbance d, and $\lambda_{i,opt}(d)$ is the corresponding Lagrange multiplier. For the case of two disturbances, with d_1 on the horizontal axis and d_2 on the vertical axis, the method can be summarized as follows:

1. Use process and problem knowledge to predict if we have any constraints that are either always active or always inactive, thus reducing the number of potential regions.
2. If possible, deduce which constraint will become active first when changing a disturbance value.
3. Predict whether some constraint region boundaries will be independent of one of the disturbances. This corresponds to a horizontal boundary (if it is independent of d_1) or a vertical boundary (if it is independent of d_2).
4. Locate the vertical or horizontal region boundaries, by finding the disturbance value for which $s_i = 0$ for constraint i. When locating a vertical boundary, we hold d_2 constant and find the value of d_1 which gives $s_i = 0$. For a horizontal boundary, we hold d_1 constant instead.
5. For the remaining region boundaries, on which there are no assumptions about being vertical or horizontal, find as many points as desired along each boundary. If a linear approximation is deemed sufficient, one needs just one or two new points for each new boundary (one, if one knows from the previous step where this region boundary intersects with another boundary).

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Table 8. Optimal Data for Selected Points in Case Study II[a]

region(s)	I	II	III	IV	V	VI	VII	VIII
F	1	1.4	1.22	1.35	1.48	1.4	1.45	1.48
p_V	0.01	0.1	0.01	0.1441	0.10	0.01	0.18	0.01
V_1	3.335	3.870	**4.008**	3.754	**4.008**	**4.008**	**4.008**	**4.008**
L_1	2.935	3.290	3.516	3.194	3.396	3.436	3.407	3.396
B_1	0.600	0.820	0.732	0.791	0.869	0.829	0.849	0.869
D_1	0.400	0.580	0.488	0.559	0.611	0.571	0.601	0.612
$x_{A,D1}$	0.9815	0.9618	0.9803	**0.9500**	**0.9500**	0.9618	**0.9500**	**0.9500**
$x_{B,D1}$	0.0185	0.0382	0.0197	0.0500	0.0500	0.0382	0.0500	0.0500
$x_{C,D1}$	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
$x_{A,B1}$	0.0127	0.0112	0.0128	0.0108	0.0127	0.0126	0.0112	0.0127
$x_{B,B1}$	0.3209	0.3061	0.3203	0.3062	0.3056	0.3116	0.3061	0.3056
$x_{C,B1}$	0.6664	0.6828	0.6669	0.6830	0.6817	0.6758	0.6828	0.6817
V_2	1.862	2.015	2.275	1.843	2.310	**2.405**	2.006	**2.405**
L_2	1.662	1.772	2.031	1.618	2.048	2.139	1.764	2.136
B_2	0.400	0.577	0.488	0.566	0.607	0.563	0.608	0.600
D_2	0.200	0.243	0.243	0.225	0.262	0.265	0.242	0.268
$x_{A,D2}$	0.0381	0.0377	0.0381	0.0379	0.0421	0.0392	0.0393	0.0411
$x_{B,D2}$	**0.9500**	**0.9500**	**0.9500**	**0.9500**	**0.9500**	**0.9500**	**0.9500**	**0.9500**
$x_{C,D2}$	0.0119	0.0123	0.0115	0.0121	0.0079	0.0108	0.0107	0.0089
$x_{A,B2}$	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000	0.0000
$x_{B,B2}$	0.0066	0.0346	0.0500	0.0066	0.0274	0.0107	0.0500	0.0176
$x_{C,B2}$	0.9934	0.9654	0.9934	**0.9500**	0.9726	0.9893	**0.9500**	0.9824
J	−0.1480	0.3452	−0.1805	0.5814	0.3699	−0.2013	0.8408	−0.2042

[a]Numbers in bold indicate active constraints.

We have used MATLAB's fmincon solver for optimization and fzero for interpolation to find the points where $s_i(d) = 0$, but in principle, any NLP solver could be used for optimization.

Numerical Issues in Optimization and Region Finding. Despite the three case studies sharing many of the same features, we found that a different optimization approach was better suited for case study II than the one used for case studies Ia and Ib. In case studies Ia and Ib, we used an algebraic equation solver to solve for $((dx)/(dt)(x,u,d)) = 0$, whereas in case study II, we used dynamic simulation to find the steady states needed to calculate $J(u,d)$ and $c(u,d)$. In addition, in case study II, we did not specify L and V directly but used these for control of compositions. This made the dynamic simulation more robust. A reason for this might be that the optimization solver would suggest negative values for V_1 and V_2 as this would obviously reduce the objective function, but this would make problems for the dynamic simulation. (The final solution would obviously not have negative flows, but the active-set method used by fmincon.f allows breaching of bound constraints at intermediate iterates. An alternative could be using the interior-point algorithm, but this algorithm was much slower for this problem.)

One point, which we did not address above, is that we may search directly for the points where two constraint lines intersect. An example is the point $(F_1, p_{V,1})$ in Figure 4. Since two constraints change at the same time in these intersection points, we could try to solve the equation set

$$c_1(d_1, d_2) + \lambda_1(d_1, d_2) = 0$$

$$c_2(d_1, d_2) + \lambda_2(d_1, d_2) = 0 \tag{15}$$

for (d_1, d_2). However, this demands a more sophisticated equation solver, and if this solver is not more computationally efficient, using this approach would defeat the purpose of the method, which is a reduced need of repeated optimizations.

More on optimal operation of a single column. In Case Study Ia, we found active constraint regions for a single distillation column where the product prices p_D and p_B were constant. To give a better understanding of how the active constraints change for differing prices, we will here show how the cost function J depends on bottom purity x_B, for three different energy prices p_V, when distillate purity x_D is fixed at $x_{D,min}$. When x_D is fixed, there is one degree of freedom left, so the remaining variables can be plotted against x_B. In Figure 7,
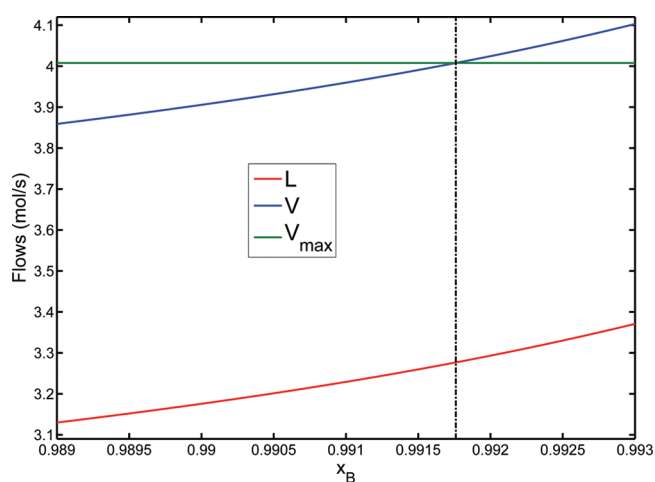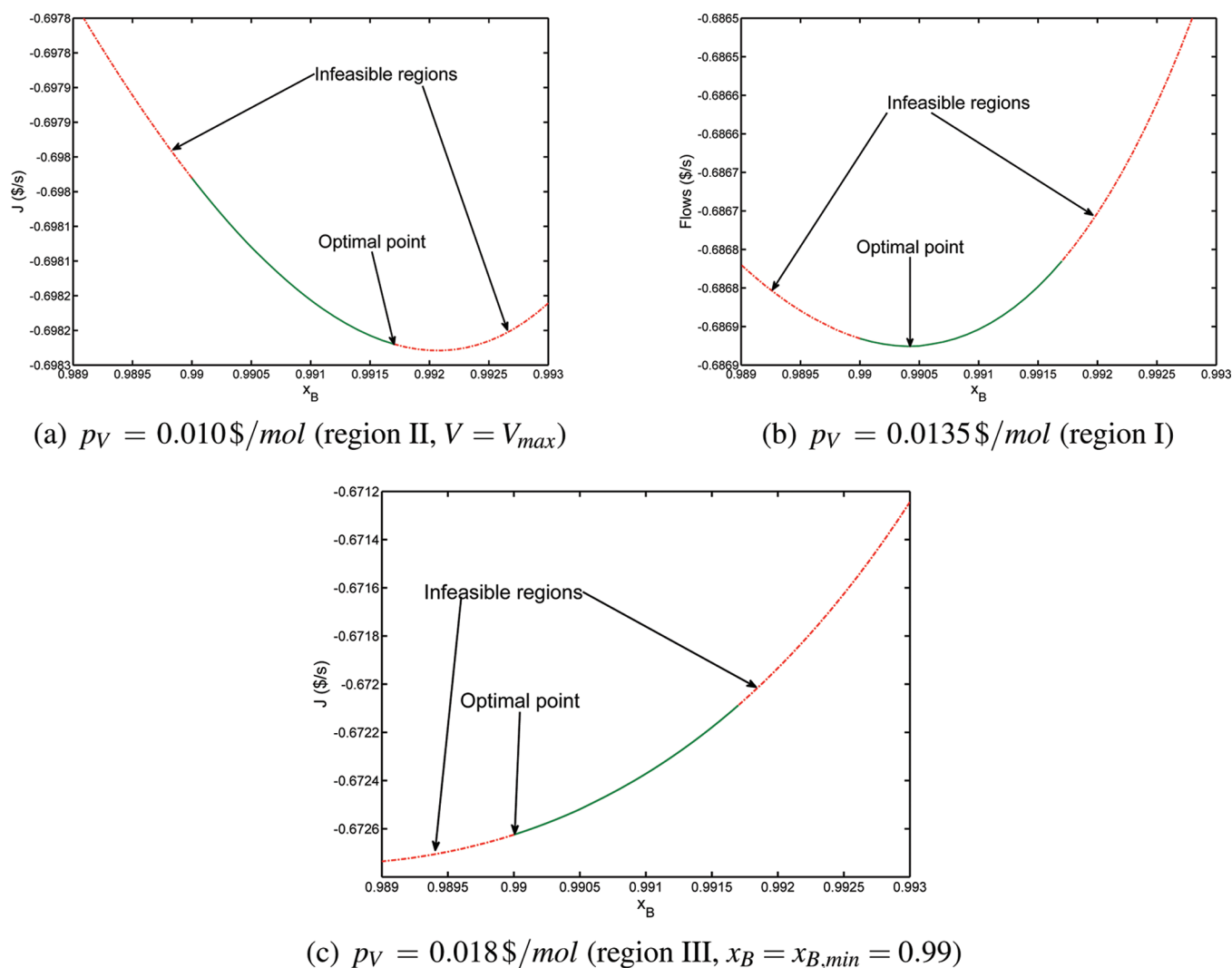

Figure 7. Single column (case Ia): L and V as function of x_B.

we show how reflux L and boilup V vary with increasing amount of light component in the bottom stream. The maximum boilup rate (V_{max}) is also included; the thin black line indicates the highest purity we can achieve without breaking the constraint $V \leq V_{max}$ (it is located at $x_B \approx 0.9917$). All calculations are done at a feed flow rate of $F = 1.4$ mol/s.

(a) $p_V = 0.010\,\$/mol$ (region II, $V = V_{max}$)

(b) $p_V = 0.0135\,\$/mol$ (region I)

(c) $p_V = 0.018\,\$/mol$ (region III, $x_B = x_{B,min} = 0.99$)

Figure 8. Single column (case Ia): Cost function J as function of amount of heavy component in column bottoms for three values of p_V.

Figure 8 shows how the objective function J varies with x_B for three different values of p_V. In each subfigure, the feasible region which lies between the constraints on V and x_B is shown in green, whereas red indicates an infeasible region where a constraint would have to be broken (V_{max} to the right, $x_{B,min}$ to the left). In Figure 8a (low energy price), we see that the objective is decreasing throughout the feasible region, meaning the optimum is at the right end of this region, i.e., at the point where $V = V_{max}$. The opposite is true for Figure 8c (high energy price), where the minimum lies at the *left* end of the green part of the curve. Here, the constraint on x_B is active. In Figure 8b (intermediate energy price), we see that the minimum lies within the green region, meaning neither V nor x_B are at their constraint values. Thus, Figure 8a,b,c corresponds to Regions II, I, and III in Figure 2.

More on the "'Avoid Product Giveaway'" Rule. In case studies Ia and II, we used what we called the "avoid product giveaway" rule[8] later used in ref 18. The rule can be stated as follows: *The purity constraint for the most valuable product is always active.*

However, this rule relies on at least two assumptions:

- A1: *The valuable product price is constant, meaning we get paid for the impurity as well, as long as the purity specification is satisfied* (this is the case in case studies Ia

and II but not Ib). If we do not get paid for the impurity, the value of the product (in $/mol or $/kg) increases when we overpurify.

- A2: *Overpurification costs extra energy.* For distillation, this seems to be a general rule, except possibly for some very nonideal mixtures. Define the overall separation factor for distillation as the ratio between light and heavy key component in the distillate and bottoms products, respectively, $S = (x_L/x_H)_D/(x_L/x_H)_B$. Then, S generally increases as we increase the internal flows L and V. For example, for the case with constant relative volatility, we have from the Fenske formula that the maximum separation factor $S = \alpha^N$ is achieved for infinite L and V, that is, for infinite energy.

With the statement "overpurification costs extra energy", we are here assuming that we fix the composition in the cheap product (e.g., x_B) while overpurifying the valuable product (x_D). Of course, if we let both compositions vary, then "overpurification" need not cost more energy. Consider, for example, a case where the separation is very simple so we use a simple flash. Assume that the feed is partly vapor and that the flash gives 95% purity in the valuable top product (x_D) without adding any energy. If the specification is only 90%, we would need to boil up more of the heavy component (use more
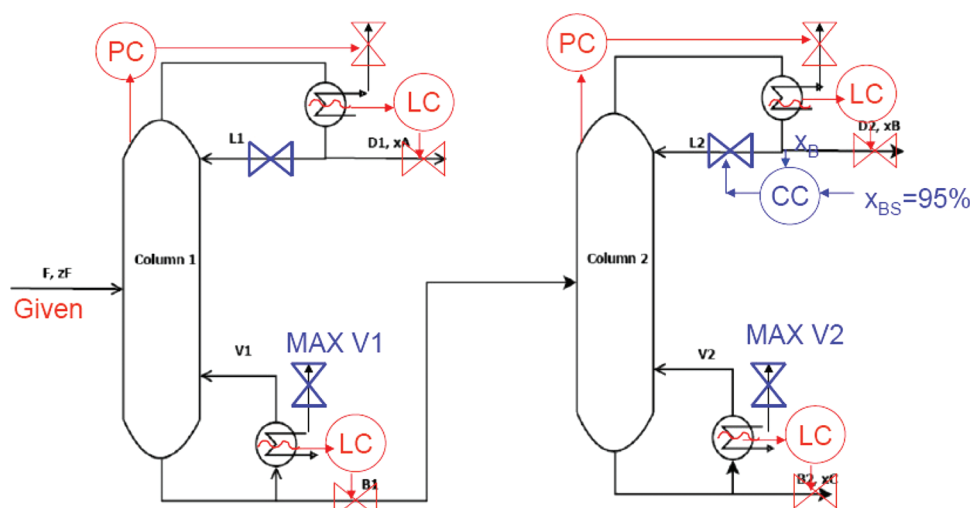
Figure 9. Control of two-column system in Region VI: Possible control structure with stabilizing layer (red) and control of active constraints (blue). The reflux flow in the first column remains as a degree of freedom.

energy) to achieve this, so overpurification (to x_D = 95%) seemingly saves energy. However, in a flash, x_B will increase when x_D increases because of the equilibrium between the products, so the bottom product becomes less pure and there is additional loss of valuable product in the bottom. In summary, even for this special case where a single flash is sufficient, it seems unlikely that it will be economically optimal to overpurify the valuable product.

Selection of Control Structure. A detailed analysis of selection of controlled variables is outside the scope of this work. However, we will discuss it briefly here. Within the self-optimizing control framework, we should:

1. Control the active constraints
2. Use the remaining unconstrained degrees of freedom to control variables whose optimal values are relatively insensitive to disturbances

For the single column case studies, there are two degrees of freedom, so we need to find two variables to control. In case Ia, with fixed prices, Regions II and III both have two active constraints, so in these regions, selection of controlled variables is straightforward (control the active constraints). In Region I, there is one active constraint (X_D) which should be controlled. The unconstrained degree of freedom can be used to control x_B; as we see from Table 2, this variable is relatively constant around 0.9900.

For the two-column case study, we have a lot of room for selecting controlled variables. There are nine regions, and thus, we may have to select as much as nine sets of controlled variables. However, one variable needs to be controlled everywhere (the purity of the most valuable product), and some are active over large parts of the disturbance space (like the purity of stream D_1, the top product of the first column).

When the active constraints are controlled, we need to select controlled variables to be associated with the unconstrained degrees of freedom. There are four degrees of freedom. From Table 7, we see that that we need to find three variables in Region I, two variables in Regions II and III and one variable in Regions IV, V, and VI. In Regions VII and VIII, all degrees of freedom are used to control active constraints.

As an example, we consider in Figure 9 the control in Region VI, where the three active constraints are x_B, V_1, and V_2. The

stabilizing control layer is based on the common LV-configuration, where reflux (L) and boilup (V) remain as degrees of freedom after having closed the stabilizing loops for levels and pressure. The control of the three active constraints is then easily implemented using the reflux in the second column (L_2) to control x_B (in the top of the second column), whereas the two vapor flows (V_1 and V_2) are simply set at their maximum.

In Figure 9, the reflux in the first column (L_1) remains as an "unused" unconstrained degree of freedom, and we would like to select an associated "self-optimizing" controlled variable. The obvious choice would be to control x_A in the top of the first column, but this is not a good choice because the optimal value of x_A changes as a function of the feedrate, from about 0.98 at low feed rates (as we enter from Region III) to 0.95 at higher feedrates (when it becomes active and we enter Region VIII); see Table 8.

A better choice may be to control the amount of impurity A which is carried through to the second column ($x_{A,B1}$) as variations in this concentration are likely to cause trouble for the second column. As seen from Table 8, the optimal value of this concentration does not vary much, which means it could be a good controlled variable in *all* regions where we have unconstrained degrees of freedom. However, to identify the best choice of controlled variables, a more detailed analysis is needed, based on evaluation of the cost for different alternatives.[9]

■ CONCLUSIONS

We have applied the method described in ref 13 to three distillation case studies. The method allowed us to find these regions using relatively few optimizations. For the cases with constant prices, we found that the purity constraint on the more valuable product was always active, as expected. For a single distillation column, we found three distinct regions (with constant prices) and five regions (with a purity-dependent distillate price). For two columns in sequence, we found eight distinct regions. In all three cases, we have found the highest feed rate for which the columns can run without violating purity constraints, i.e., the physical bottleneck. We have also described the assumptions under which the "'Avoid Product Giveaway Rule'" is correct.

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